BABEŞ-BOLYAI UNIVERSITY

FACULTY OF ENVIRONMENTAL SCIENCE

THE GEOTHERMAL PHENOMENON FROM THE WESTERN PART OF ROMANIA. THE ENVIRONMENTAL IMPACT CAUSED BY THE EXPLOITATION OF THE GEOTHERMAL WATERS FROM ORADEA AND SĂCUIENI PERIMETERS

PhD Student Thesis

(Summary)

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CONTENT

Key words: geothermal phenomenon, geothermal recourses in Romania, waste geothermal withdraw, inorganic contaminants, organic contaminants, radionuclides, risk, environmental impact assessment

INTRODUCTION

Throughout its existence, man was in a permanent relationship with the environment, adapted them, and exerted a major influence on them too, especially on climate, having both positive and negative consequences. In the last century, the use of the energy produces by fossil fuels (oil, gas, coal) combustion has had severe environmental impacts, higher than any human activity in history.

Furthermore, the economic development in the new century seems to be limited due to geological resources decline. There is a direct link between the energy use per person and the life standard. To improve the people life standard is necessary to find clean energy resources at affordable prices for the majority of the people.

Currently, in the energy sector of most countries, there is a priorities reconsideration 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 guaranteed. As such, the use of alternative energy sources becomes increasingly important/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.

For the future, the World Energy Council estimates a 30-80% increase in the use of unconventional energy resources, particularly geothermal resources. Geothermal energy is an inexhaustible source of energy, whose use gives several advantages such as minimal impact on the environment, requires limited space for development, is available 24 hours from 24, can be used as a viable alternative to fossil fuels.

Geothermal energy can, at least locally; to contribute significantly to the reducing of fossil fuel consumption, being economically competitive, and reducing the fossil fuels imports the pollutants emissions due to their combustion.

The present paper is a comprehensive study on the geothermal phenomena from western Romania, and on the assessment of environmental impact due to the exploitation of geothermal water originating from two perimeters (Oradea and Săcuieni) located in Bihor County. The paper is structured in five chapters as follows:

- The **first chapter** presents general information on the geothermal exploitation history, and the areas with geothermal potential from the world and from Romania.

- The **second chapter** presents in details the geological, hydrogeological and geothermal features of the investigated perimeters (Oradea and Săcuieni).
- In **chapter three** are shown the experimental results of physico-chemical analyses of water samples originating from the two perimeters. There were analyzed some physico-chemical parameters (pH, temperature, electrical conductivity, redox potential, indicating manganese), major dissolved ion content, content of organic matter (humic acids, phenolic compounds and petroleum hydrocarbons) and radionuclides activity (^{222}Rn and ^{226}Ra). For each analyzed parameter, the collecting and preserving water samples procedures and the used methods are described. At the end there are the results and discussions.
- **Chapter four** is devoted to environmental impact assessment and human health arising from the exploitation of geothermal water originating from Oradea and Săcuieni perimeters. The impact assessment was made based on the physical and chemical characteristics of geothermal waters, comparing the analysis results and the maximum allowable concentrations normal law.
- In **chapter five** are given the final conclusions of the study.

CHAPTER I.
INTRODUCTION

1.1. Short history on the geothermal exploitation

An intense use of geothermal resources in ancient times was mentioned during the maximum flowering period of the Roman Empire. The interest for this domain registered a regression during the Middle Ages, then at the end of the XIX century started an intensive exploitation of the geothermal sources. This phenomenon increased in the early of XX centuries, marked by the development of the technologies used in this domain.

1.2. Areas in the world with geothermal potential

The majority of geothermal fields are located near the lithospheric plates contact areas. Europe has a high geothermal potential associated with the presence of hot rocks areas. Geothermal flux distribution map (Fig.1) indicates the existence of areas where this parameter exceeds 150 mW/m^2 (Iceland, France, Italy, Greece and Turkey) [Hurter and Haenel 2002].

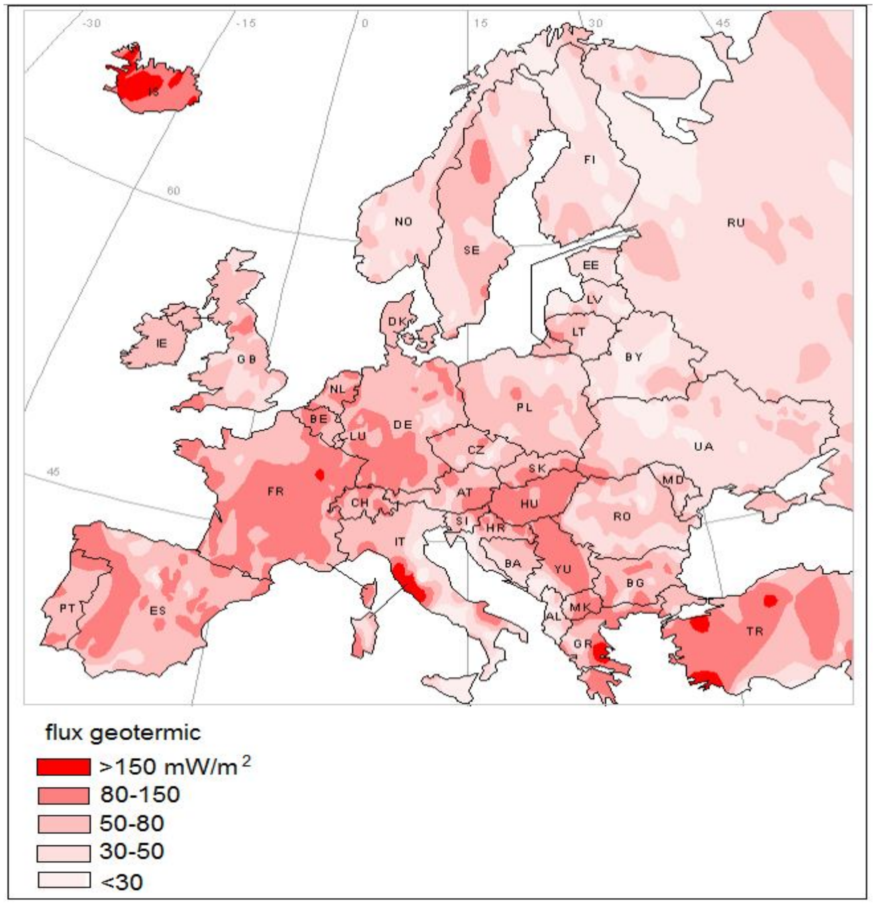


Fig.1. Geothermal flux distribution in Europe [Hurter and Haenel 2002].

1.3. Geothermal potential areas from Romania

The exploration of geothermal resources in our country began in the 60s, when an extensive research project of hydrocarbon reserves began. In that time were found eight geothermal areas: six of them are located in the western parte of Romania, and the other two in the south of the country [Airinei 1981]. In these areas were drilled over 200 wells at depths of 800-3500 m, which proved the existence of geothermal resources of low (25-60 ° C) and average enthalpy (60-120 ° C) [Airinei 1981]. The antecedents are much older, the first geothermal well was drilled in Romania in 1885, in Felix Spa, near Oradea. The well had a depth of 51 m, a flow of 195 l/s, and a weelhead temperature of 49^0C. Afterwards, it were drilled the wells from Căciulata (1893 - 370C), Oradea (1897 - 290C), Timisoara (1902 - 310C), etc.

The geothermal fields of Romania are located mainly in the west of the country (Fig.2) [Negoitza 1970 Bandrabur et al. 1982, Fall 1985, Burchiu et al. 1998 Cohut and Benda 2000].

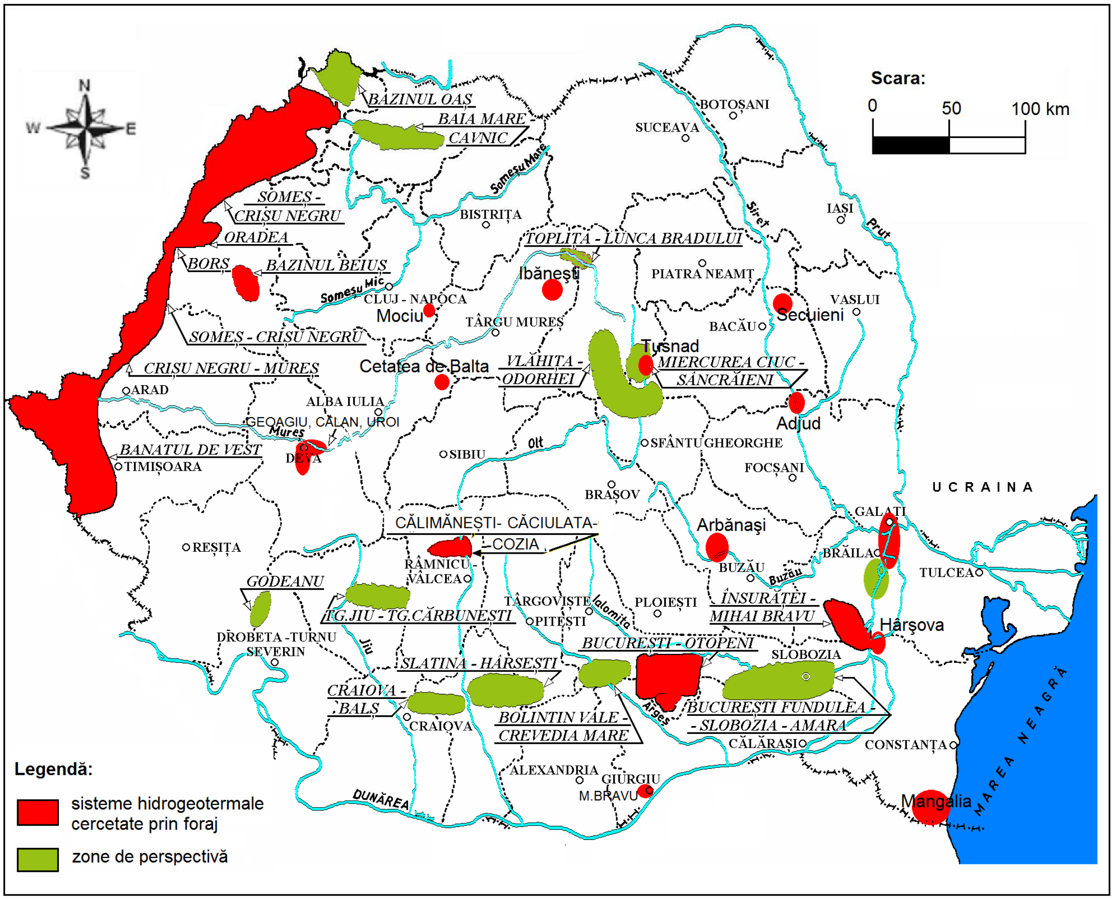


Fig.2. Geothermal areas explored by wells and perspective areas, amended Negoiţă 1970 Bandrabur et al. 1982, Fall 1985, Cohut and Benda 2000.

The use of the geothermal resources identified in our country during 1995 and 2005 is shown in Fig. 3. [Panu et al. 1996 Cohut and Benda 2000, Rosca et al.2005].

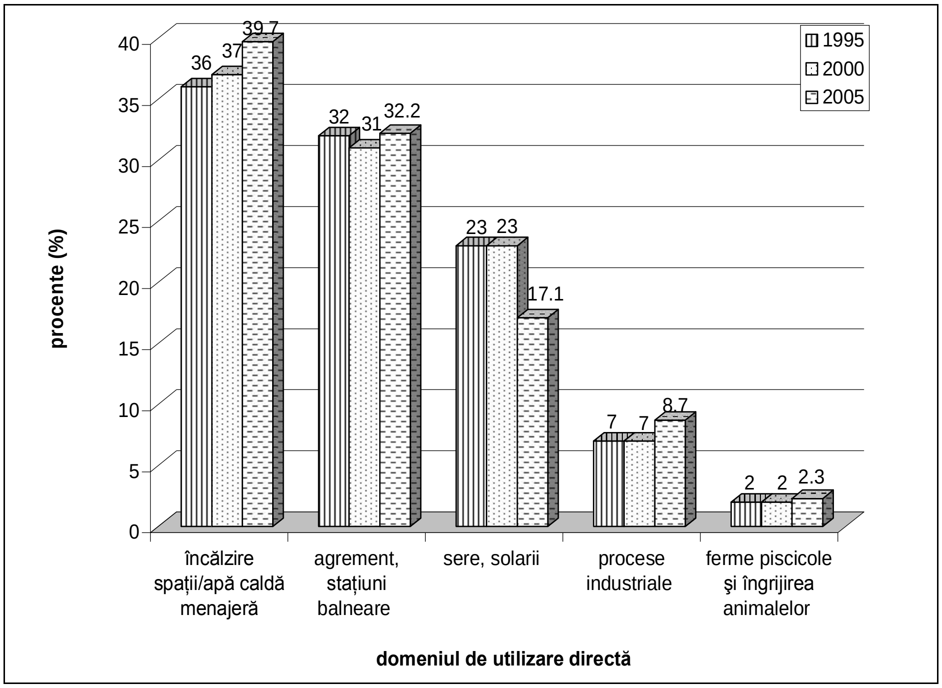


Fig.3. The direct use of geothermal resources in Romania, during 1995-2005 [after Panu et al. 1996 Cohut and Benda 2000, Rosca et al. 2005].

CHAPTER II

CHARACTERIZATION geological, hydrogeological and geothermal AREA OF STUDY

In this paper we have investigated two geothermal aquifers located in Bihor County, whith contrastant hiderogeological and hydrochemical features, which are the Triassic geothermal aquifer of Oradea and the Lower Pontian geothermal aquifer from Săcuieni. The two thermal aquifers are located in Pannonian Basin, the richest geothermal resources region in the country.

The presence of geothermal deposits on our country territory is the result of the presence of high heat flow areas, as shown in Fig.4. [Negoitza 1970, Fall 1985, Milcoveanu 1984 Veliciu 1987, 1998]. The heat excess, manifested by an increased heat flux, is derived from the subcrustale magmatic processes and regional characteristics of lithosphere structure. The presence of this heat flux is mainly due to the thinning of terrestrial crust from the Intra-Carpathian Basin, as a consequence in the Pannonian Basin the Mohorovicic discontinuity is located at a depth of 20-25 km, compared with 30-35 km

- average depth at which this discontinuity is located in Europe [Paal 1975, Paraschiv 1975].

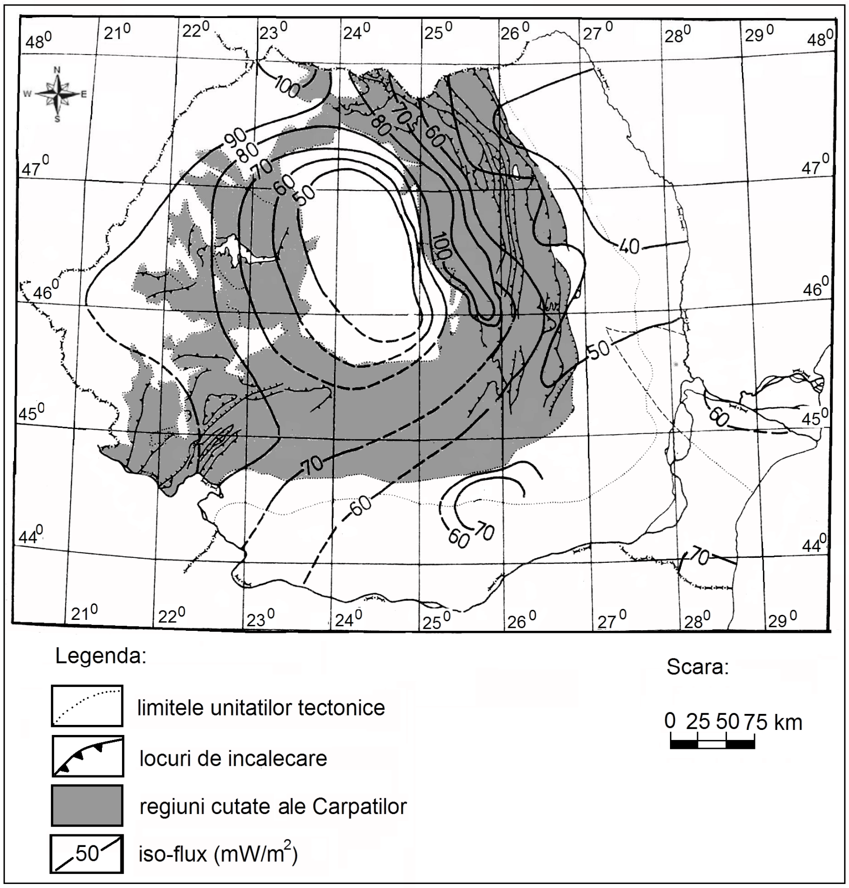


Fig. 4. The distribution of geothermal flux in Romania, modified after Milcoveanu 1984 and Veliciu 1987.

2.1. The Triassic geothermal aquifer from Oradea

Geothermal deposit is located almost entirely in the basement of Oradea, covering an area of approximately 75 km^2 [Paal 1975 Țenu 1981].

2.1.1. Geological framework of the Oradea perimeter

The data from the drills located in the area, correlated with the results of the geological research carried out in adjacent areas [Vasilescu and Nechita 1968 Istocescu[ab], 1970, Bleahu et al. 1971, Michael 1971, Michael 1972, Țenu 1981 Cohut 1986 Veliciu 1987, Miklos 2000] showed that the geological structure in the area is consists of formations belonging to the Quaternary, Neogene, Cretaceous, Jurassic and Triassic, the basement consists of metamorphic rocks.

The metamorphic basement consists of a complex of michasisth with granite, biotite, tourmaline and a paragneiss complex with muscovite and biotite belonging to the Someş Series [Vasilescu and Nechita 1968 Mutihac 1975 Țenu 1981, Săndulescu, 1984].

The metamorphic basement is covered by the sedimentary formations belonging to Bihor Unit of Triassic, Jurassic and Cretaceous age, plus parts of the sedimentary filling of the Pannonian Basin of Miocene and ?Pliocene age [Vasilescu and Nechita 1968 Țenu 1981] (see Fig. 5).

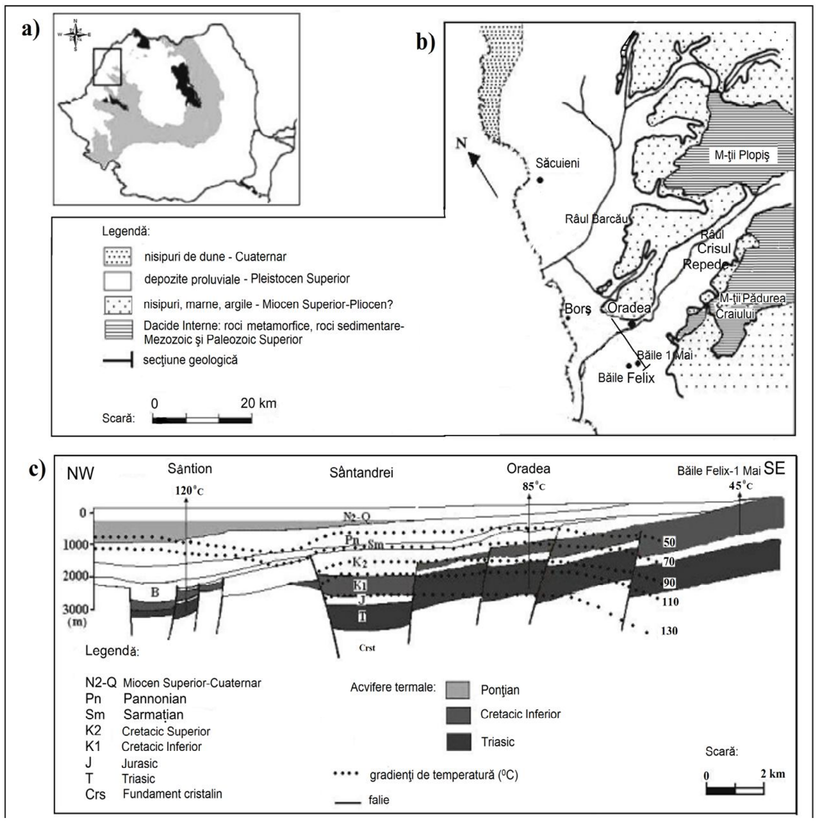


Fig.5. Geology study area: a) location in Romania, b) geological aspects of the area, with amendments after 1981 Țenu c) geological section of the Triassic aquifer of Oradea, modified after Cohuț 1986.

The Triassic deposits lay over the metamorphic basement and consist on significant vertical lithological variations, which made possible the delineation of some subdivisions [Vasilescu and Nechita 1968 Țenu 1981].

The Lower Triassic consists on detritic Werfenian formations, represented by siliceous sandstone, very compact, light purple or gray, alternating with sandstones and clay schists, encountered in wells 4004, 4006, 4796, 1715, 1717 [Vasilescu and Nechita 1968]. The Middle Triassic sequences are represented by a set of dolomites (with thickness of approx. 49 m) very compact, gray, blackish belonging to Anisianului. Ladinian consists on Bucea black limestone, of Guttenstein type, with many fine crystals of calcite and pyrite, disseminated in limestone mass [Răileanu1957, Preda 1962, Doe and Nechita 1968]. Lower and middle Triassic sedimentation in the area is completed by white limestone of Weterstein (Ladinian) type, intercepted by drilling 4006, 4004, and in some areas (probe 4767) they were replaced by gray limestone dolomite [Țenu 1981]. Over the Guttenstein black limestones lays a powerful package of gray dolomites, with thicknesses of 394 m (4006 drilling), or 120 m (4005 drilling). The Carnian consists on a series of compact blackish sandstone limestone, clay schists, dark and compact limestone, with a thickness of 220 m (4006 drilling), 206 m (4005 drilling), and a limestone compact package, gray, with calcite diaclase, of approx. 214 m thickness, placed over limestone and dolomite [Răileanu1957, Preda 1962, Doe and Nechita 1968]. The Triassic sedimentation of the area closes with formations belonging to Norian, represented by whitish gray limestone with pink spots, in some core drills were identified large areas of limestone of brick color. The total thickness of Triassic deposits range from 500 to 1300 m [Țenu 1981].

The Jurassic deposits consist of red shales, quartzite sandstones, limestones, shale, clay shale [Mészáros et al. 1999, Popa et al. 2003]. The total thickness of the Jurassic deposits is of 50 ÷ 350 m, lower than the Triassic one.

The last Mesozoic sedimentation cycle from the Oradea perimeter belongs to the Cretaceous. Lower Cretaceous (Barremian-Aptian) - is represented by limestone gray - brownish yellow with diaclase calcite, limestone and marly-blackish gray [Vasilescu and Nechita 1968 Țenu 1981]. The Upper Cretaceous (Senonian)- is represented by marls, fine sandstone, limestone, sandstone, conglomerates and clay schists, having 300 to 1000 m thicknesses [Țenu 1981, Sandulescu 1984 Cohut 1986].

The Neogene is represented by Miocene and ?Pliocene deposits [Petrescu 1979]. Lower Miocene deposits include Badenian and Sarmatian deposits of 50 ÷ 250 m thickness and contain marl, marl sandstone, gray sandstone with thin intercalations of limestone and gray sandstone, calcareous sandstone [Țenu 1981]. Upper Miocene deposits contain Pannonian ss sequences, consisting of fine sandy marl-sandstone, gray-green sandy clay, with thickness of 415 m (drill 4006) or 709 m (drill 4005). In the Oradea perimeter it can be present some Pliocene deposits, but their age is uncertain [Voitești 1936, Petrescu 1979].

The last deposits from the perimeters belong to Quaternary and consist of sands and gravels from Crişul Repede River alluviums [Vasilescu1968, Țenu 1981 Codrea 1996].

2.1.2. Structural elements and regional tectonics

As a result of late Cretaceous/Paleogene tectogenesis and Neogene subsidence [Săndulescu 1984] which took place in the area, a system of faults and thrusts was developed. The main fault which outlines Oradea Triassic aquifer are: Velenta fault system, fault Santandrei, Fault Nojorâd [Paal 1975 Cohut 1986].

2.1.3. The hydrogeology of the area. Observations on regional-scale hydrodynamics Triassic aquifer thermal Oradea

The main aquifer systems in the area are located in Holocene, Pleistocene, Pliocene, Pontian, Lower Cretaceous and Triassic deposits. The first systems contain cold water while the last three contain thermal water [Țenu 1981].

The Triassic collector contains limestones and dolomites at depths of 2,200 to 3,400 m and belongs to the Inner Dacides [Săndulescu 1984], and to be more precise to the Bihor Unit, as an underground extension of the rocks exposed in the Pădurea Craiului Mountains and covered by the Cenozoic formations of the Pannonian Basin. The aquifer probably has an active recharge; and its waters likely originate from the Apuseni Mountains, located approximately 80 km east of Oradea. The main flow of these waters is orientated from the northeast towards the west [Țenu, 1981]. A secondary flow direction, likely connected to the main flow trends north-northwest towards the south-southeast [Țenu 1981].

2.1.4.The geothermal features of Oradea perimeter

In the area of Oradea, at 2400 m depth thermal gradient has an average value of 25-42.5°C/km (Fig. 6).

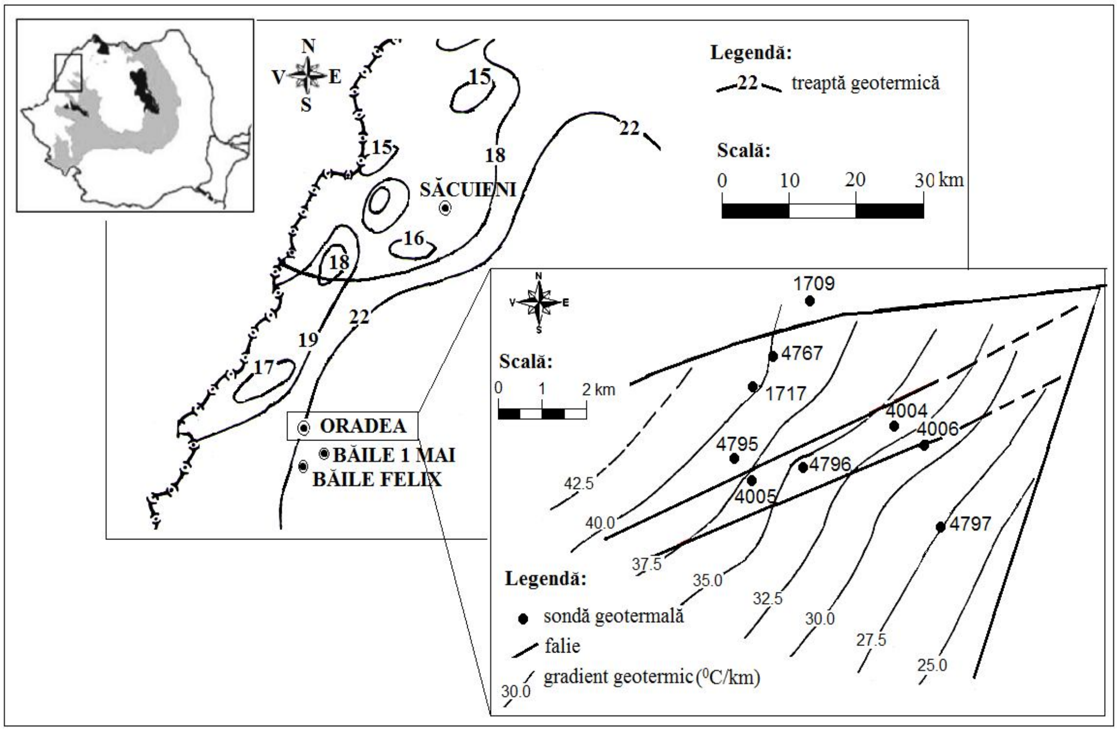


Fig.6. Geothermal features of the Triassic geothermal aquifer of Oradea, with changes after 1973 and Butac Paraschiv 1985.

The 12 existing wells within the perimeter of Oradea have an artesian flow ranging between 2.5 and 30 l/s [Țenu 1981]. The wellhead temperature range between 72 and 105^{0}C, decreasing from West to East.

2.2. The Pontian geothermal aquifer from Săcuieni

Săcuieni City is located at a distance of 45 km northwest of Oradea. Within the Săcuieni perimeter, geothermal waters originating from lower Pontian aquifer are exploited. Thermal aquifer covers an area of 2500 km^2 and was investigated by the 13 wells drilled in the region (including oil drills). The deposit is now exploited by two wells (4691 and 1704).

2.2.1. The geological framework of Săcuieni perimeter

The basement was intercepted by numerous deep wells and is generally composed of metamorphosed rocks and quartzite schists, ortho-gneisses, micaschists, crystalline limestone, and eruptive rocks (Fig. 7) [Beca 1983 Filipescu 1979].

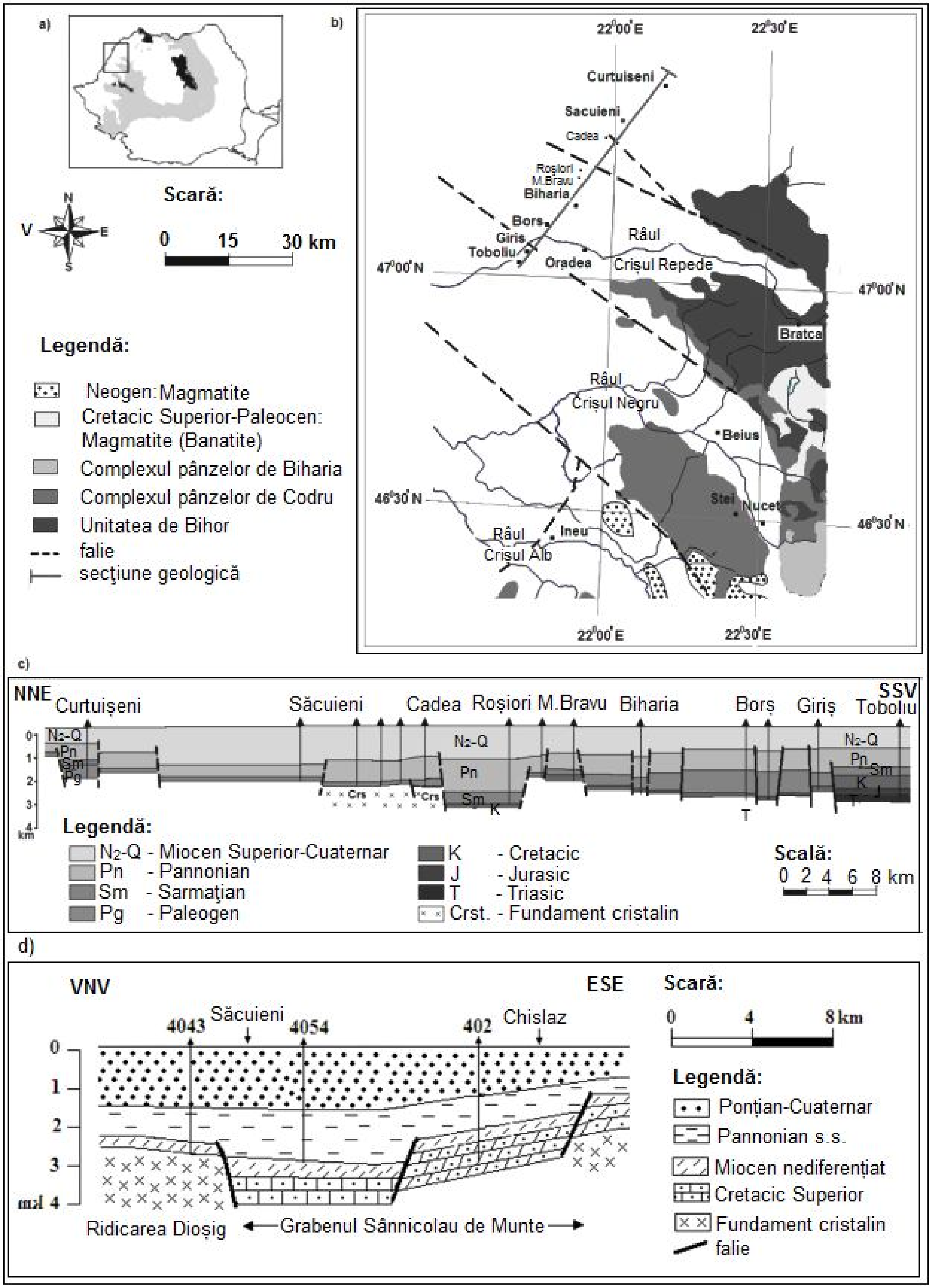


Fig.7. Geology of the study area: a) location in Romania, b) geological aspects of the area, with amendments after 1981 Țenu c) Geological section of thermal aquifer from Săcuieni, with changes after Butac 1985, d) detailed geological section from Săcuieni, modified after Țenu 1981 .

Over the basement lay the Upper Miocene deposits belonging to Upper Badenian, Sarmatian ss, Pannonian ss and Pontian [Giurgea 1972 Bessarion 1979 Țenu 1981 Butac 1985]. The Badenian sequences consist of conglomerates and sandstone marls [Filipescu 1979 Țenu 1981]. The Sarmatian ss deposits consists of conglomerates, marls and sandy marls [Giurgea 1972 Filipescu 1979 Țenu 1981]. Pannonianul s.s. deposits from Săcuieni are characterized by a lithological monotony, and consists of marl and clays, sands are intercalated shale, sometimes even layers of lignite [1969 Mihaila, Giurgea 1972, Beca 1983]. Unlike Pannonianul ss, characterized by a clear pelitic dominance, the Pontian is characterised by a general and sudden change of lithology, resulting in a roughly cyclic sedimentation with cycles that have coarse sand or gravel base, followed by sand increasingly fine, then clay and marl [Giurgea 1972 Țenu 1984]. In the area the sedimentation process continued with the submission of Pliocene sequences belonging to Dacian and Romanian. In the north sector of Pannonian Basin were identified [Filipescu 1979], a lower complex which consists of marl, sand and gravel, and aupper complex made by sands, sandstones and intercalations of marl. Because of tle lack coredrillings and micropaleontological there it can not be set a clear delimitation of Pon'ian/Dacian and Dacian/Romania limits.

The Quaternary deposits are of great extension, covering the entire area, see Fig.7. The thickness of these deposits decreased from NE to SW, becoming thinner [Giurgea 1972]. The Pliocene/Quaternary limit is difficult to delimitate because of the lithological similarities of bouth deposits [Țenu 1981]. The Quaternary deposits consist of Pliocene (yellowish-reddish clayly sands) and Holocene (sand dunes and marsh) deposits [Mihăilă 1969, Giurgea 1972].

2.2.2. Structural elements and regional tectonics

In the area a system of faults and thrusts was developed. These systems of fractures and faults led to the formation and preservation of a thick stack of sediments.

2.2.3.Hidrogeological features of the area. The hydrodynamic features of the lower Pontian aquifer from Săcuieni.

The hydrogeological resarches from the area, evidenced the presence of several aquifers: the freatic, located in Holocene and Upper Pleistocene deposits, and deep aquifers located in Lower Pleistocene, Lower Pontian, Pannonian ss and Sarmetian deposits [Giurgea 1972].

The Loer Pontian thermal aquifer from Săcuieni is located at depths between 1250 and 1700 m. The aquifer is multylayerd and consists of sands with marls and clay interbeddings. The individual thickness of permeable layers is negligible in relation to their extension, forming a multilayerd collector type. This complex involves 1 to 100 permeable layers (in areas with high basement) and 40-50 layers (in areas with an active subsidence during Pontian). Cumulative thickness of this layer amounts to 1.5 to 150 m, common values are those between 2 and 25 m [Țenu 1981]. The first 4-8 levels of basal horizons have a general distribution, while the upper layers occupy limited areas, especially in central areas. The number of the layers intercepted by the wells range between 30 and 40. The total thickness of the complex, including impermeable intercalations, is reaching 500-800 m [Țenu 1981]. The lower level of the complex correspond Pannonian ss/Pontian limit and the upper level of the complex correspond to Medium Pliocene, where marls with rare intercalations of thin sandy or clay layers predominate [Țenu 1981]. The wells have an flow of 17-20 l s in winter and 3-4 l/s in summer, and the wellhead temperature range between 80 to 84^{0}C [Țenu 1981]. The use of geothermal energy can assure an energetic equivalent of 2600 toe (tons oil equivalent), of which 1300 toe (50%) are currently being used.

The hydrogeological studies [Țenu 1981, 1983, Plavi 1992] proved the low recharge of geothermal aquifer. It was assumed that there are some natural ways for aquifer recharge in Marghita-Derna area, and two major drainage axes arranged from NNW to SSE (Oradea-Carei) and NNW –SSV (Satu-Mare area). The presence of two axes of drainage is the result of structural-tectonic changes that are felt by similar effects to the Quaternary. The recharge volumes are very low and the thermal aquifer has practically a closed circuit.

2.2.4.*The geothermal features of Săcuieni perimeter*

The wellhead temperature of the waters originating from Lower Pontian aquifer from Săcuieni range between 70 and 92 °C, in the weel the values are higher by 10-15 °C. these high tempearurea are colrrelated to the pronounced geothermal anomaly from Pannonian Basin, which reaches maximum values along the Tisa Valley where the heat flux density has an average of 2.7 μcal/cm^2·s, compared to 1.5 μcal/cm^2·s which is the global average value [Paraschiv 1975 Airinei 1981, 1987]. The geothermaşl gradients in the area have an average value of 52^0C/km, the temperature isotherms range between 90-105^0C (see Fig.8), and the geothermic step is approx. 1^0C/18 m compared to the average value recorded in Europe 1^0C/33 m [Airinei 1987].

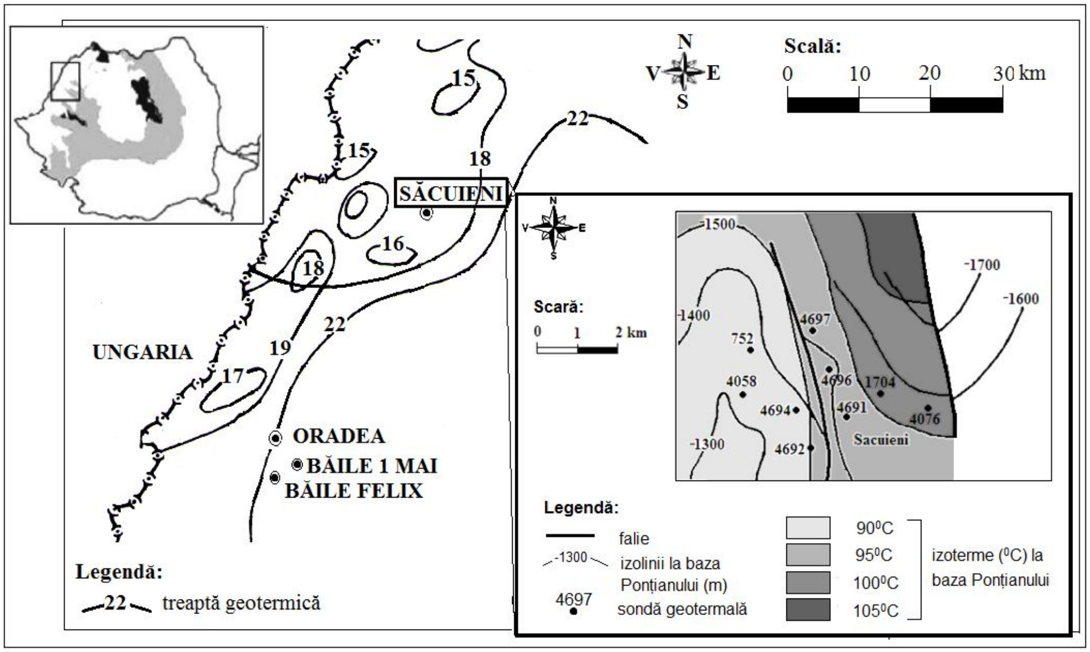


Fig.8. The geothermal features of the Lower Pontian aquifer from Săcuieni, with changes after Paraschiv 1973 and Plav 1992.

CHAPTER III.

The physico-chemical features of the geothermal waters originating from Oradea and Săcuieni perimeters

In the present study was carried out physico-chemical characterization of geothermal water originating from the Triassic aquifer from Oradea and the Lower Pontian aquifer from Săcuieni, during 2007-2010. It were investigated some physico-chemical parameters (pH, temperature, electrical conductivity, redox potential, indicating manganese), major dissolved ion content, organic matter (humic acids, phenolic compounds and petroleum hydrocarbons) content, and and the radionuclides (^{222}Rn and ^{226}Ra) activity.

To assess the possible effects induced by the metheorological cganges and the exploiations regime of geothermal wells, the sampling campaigns were conducted during the cold season (December-February months), when the geothermal plants operated at high capacity and the hot season (July-August).

Results have highlighted significant differences in the hydrochemical features of the two aquifers. The water is slightly acidic to alkaline pH in the range of 6.3-8.15. The pH level is due to contact with basic rocks such as limestone and dolomite. The electrical conductivity of geothermal waters in the aquifer is relatively low in Oradea (1107-2100 mS/cm) than in Săcuieni (6201-7540 mS/cm), the value of this parameter is directly correlated to the concentration of dissolved ions. Water hardness of Oradea aquifer range between 28.95-52.04 gr.ger compared to Săcuieni where this parameter range between 1.45-2.50 gr.ger. Hardness level reflects the geological characteristics of the two aquifers. The high water hardness of Oradea is a consequence of the presence of limestone and dolomite [He et al. 1999 Scrădeanu and Alexander, 2007] in the Triassic aquifer, rocks which have a high content of calcium and magnesium salts, compared with Săcuieni aquifer where sands dominate. The water from the two aquifers are moderately reducing, with redox potential up to -50.5 mV (Triassic aquifer) and -74.35 mV (Pontian aquifer). These low values of redox potential may indicate the presence of an anaerobic environment and reducing conditions in the two aquifers.

The dissolved anions content is dominated by the presence of sulfate ions (339.91-730.8 mg/l) and bicarbonate (165.03-275.36 mg/l) for Oradea, respectively bicarbonate ions (2195.37-2403.65mg/l) and chlorine (542.87-625.21 mg/l) for Săcuieni aquifer (Fig. 9). Major dissolved cations are calcium (134.28-274.95 mg/l) and magnesium (32.71-71.22 mg/l) for Oradea aquifer, and sodium (1239.24-1396.21 mg/l) and potassium (39.92-58.15 mg / l) for Săcuieni aquifer.

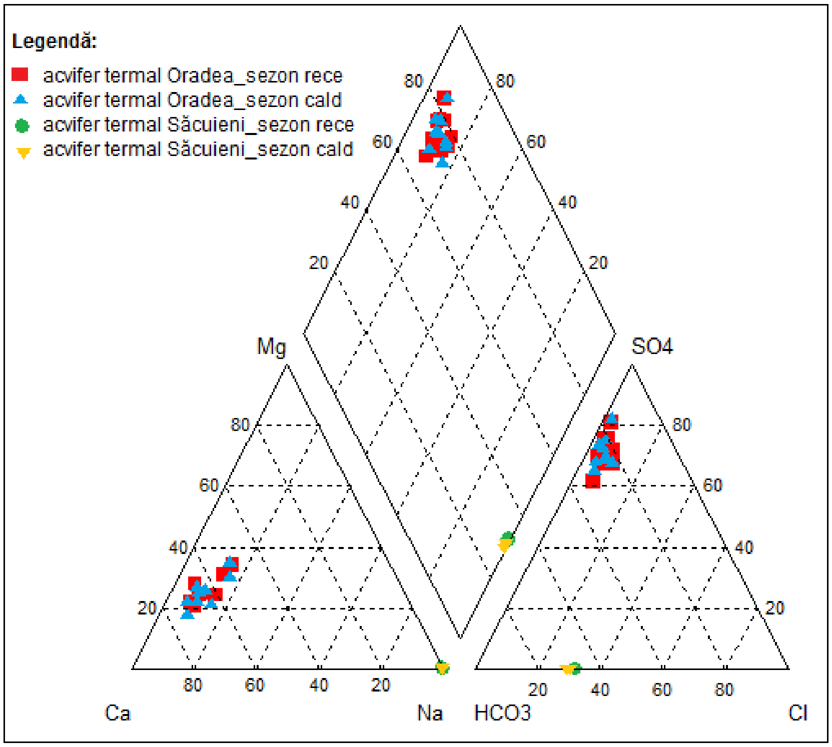


Fig.9. Piper diagrams showing the abundance of dissolved ions in the two geothermal aquifers.

As shown by in Fig.9 the waters from Triassic aquifer of Oradea can be classified as sulfate-bicarbonate-calcium-magnesium type, and those from Lower Pontian aquifer of Săcuieni are bicarbonate-sodium-chloride type. The high content of Ca^{2+}, Mg^{2+}, HCO_3^- and SO_4^{2-} for Oradea is the consequence of the presence of limestone ($CaCO_3$), dolomite ($CaMg(CO_3)_2$) and anhydrite ($CaSO_4$) in the aquifer [Gilău 1997 Stănăşel et al. 2003, Baciu 2004, Muraoka 2010]. High content of bicarbonate in Pontian aquifer can be correlated with reduced dynamics of these waters, which something may lead to high levels of bicarbonate and sulfate reduction [Ţenu, 1981, Qin 2005]. The high sodium concentration from Săcuieni can be correlated to the presence of clay layers în the area [Plavi and Cohut 1992], or may result from mixing with connate waters from deep reservoirs [Ţenu, 1981]. Low Ca^{2+} and Mg^{2+} content of Săcuieni aquifer can be the result of monovalent/bivalent ion exchange between Ca^{2+} and Mg^{2+} ions and Na^+ ions from caly [Ţenu 1981 Gilău 2001].

By comparing the concentrations of major dissolved ions from the two thermal aquifers, can shape their areal distribution (Fig. 10). Thus, for the thermal aquifer from Oradea the concentrations of Ca^{2+} and $SO4^{2-}$ ions have an upward trend NW-SE direction, compared with HCO^{3-} ions showing a downward trend in the same direction. This may confirm the hypothesis of the existence of drainage from W to E for the Triassic aquifer [Țenu 1981].

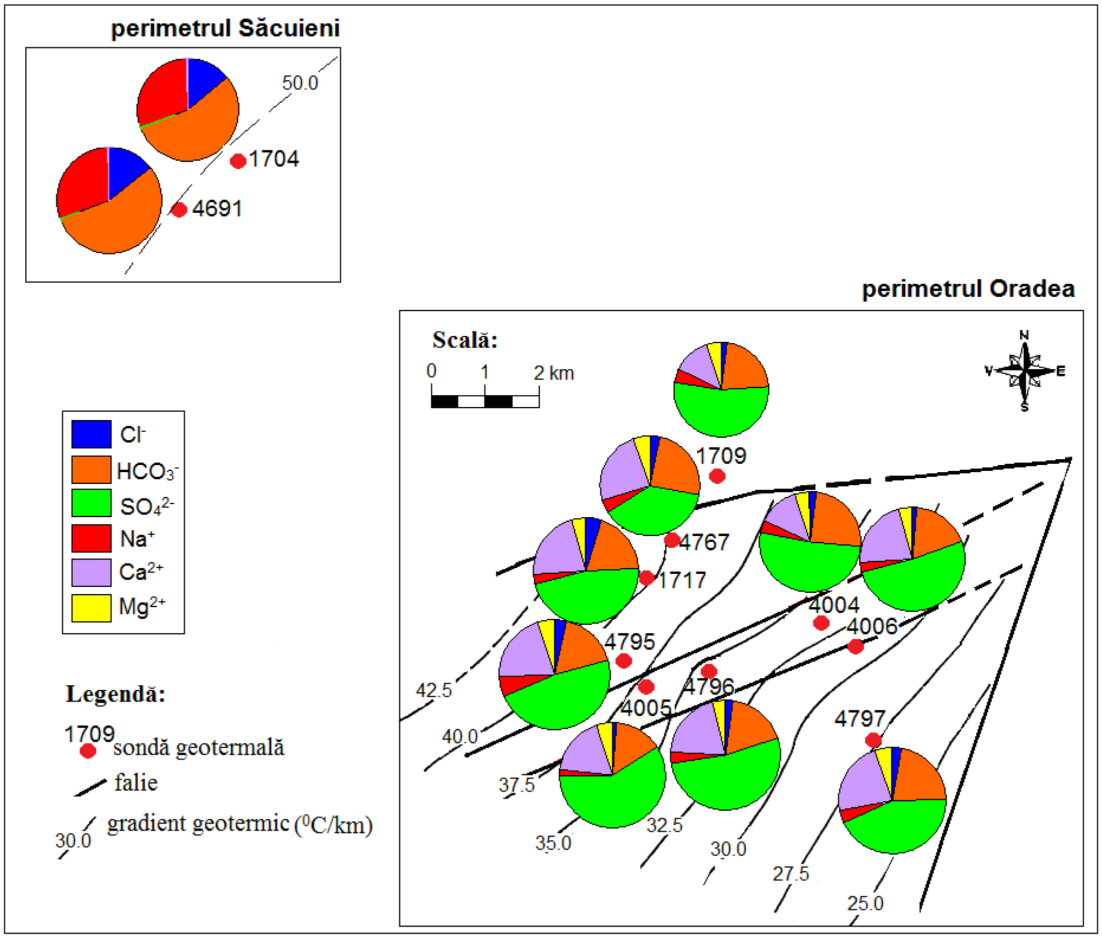


Fig. 10. Areal distribution of major dissolved ions identified in the investigated thermal aquifers.

Chemical data revealed a great similarity in terms of chemical composition of water from the same aquifer, indicating a common origin and same aquifer recharge [Țenu 1981]. Tests conducted on water samples collected in different sampling campaigns, showed no significant seasonal differences, demonstrating that climatic changes have a minor impact on thermal aquifers located at such great depths. For some samples can be identified some minor seasonal fluctuations. A slight increase was registered for concentration of Na^{+}, K^{+}, Mg^{2+}, HCO_3^{-}, SO_4^{2-}, PO_4^{3-} ions during the warm season, while the concentrations of ions Ca^{2+}, Cl^{-} have slightly decreased over the same period.

3.2. Determination of humic acids

Currently there are no national specific regulations on maximum concentration limit and/or analzse methods for the determination of humic substances/humic acids in water samples, or samples with complex matrices. This paper aims to contribute to this field, by following to develop and implementate a simple method to analyse humic acids (HA) in water samples. The analysis method consists of HA separation by solid phase extraction technique (SPE), followed by HA quantification by UV-Vis spectrophotometry. There was lso performed a qualitative analysis of HA by high performance liquid chromatography (HPLC), aiming to separate the hydrophilic and hydrophobic component of humic acids. The method was then applied to natural water samples originating from different aquifers located in Oradea and Săcuieni perimeters.

The Ha content in geothermal waters from Oradea range between 0.55 and 1.38 mg/l, having significantly lower values than those registered in Săcuieni where HA concentration range between 2.13 and 2.61 mg/l (Fig.11). The same trend is maintained, and in the case of drinking waters: 0.58-0.73 mg/l in Oradea, compared to 0.91-1.49 mg/l in Săcuieni. The highest HA levels were registered in the surface waters from Săcuieni (2.39- 4.02 mg/l). The high HA level from Săcuieni aquifers is a consequence of geological and hydrogeological characteristics of this area. The presence of coal layers and petroleum deposits in the perimeter Săcuieni [Țenu 1981] represents a rich source of natural organic matter which leads to high HA concentrations.

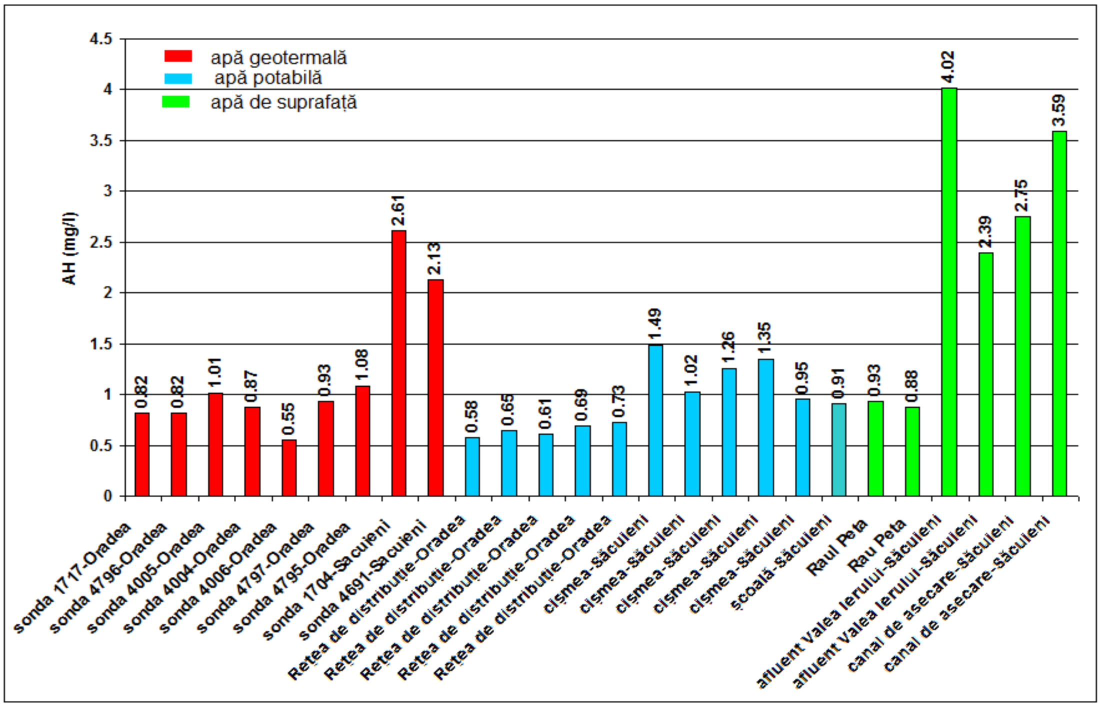


Fig.11. The HA level in different aquifers located in Oradea and Săcuieni aquifers.

In all water samples were identified the hydrophilic and hydrophobic fractions, the last one was the dominant one. For drinking water and surface water samples two hydrophobic fractions (HOB_1 and HOB_2) were identified. The share of hydrophilic fraction was significantly higher for thermal waters from Săcuieni.

3.3. Determination of phenol and phenolic compounds

The present study aimed to improve a method of isolation/concentration (by solid phase extraction and liquid-liquid extraction) and chromatographic analysis (RP-HPLC) of phenol and a set of 13 phenolic compounds in water samples. Much of the phenolic compounds analyzed are included on the list of the 11 phenolic compounds considered priority pollutants for the environment, issued by the US-EPA [U.S. EPA, 2002].

The data showed the presence of phenolic compounds in water samples originating from Săcuieni perimeter. Phenolic compounds were identified in geothermal water samples and emissaries which collect the waste geothermal waters. The presence of phenols can be correlated to the high content of petroleum hydrocarbons from Săcuieni aquifer, becaue phenols are compounds which can be found in petroleum residues.

3.4. Determination of n-hexane extracteble petroleum hydrocarbon, by fluorescence spectrometry

The waters originating from Oradea aquifers had lower petroleum hydrocarbons than the waters from the Săcuieni aquifers. The lowest content of petroleum hydrocarbons occurred in drinking water, and it was below the detection limits (<1 mg/l) for Oradea aquifers, while for Săcuini the level ranged between 1.6 and 3.4 mg/l. The waters originating from thermal aquifer from Oradea had petroleum hydrocarbon content below the detection limit, except for well 4797, which had a petroleum hydrocarbon concentration of 1.5 mg/L, while in Săcuieni thermal aquifer, the petroleum hydrocarbons level was 30-40 times higher (32.27 to 41.18 mg/l). The highest level of petroleum hydrocarbons (35.79-74.62 mg / l) was detected in surface waters of Săcuieni, while in Peța River (Oradea) the level was considerably lower (4.4 to 4.9 mg/l). The high concentration of petroleum hydrocarbons in Săcuieni perimeter is a feature that is found in other surrounding localities within the country and may be correlated with the presence of petroleum deposits identified in the area [Filipescu 1979].

3.5. Determination of radionuclides (^{222}Rn and ^{226}Rn) activities

In the present study two of radionuclides (^{222}Rn and ^{226}Rn) formed by alpha decay of ^{230}Th in the ^{238}U decay series were analyzed. The level of these two radionuclides is shown in Fig.12.

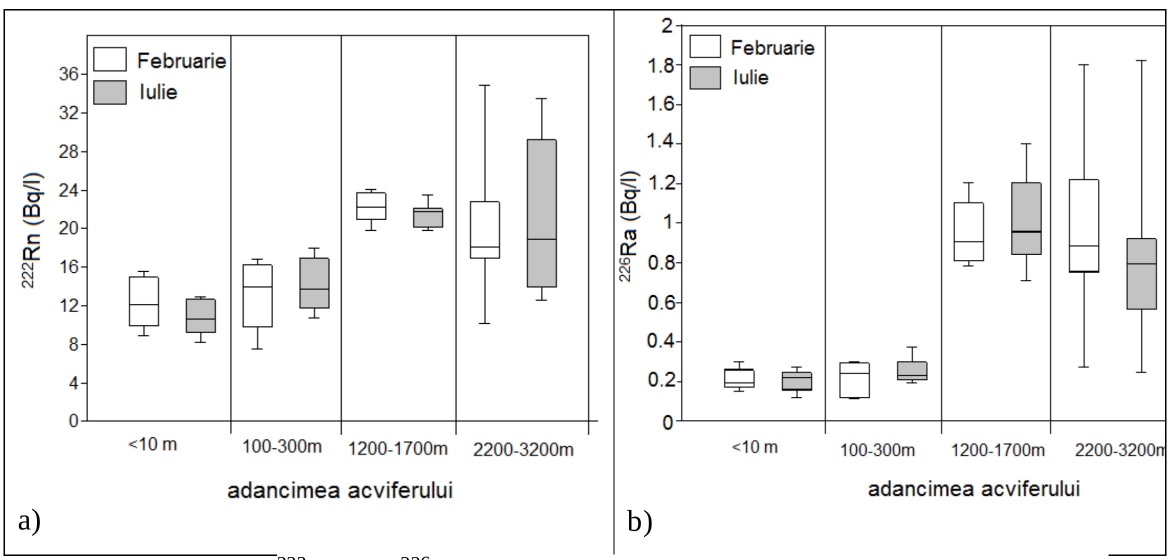


Fig.12. ^{222}Rn and ^{226}Ra activities for different aquifers located in Oradea and Săcuieni perimeters, during the cold and hot season.

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.

CHAPTER IV.

Environment impact assessment due to the exploitation of geothermal waters originating from Oradea and Săcuieni perimeters

In the present study was assessed the possible impact on the environment and human health due to geothermal exploitation of thermal waters from Oradea and Săcuieni perimeters. Oradea geothermal area overlaps the Crişul Repede Rriver hydrographic basin. The only affluent with a permanent water course is Peța River, which collects the waste geothermal waters exploited in the area, and lead these waters towards the Crişul Repede River. Săcuieni geothermal area overlaps the hydrographic basin of Ier River. The geothermal wastewater discharge into surface emissaries promotes migration of pollutants through these rivers.

The environmental impact caused by the exploitation of geothermal waters from Oradea and Săcuieni was estimated based on the results of analysys of water samples prelevated at the discharge place and upstream (100 m) and downstream (100 m) from the discharge place. The results were compared with the maximum allowed concentrations set by the present legislation. It as evaluated and the quality of the surface emissaries wich collect the geothermal waste waters, by establishing the quality grades of these waters.

4.1. The impact caused by the presence of inorganic compounds in geothermal waters

In order to evaluate the environmental impact there were selected several location where geothermal wastewaters are discharged in surface waters (Fig.13).

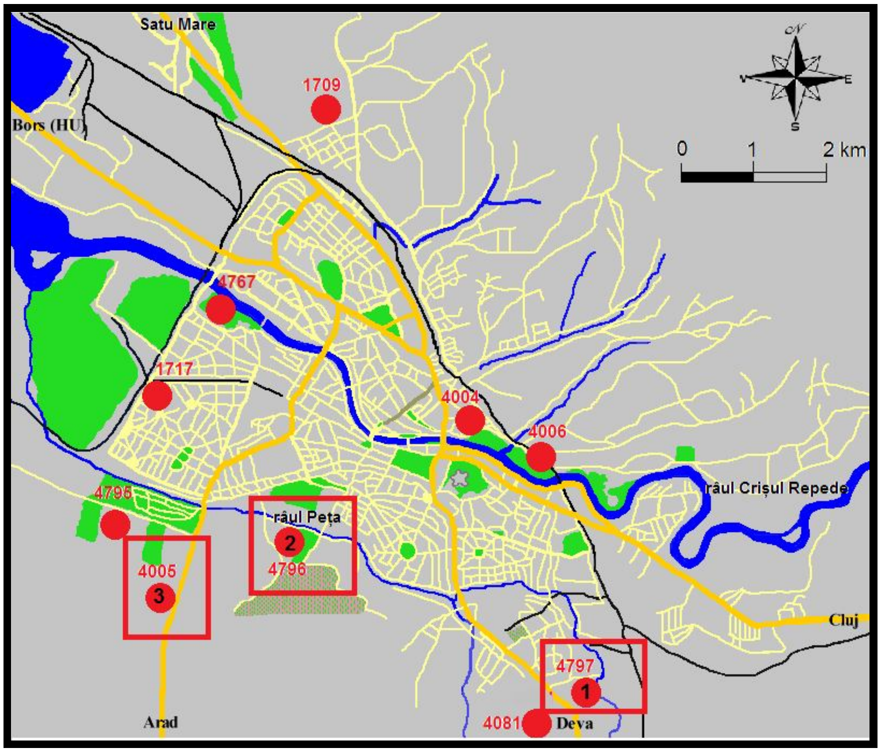
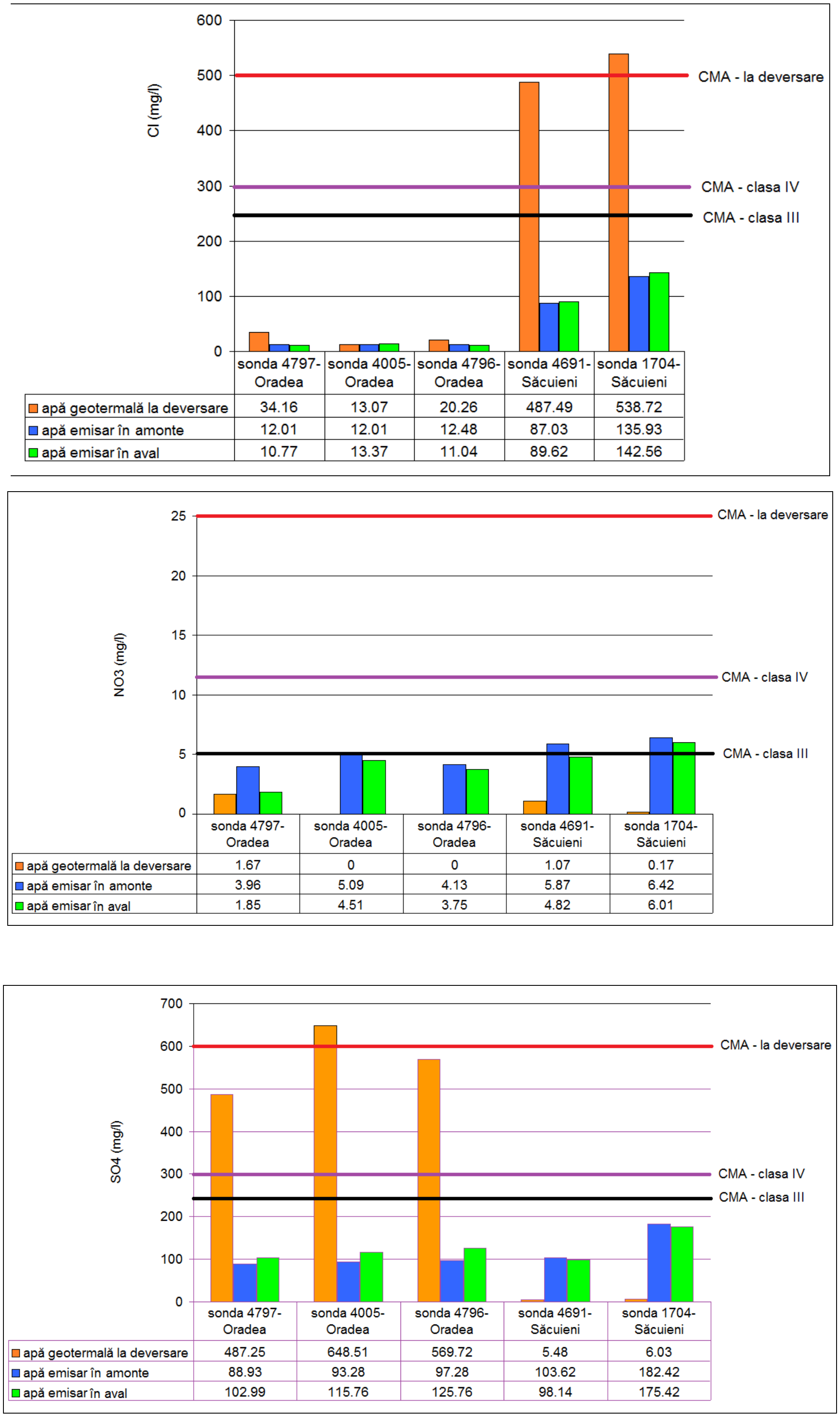


Fig.13. Location of geothermal wells in Oradea and the selected points for the evaluation of the environmental impact.

On discharge, all geothermal water samples had pH between 6.5 and 8.5, and temperatures were below 35^0C, thus respecting the limits imposed by low (NTPA 001/2005). The maximum limits for certain dissolved ions, were exceeded in some cases (Fig.14). It was exceded the maximum admissible concentration for chloride ions (well 1704-Săcuieni), sulfate ions (well 4005-Oradea), and ammonium ions (4691 and 1704 wells in Săcuieni). As a consequence these parameters require careful monitoring and solutions should be found in order to reduce their concentrations. The levels of the other dissolved ions are within the acceptable limits imposed by the low.



	sonda 4797-Oradea	sonda 4005-Oradea	sonda 4796-Oradea	sonda 4691-Săcuieni	sonda 1704-Săcuieni
apă geotermală la deversare	34.16	13.07	20.26	487.49	538.72
apă emisar în amonte	12.01	12.01	12.48	87.03	135.93
apă emisar în aval	10.77	13.37	11.04	89.62	142.56

	sonda 4797-Oradea	sonda 4005-Oradea	sonda 4796-Oradea	sonda 4691-Săcuieni	sonda 1704-Săcuieni
apă geotermală la deversare	1.67	0	0	1.07	0.17
apă emisar în amonte	3.96	5.09	4.13	5.87	6.42
apă emisar în aval	1.85	4.51	3.75	4.82	6.01

	sonda 4797-Oradea	sonda 4005-Oradea	sonda 4796-Oradea	sonda 4691-Săcuieni	sonda 1704-Săcuieni
apă geotermală la deversare	487.25	648.51	569.72	5.48	6.03
apă emisar în amonte	88.93	93.28	97.28	103.62	182.42
apă emisar în aval	102.99	115.76	125.76	98.14	175.42

Fig.14. Surface water contamination.

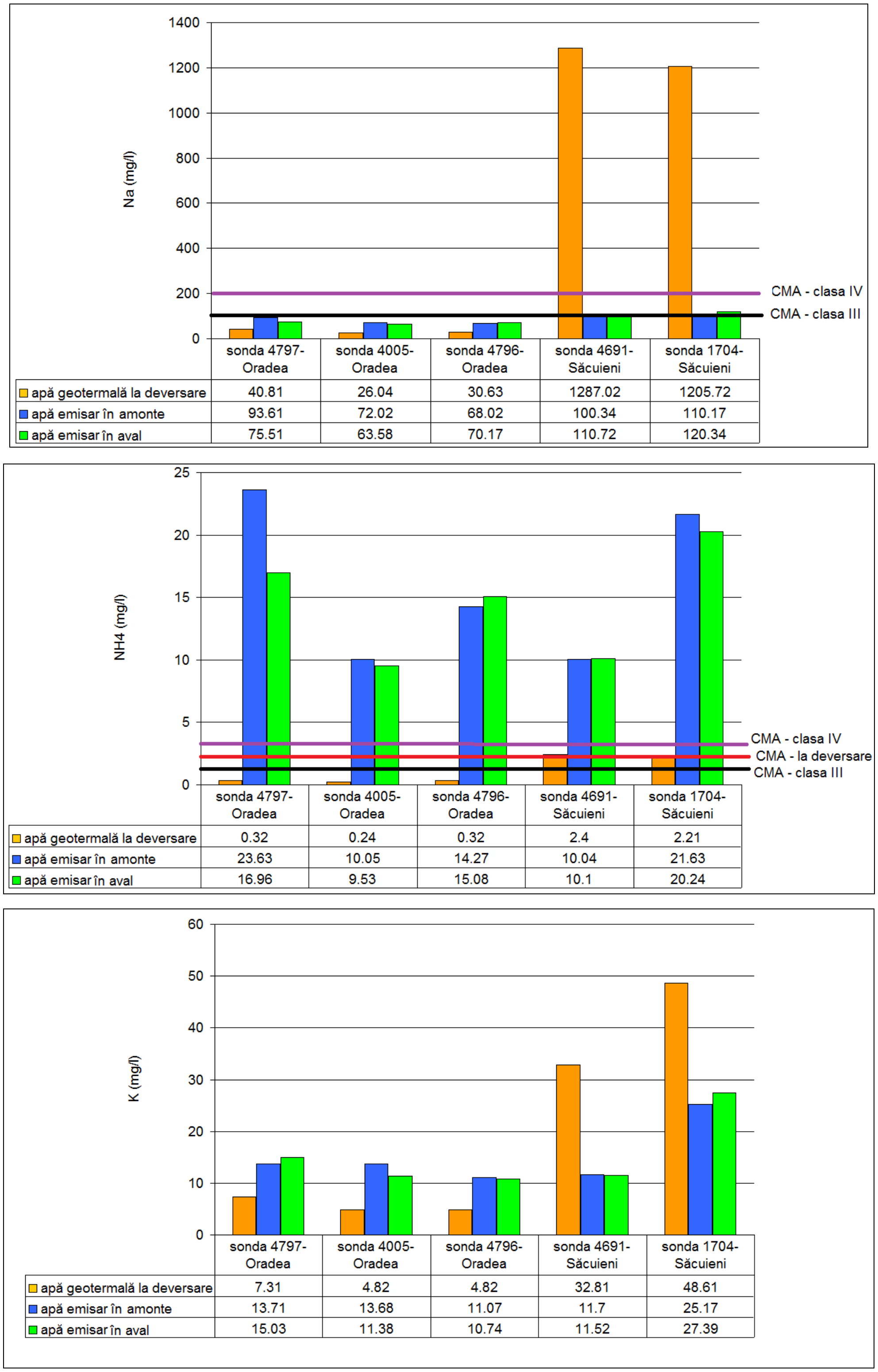


Fig.14. Surface water contamination.

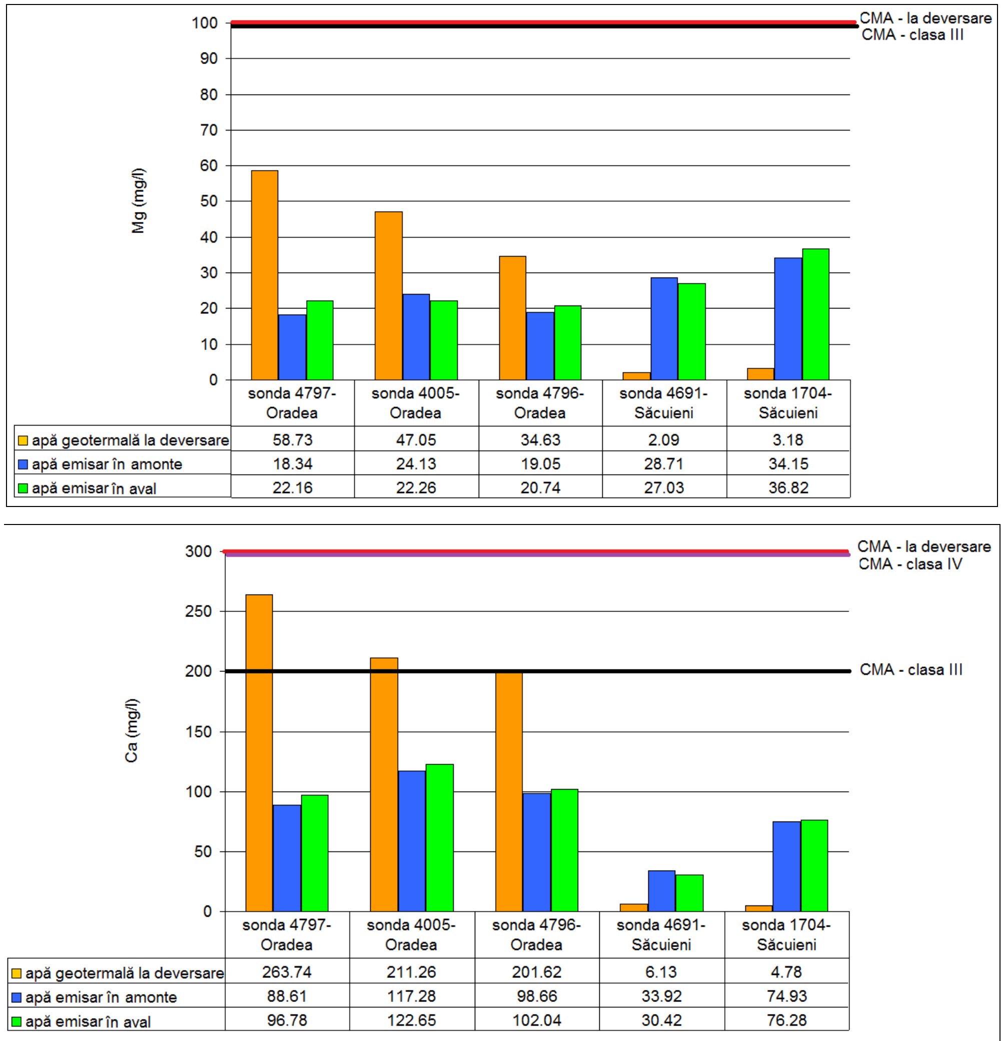


Fig.14. Surface water contamination.

Analyzing the hydrochemical features of the water sampled downstream of geothermal waste water discharge and the given maximum acceptable concentrations for each quality class, it was noticed that in the case of some parameters the surface waters belong to IV and V quality class, having a poor quality (Table 1).

Table 1. The quality of surface emissaries which collect the thermal wastewaters.

Locația	Emisar de suprafață	Cl^- (mg/l)	SO_4^{2-} (mg/l)	Na^+ (mg/l)	NH_4^+ (mg/l)	Ca^{2+} (mg/l)	Mg^{2+} (mg/l)
Oradea	râul Peța	I	II	III	V	II	II
Oradea	râul Peța	I	II	III	V	III	II
Oradea	râul Peța	I	II	III	V	II	II
Săcuieni	afluent Valea Ierului	III	II	IV	V	I	II

Săcuieni	canal de asecare	III	III	IV	V	II	II
CMA-clasa I		*25*	*60*	*25*	*0.4*	*50*	*12*
CMA-clasa II		*50*	*120*	*50*	*0.8*	*100*	*50*
CMA-clasa III		*250*	*250*	*100*	*1.2*	*200*	*100*
CMA-clasa IV		*300*	*300*	*200*	*3.2*	*300*	*200*
CMA-clasa V		*>300*	*>300*	*>200*	*>3.2*	*>300*	*>200*

The contamination of these surface waters has cumulative causes due to the geothermal waste water discharges and other upstream activities. The pollutants are transported by water currents, at different distances, depending on seasonal variation in river flow.

4.2. The impact caused by the presence of certain organic compounds in geothermal waters

- *Humic acids*

Due to the low HA content (0.55-1.38 mg/l) the geothermal wastewaters withdraw do not affect the quality of surface emissaries from Oradea. The situation is different in the case of Săcuieni, where the discharge of geothermal wastewaters may lead to adverse effects on aquatic ecosystems due to high HA contents (2.13-2.61 mg/l). As a consequence the Ier Valley, which collect the wastewater from well 4691 (Săcuieni), the HA content was 4.02 mg/l, compared to Peța River, which collect the wastewaters from several wells from Oradea, the HA level was considerably lower (0.93 mg/l).

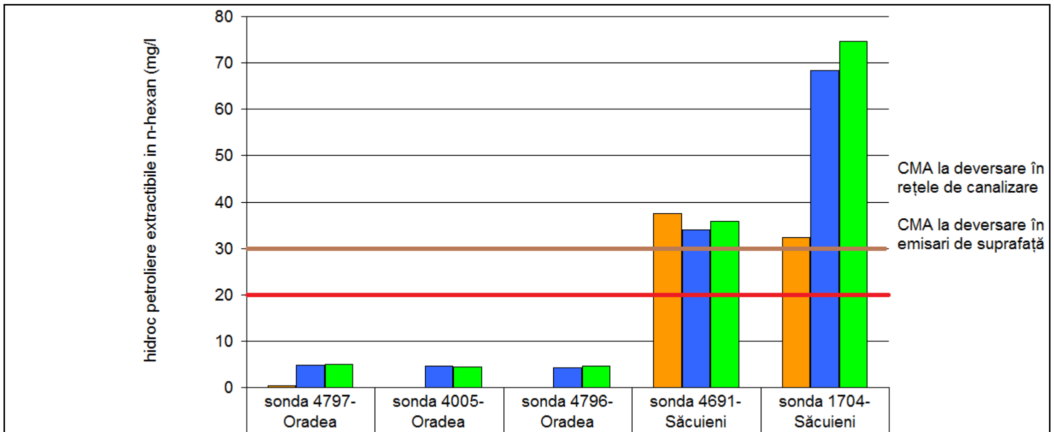
- *Phenolic compounds*

The presence of phenol and some nitro and chlorophenols in some samples originating from Săcuieni perimeter represent a risk factor for the environment, because these chemicals are an important class of environmental contaminants, their presence in low concentrations (μg/l) has severe consequences on the environment and human health. Some of the identified phenols are part of the 11 phenolic compounds included on the list of priority pollutants in the Clear Water Act [US-EPA 2002] and EU Commission Decision 2455/2001/EC.

- *Petroleum hydrocarbons*

The discharge into the environment of the geothermal wastewaters from Oradea has no negative impact because of the low level of petroleum hydrocarbons, while in Săcuieni the this activity has a negative impact due to the high content of petroleum hydrocarbons which can lead to surface waters contamination [McIntosh et al. 2010].

In Săcuieni the petroleum hydrocarbons level of the wastewaters discharged in surface waters exceeded the maximum admissible concentration (20 mg/l for surface emissaries and 30 mg/l for sewage systems) established by the law (NTPA 001/2005 and NTPA 002/2005) (Fig.15).



	sonda 4797-Oradea	sonda 4005-Oradea	sonda 4796-Oradea	sonda 4691-Săcuieni	sonda 1704-Săcuieni
apă geotermală la deversare	0.41	SLD	SLD	37.49	32.27
apă emisar în amonte	4.8	4.7	4.3	34.03	68.39
apă emisar în aval	4.9	4.4	4.6	35.79	74.62

Fig.15. Contamination with petroleum hydrocarbons of surface waters due to the geothermal wastewaters discharge.

4.3. The impact caused by the presence of radionuclides in geothermal waters

4.3.1.Drinking water contamination

Considering that the geothermal waters are used as heat agent and domestic hot water preparation, it was investigated the possible radionuclides contaminations of domestic waters. For this, geothermal and drinking water were sampled at the entrance and exit of heat exchangers. The results are shown in Fig. 17. The data proved that the radionuclides content of domestic waters did not suffered significant changes during the heat process.

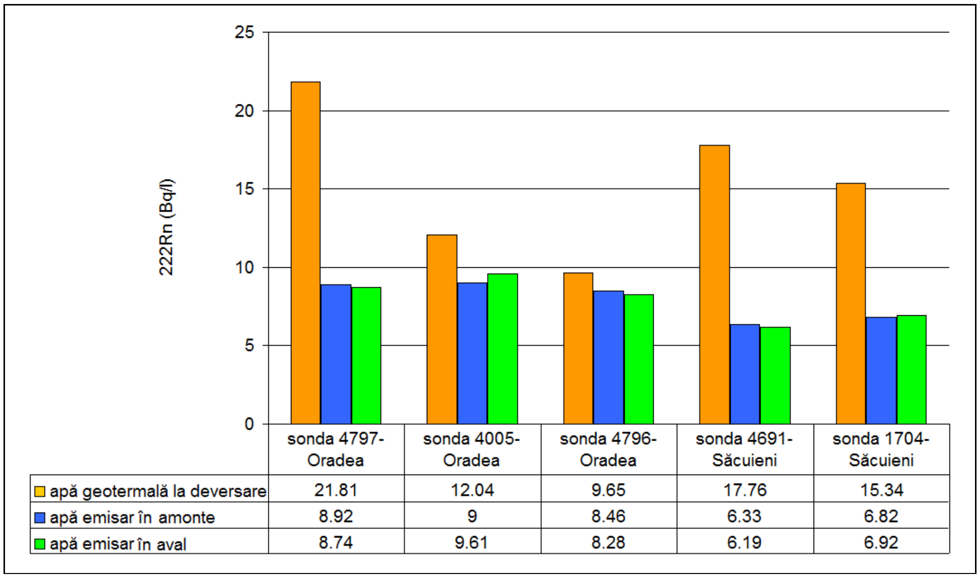


Fig.4.16. The contamination of domestic hot water with radon

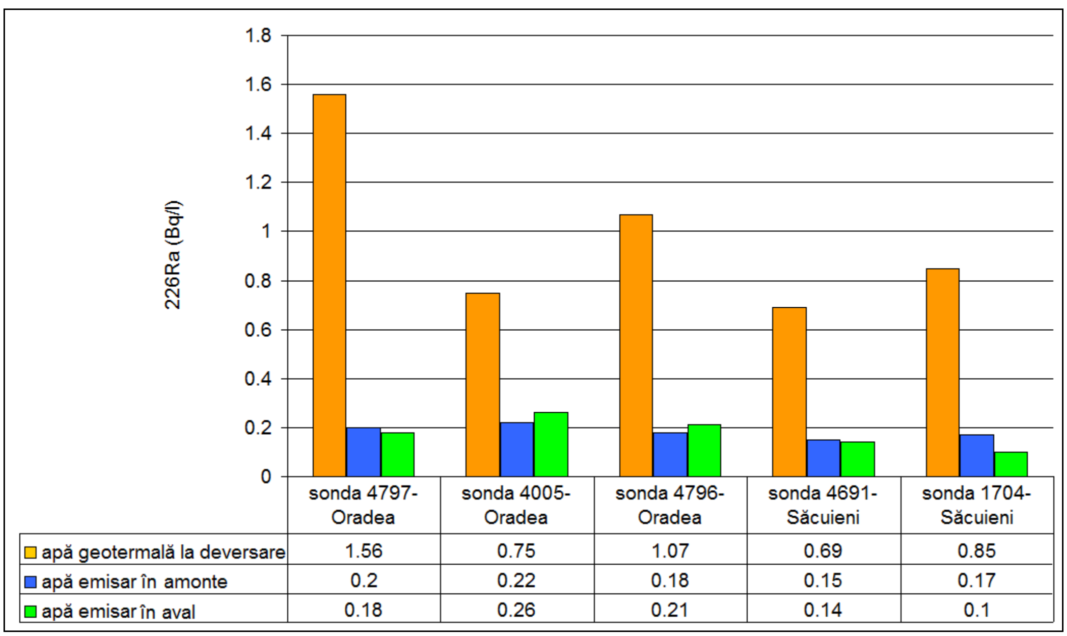


Fig.17. The contamination of domestic hot water with radium

Because the analyses proved the presence of radionuclides in the drinking waters, it was evaluated the impact on human health caused by the ingestion of drinking water originating from Oradea and Săcuieni aquifers. We must mention that radon level in drinking water samples from Oradea (9.59 to 17.05 Bq/l) and Săcuieni (9.01 to 15.05 Bq/l) was lower than the reference level set for drinking water by EU law (100 Bq/l).

To assess the impact on human health due to consumption of drinking water from Oradea and Săcuieni the effective dose due to radon and radium ingestion was calculated. The results showed that the effective dose due to ingestion of radium and radon are within the maximum levels imposed by the international law.

4.3.2. Surface water contamination

In order to evaluate the impact of the geothermal wastewaters withdrow on surface water due to the radionuclides content, it were analyzed water samples taken both upstream and downstream of the discharge point. The results did not show significant changes in radionuclides levels (see Fig. 17). Therefore in terms of content of radionuclides, geothermal wastewater discharged in the surface waters do not present risk to the environment.

4.3.3.Indoor air contamination with radon

The results showed the presence of low levels of radon in the houses heated with geothermal waters. The radon level was higher in the houses from Oradea (5.84-80,77 Bq/m^3) than in Săcuieni (17.43-30.14Bq/m^3). In both cases the radon lavel was under the maximum limit recommended by WHO, which is 100 Bq/m^3 [WHO 2009], or 200 Bq/m^3 the level recommended by ICRP [ICRP 65, 1994]. In a single case was registered a value exceeding 100 Bq/m^3, this value was registered inside the Nufãrul building, where thae radon level was 111.95 Bq/m^3. This value is below the maximum permissible limit for exposure to radon at work, which is 163 Bq/m^3.

CHAPTER V.
CONCLUSIONS

- The presence of geothermal areas in Pannonian Basin is the result of the presence of high heat flux areas. The heat excess is caused by the subcrustal magmatic processes and by the regional features of lithosphere structure.
- In this paper it were investigated two geothermal aquifers located in the eastern extremity of the Pannonian Basin: the geothermal Triassic aquifer from Oradea and the Lower Pontian aquifer from Săcuieni.
- The results of the present study have highlighted significant differences in the hydrochemical features of the two aquifers. The levels of some parameters differed by tens orders of magnitude depending of the aquifer.

- The waters originating from the two thermal aquifers are slightly acidic (Oradea: pH 6.3 to 7.61), respectively slightly alkaline (Săcuieni: pH = 7.55 to 8.15).
- The water electrical conductivity is relatively low (1107-2100 mS/cm) for Oradea, than in Săcuieni (6605-6760 mS/cm). The value of this parameter is directly correlated to the concentration of dissolved ions.
- In Oradea aquifer the water hardness is considerably higher (28.95-52.04 gr.ger) as compared to Săcuieni (1.45-2.50 gr.ger.). The high hardness of waters from Oradea is a consequence of the presence of limestone and dolomite in the area, rocks which high content of calcium and magnesium salts, as compared to Săcuieni aquifer where sand predominates.
- The content of the dissolved anion is dominated by the presence of sulfate (339.91-730.8 mg/l) and bicarbonate (165.03-275.36 mg/l) for geothermal waters from Oradea, while in Săcuieni aquifer predominate the bicarbonate (2195.37-2403.65 mg/l) and chlorine (542.87-625.21 mg/l). The major dissolved cations of the Oradea thermal aquifer are calcium (134.28-274.95 mg/l) and magnesium (32.71-71.22 mg/l), while for Săcuieni the dominant cations are sodium (1239.24-1396.21 mg/l) and potassium (39.92-58.15 mg/l).
- Considering the ratio (%) of major dissolved ions present, the thermal aquifers can be classified as bicarbonate-sulphate-calcium-magnesium (Triassic aquifer from Oradea) and bicarbonate-chloro-sodium (Pontian aquifer from Săcuieni) type.
- The humic acids level of the aquifers from Oradea is considerably lower (0.55 and 1.38 mg/l) than in Săcuieni (2.13 and 2.61 mg/l). The high content of humic acids found in aquifers of Săcuieni may be a consequence of the presence of coal layers in the area.
- The results proved the presence of phenol and phenolic compounds in waters originating from Săcuieni.
- The level oh petroleum hydrocarbon is considerably lower in Oradea (0.4-1.05 mg/l) than in Săcuieni (32.27-41.18 mg/l) aquifer.
- The radionuclides level was higher in Oradea geothermal aquifer (12.95-34.82 Bq/l for ^{222}Rn and 0.25-1.82 Bq/l for ^{226}Ra) than in Săcuieni thermal aquifer (21.05-22.88 Bq/l for ^{222}Rn and 0.83-1.19 Bq/l for ^{226}Ra).
- At the geothermal wastewaters discharge in surface waters it were exceeded the maximum admissible concentration for chloride (well 1704-Săcuieni), sulfate (well 4005-Oradea), ammonium ions (wells 4691,1704-Săcuieni) and petroleum hydrocarbons (wells 4691,1704-Săcuieni).

- For some parameters (sodium and ammonium) the surface waters which collect tha geothermal wastewaters are classified as poor quality class (IV and V).
- The contamination of the surface waters from the geothermal perimeters is the result of cumulative factors: both the activities from upstream of the geothermal perimeter and the geothermal wastewaters withdraw.
- The results proved that there is no contamination of domestic hot water due to the presence of radionuclides in geothermal waters.
- The results proved that there is no contamination of surface waters due to the geothermal wastewaters withdraw.
- The indoor radon levels in the houses heated with geothermal waters (up to 30.78 Bq/m^3 in Oradea and 23.22 Bq/m^3 in Săcuieni) is lower than maximum level recommended by WHO (100 Bq/m^3), or 200 Bq/m^3 level recommended by ICRP.
- Although the geothermal energy is classified as a clean energy, the management and exploitation of geothermal resources should be done with utmost care to avoid negative effects on the environment and human health that could result from chemical or physical contamination.
- This study indicates the presence of some potential risk factors for environmental and human health due to exploitation of the geothermal waters originating from the two areas. The risk factors are represented by the presence of high amounts of chloride (Săcuieni), sulfate (Oradea), ammonia ions, petroleum hydrocarbons (Săcuieni), phenolic compounds (Săcuieni) and radium (Oradea) in the geothermal waters.

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