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THE MINERAL WATERS FROM THE EASTERN CARPATHIANS: A CHEMICAL REVIEW

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ABSTRACT. The Eastern Carpathians are renowned for their mineral water resources, exploited since historical times for their therapeutic value. This paper summarizes the present state of knowledge on the chemical type of mineral waters from the point of view of dissolved constituents and associated gases, such as CO_2, CH_4 or H_2S. The chemistry of mineral waters is highly influenced by the type of rocks they are leaching. Considering this aspect the mineral waters from the Eastern Carpathians are mainly influenced by the Neogene to Quaternary volcanic rocks of the Călimani-Gurghiu-Harghita Mountains, the sedimentary deposits of the Transylvanian Basin and the flysch deposits of the Carpathian arc.

Four major chemical types were defined: sparkling, CO_2-rich waters, NaCl-rich waters, sulfate or H_2S-rich waters and geothermal waters. The overlapping chemical features give birth to the mixed type of waters found in the southern part of the Eastern Carpathians.

Key words: *mineral waters, chemical type, major ions, CO_2*

INTRODUCTION

The legend on the origin of mineral waters, with the shepherd, whom injured leg got healed by stepping into a bubbling pool, is told in almost every place in Transylvania where sparkling mineral waters occur.

When speaking about mineral waters or defining the notion of “mineral water”, three aspects must be taken into consideration: geological, medical and social aspects or recent trends on water consumption. These aspects, together with the actual definition of the „mineral water” have changed several times during history from the past until the present.

The first definitions of the notion of “mineral water” appeared from Hintz and Grünhut, in 1907 in the German book of baths, Deutscher Bäderbuch, defining the ”mineral water” as water containing dissolved solids above 1000 mg/l, special chemical elements, higher temperature and/or dissolved gases. Based on these assumptions

the definitions adopted by the Transylvanian authors (Bányai, 1934, Hankó 1896, Pricăjan, 1972, Straub, 1950) focused more on the origin and geological aspect of the groundwater, together with the chemical limitation of the 1000 g/l.

Nowadays the European Parliament and Commission adopted the Directive 2009/54/EC that defines the notion of mineral water as a: „microbiologically wholesome water originating in a groundwater table or deposit and emerging from a spring tapped at one or more natural bore exits. Natural mineral water can be clearly distinguished from ordinary drinking water: a) by its nature, which is characterized by its mineral content, trace elements or other constituents and, where appropriate, by certain effects; b) by its original purity. The characteristics referred to in point 1, which may give natural water properties favorable to health, shall have been assessed from the following point of view: geological and hydrological, physical, chemical and physico-chemical, microbiological, if necessary, pharmacological and clinical."

Three types of mineral waters were defined according to the total dissolved constituents: low mineral content with mineral salt content as a fix residue, not greater than 500 mg; very low mineral content, not greater than 50 mg/l and rich in mineral salts, value greater than 1500 mg/l.

HISTORICAL OVERVIEW ON THE RESEARCH OF MINERAL WATERS

In Romania, the presence of mineral waters has strong social and cultural impacts, whether the water is used for drinking water supply or for its therapeutic value. In Romania, depending on its location, special terminology is applied when speaking of sparkling mineral waters. The expressions „borcut" is used is the northern part of the country, while „borvíz" in the central parts, both meaning the same thing: CO_2 rich mineral water, similar to the expression „cevice" used in Ucraine.

The mineral water resources of the Eastern Carpathians are renowned since historic times when the first spas, like Sărățel, Anieș (Chintăuan, 1998), Borsec, Vâlcele, Odorheiu Secuiesc and the salty fountains of the Transylvanian Basin (Fischer, 1887) were constructed at several places by the Romans.

For a proper harness of these resources, they were highly examined from different perspectives: chemical, geological, medical etc., using the opportunities and technology of the era. Results are reported in different synthesizing studies, books, monographs, which discuss the therapeutic value, geology and geochemistry of the mineral waters.

The first studies on the location and therapeutic value of the mineral waters from the study area date back to the 18th Century. In this period of time the study area belonged to the Habsburg Monarchy, so the first description of the therapeutic effect, based on empirical data, of the mineral water springs and spas from Transylvania appeared in 1777, in German, written by a doctor from the court of Maria Theresia, named Crantz. In his complex database (Crantz, 1777), several locations from Transylvania, like Bodoc, Borsec, Băile Harghita, Băile Homorod, Corund, Covasna, Lueta, Băile Malnaș, Sângeorz Băi are mentioned, hosting mineral water resources.

The first correlation between the chemical composition and therapeutic effects of several mineral waters from Borsec, Bățanii Mari, Băile Chirui, Băile Ozunca, Băile Homorod, Dobogeni and Corund was reported by Kibédi, in 1766.

Similar important contributions on the location and therapeutic effect of mineral waters are given by several authors in the second half of the 18th Century and beginning of the 19th Century: Fichtel (1780) describes the area of Rodna and Anieș, Wagner (1783) provides descriptions on the therapeutic effects, Barbenius (1781) investigates the mineral waters from Sfântu Gheorghe, Covasna and surroundings, Nyulas (1800) gives information on the mineral waters from Rodna Mountains and surroundings, Gergelyffy (1811), Bélteki (1818), Pataki (1820) give important details on physico-chemical aspects on the mineral waters from Transylvania and finally Knopfler (1856) reports the first colored geological map of Transylvania highlighting the most important mineral water spas and springs.

The balneology and use of mineral waters in the 18th Century was characterized by the existence of some small important local spas for nobles: Anieș (Băile Dombhat), Rodna, Sângeorz Băi, Borsec, Vâlcele, Băile Tușnad, Sovata, Băile Selters, Lueta, Băile Chirui and different small bubbling pools, where local people met for treatment, healing or for entertainment. Often these local spas were compared to German spas, based on the similarity of the water's chemical composition or to advertise local values (Boleman, 1887, Hankó, 1891, Orbán, 1871). Some of the mineral waters were used for drinking cures and also bottling industry began to flourish at Corund, Băile Seiche, Odorheiu Secuiesc, Băile Homorod (Chintăuan, 1998, Zepeczaner, 2009)

From the late 19th Century and the beginning of the 20th Century the study of mineral waters focused more on hydrogeology and an attempt to synthesize and define the mineral water types, based on chemistry and geology was carried out. With the improvement of analytical methods new analyses were performed by Hankó (1896) and Straub (1950). A detailed comparative study on the chemical composition and origin of NaCl-rich ground waters from Transylvania was performed by Fischer (1887). Geological and hydrogeological aspects on the genesis of mineral waters were discussed (Bányai, 1929, 1934) together with the first classification of mineral waters from the Eastern Carpathians into four main groups (Bányai, 1934). For the very first time the correlation between the geological background and the chemical features of the mineral waters was discussed.

In the 20th Century important contributions to the classification of mineral waters and detailed prospecting studies on mineral water resources are given by Pricăjan, (1972), Molnár-Amărăscu, (1961), Bandrabur and Slăvoacă (1971), Vasilescu and Avramescu (1965), Pascu (1983), Kisgyörgy, (1978) based on a hydrochemical survey carried out on more than 2000 mineral water resources from the Eastern Carpathians. These analytical data are embedded in monographic descriptions in order to make it comprehensive for a wider audience Airinei and Pricăjan (1972) define geographically the area with the most active CO_2 emissions as free gas and/or dissolved CO_2, as the Post-Volcanic Manifestations Area.

Nowadays new monographic studies appeared using the previous investigations combined with chemical data (Jánosi et al., 2005, Berszán et al., 2009). Finally few data on the stable isotopic composition complete the present day state of knowledge

on the origin of mineral waters and gases from the Eastern Carpathians (Crăciun et al., 1989, Blaga et al., 1984, Berdea et al., 2005, Vaselli et al., 2002, Cuna et al., 2007, Papp, 2000, Papp and Nițoi, 2006, Magdaș et al., 2009, Fórizs et al., 2011).

MINERAL WATER TYPES IN THE EASTERN CARPATHIANS

The origin of mineral waters was highly debated within scientist. The juvenile origin of mineral waters was based on the assumption that mineral waters are fluids degassing from the hot arising magma and are new in the hydrological cycle. The more popular hypothesis of the meteoric origin, considers mineral waters as ground waters that during their underground path, as a result of water-rock interaction occurring at a certain depth and temperature have attained high mineralization and dissolved gases.

The mineral waters from Romania have a high hydro-chemical variety determined by the geological structure of the country. In the Carpathian region there are three major units influencing the chemical composition of mineral waters: the Carpathian thrust-and-fold belt, the Transylvanian Basin and the Neogene to Quaternary volcanic arc.

Several authors tried to group the mineral waters based on hydro-chemical features (fig.1, fig. 2).

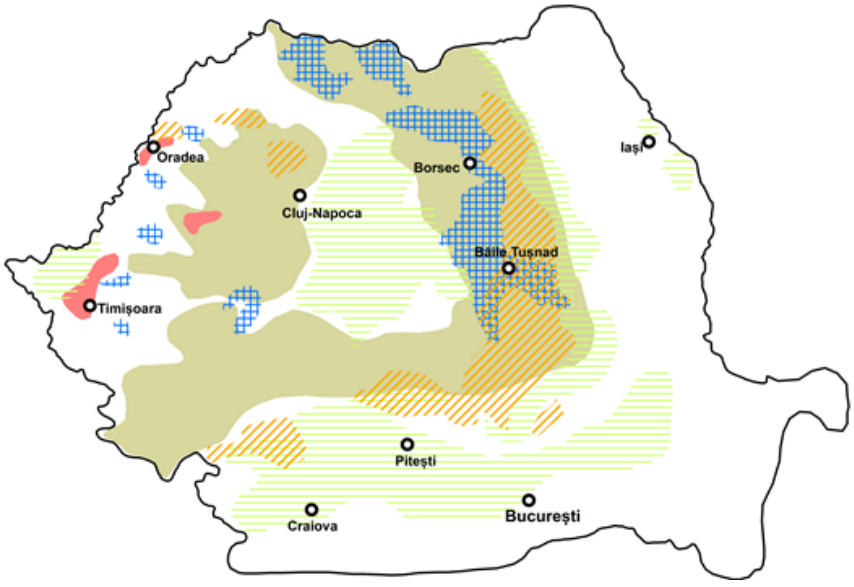

Fig. 1. *Sketch figure of the main mineral water types of Romania with special focus on the Eastern Carpathians: CO_2-rich mineral waters (blue grid), NaCl-rich mineral waters (green lines), sulfate and H_2S-rich mineral waters (orange lines), mixed waters (overlapping of different types, ex. in the SE Carpathians), geothermal waters (red areas), modified after Pricăjan, 1972.*

Bányai (1934) defines 5 mineral water types: CO_2-rich mineral waters in the proximity of the volcanic chain, NaCl, high salinity waters, in the Transylvanian Basin, H_2S-rich mineral waters in the Carpathian flisch area, mixed type of waters and geothermal springs. Airinei and Pricăjan (1972) and Molnar-Amărăscu (1961) define only 3 main groups based on the hydro-chemical study of more than 2000 mineral water resources from Romania, prospecting the mineral water resources suitable for balneal therapy having particular features (specific ion/trace element content, temperature, radioactivity) depending on the geological setting. A series of dissolved gases, such as CO_2, CH_4, N_2, H_2S, Ar and He, are associated to the mineral waters.

Sparkling (CO_2-rich) mineral waters

In the Eastern Carpathians more than 2000 mineral water springs are estimated according to the study of Bányai (1934) and Pricăjan (1972). Based on the study of Pricăjan (1972), the total area covered by igneous rocks in Romania is 7370 km^2. Where the most abundant natural CO_2 emissions were measured, either as free gas or dissolved in the water, the term Post-Volcanic Manifestations Area was introduced Airinei and Pricăjan (1972). The area of the whole Post-Volcanic Manifestations Area is 13000 km^2 and half of which belongs to the Călimani-Gurghiu-Harghita Mountains. This covers also the neighboring places of the volcanic edifices. The most powerful manifestations appear in the neighborhood of the volcanoes, especially in the eastern part, in Gheorghieni, Ciuc and Baraolt Basins. Due to the highly tectonized region and the presence of faults which facilitate fluid circulation, the sparkling mineral waters are found up to 50 km distance from the volcanic ranges (Bányai, 1934).

The CO_2 appears as “dry” emanations (mofettes) or as dissolved gas in the groundwater. Depending on the type of rocks water is leaching, in the CO_2-rich mineral waters the dominant ions are HCO_3^-, (present as CO_2 dissolved in the water). The presence of CO_2 in the water increases its dissolution capacity, and water gets enriched in dissolved salts (Pricăjan and Airinei, 1979). The main cations are usually Na^+, Ca^{2+}, Mg^{2+} and Fe^{2+}.

In the proximity of the volcanic chain or the sub volcanic area of Bârgăului Mts. the mineral waters are hosted either in the lavas and massive volcanic rocks or in volcano-sedimentary formations, pyroclastic flow, tuffs and Quaternary deposits. These are characterized either by low discharge, of Ca-Mg-Fe-HCO_3 type with high Fe and H_2S content (eg. mineral waters from Valea Vinului, Anieș, Băile Harghita, Sântimbru-Băi, Vlăhița, Băile Homorod, Băile Chirui) or with high discharge, suitable for bottling, represented by mineral waters from the Ciuc Basin, Sâncrăieni, Băile Tușnad, Baraolt Basin, Biborțeni of Ca-Mg-HCO_3 type (Pricăjan and Airinei, 1981).

The mineral waters get enriched in Fe due to the alternation of several minerals, such as augites, amphiboles, pyroxenes, sandstones with spherosideritic nodules (Bányai, 1934). In particular cases some rare elements like As is present in the composition of mineral waters (Pricăjan, 1972). Mineral waters are present in the crystalline area, circulating through the fractures of dolomitic limestone (eg. Borsec, Bilbor) of Fe-HCO_3 and Ca-Mg-HCO_3 type, with high $CaCO_3$ content. As the mineral waters come to the surface the underground pressure of the water decreases, Ca and Mg precipitate from the solutions and create travertine deposits (Pricăjan and Airinei, 1981).

In the Carpathian flysch area mineral waters with high Na and Cl content are present of Ca-HCO_3-Cl and Na-HCO_3-Cl type (eg. Cașin, Repat, Bodoc, Malnaș, Covasna, Vâlcele, Zizin, Turia-Balvanyos, Slănic Moldova (Bányai, 1934, Pricăjan and Airinei, 1981).

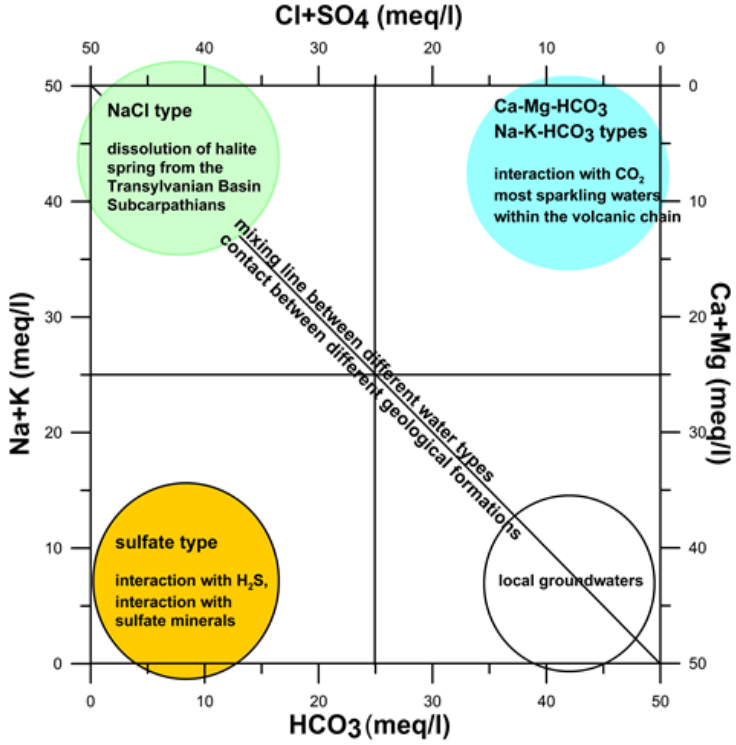

Fig. 2. *The mineral water types on Langelier-Ludwig diagram, modified after Pricăjan, 1972 and Vaselli et al., 2002.*

NaCl-rich mineral waters

The NaCl-rich waters are largely represented in the Transylvanian Basin, and are related to two major sources: the Miocene sedimentary deposits (salt diapires) and the hydrocarbons (Bányai, 1934, Pricăjan and Airinei, 1972).

In the area of the Miocene (Badenian) salt deposits the ground waters get enriched in NaCl by leaching directly the salt diapires or salty clays and marls. Mineralization of such brines increases almost until NaCl saturation, reaching values of more than ~250 g/l. They can be found mostly in the edges of the Transylvanian Basin, where the salt outcrops are present at several locations eg. Băile Someșeni, Bistrița, Dumitra, Tăure, Mintiu, Ideciul de Jos, Jabelnița, Sovata, Praid, Corund, Lueta,

Mărtiniș, Merești, Crăciunel, Sânpaul, Racoș etc. The groundwaters come to surface as natural springs, fountains, lakes and through deep boreholes (Fischer 1887, Bányai, 1929, 1934). Besides the high NaCl content the groundwaters are enriched in other ions, such as Ca, Fe, SO_4, CO_3, Br, I most probably due to the diversity of rocks they are leaching; however the hypothesis of being remnants of old seawater captured during the basin sedimentation processes, traced by the presence of Br and I content, was not excluded (Molnár-Amărăscu, 1961).

Another specific group of NaCl-type of waters is represented by groundwaters of salinity around 50 g/l very briefly discussed by some authors (Bányai, 1934, Molnár-Amărăscu, 1961). The hypothesis of Bányai (1929) and Molnár-Amărăscu and Pricăjan (1961) on the origin of such mineral waters, identified at Praid, Cound, Odorheiu Secuiesc, Cristuru Secuiesc, Homorod Brașov is that their source is from Sarmatian deposits and can be considered old formation waters typical of CH_4 deposits and mud-volcanoes. The geological studies confirmed the presence of CH_4-domes and bituminous shale at several areas where such ground waters come to surface. NaCl, oil-field-brines are present also in the Carpathian flysch area having similar chemical composition.

Sulfate and H_2S-rich mineral waters

Present in a wide range of geological formations, the sulphate-rich mineral waters are discussed as having a twofold origin: post-volcanic and organic. The post-volcanic CO_2 exhalations are often accompanied with H_2S. Within the volcanic chain there are several places where the sparkling mineral waters have the typical smell of sulphur suggesting the presence of H_2S eg. Balvanyos, Lazaresti, Sântimbru Băi. Sulphur rich NaCl type waters may be related to organic matter, hydrocarbons eg. springs from the Transylvanian Basin where CH_4 is often accompanied by H_2S.

Waters containing H_2S are present in the Carpathian flysch area, in sedimentary and crystalline structures (Micloșoara, Belin, Covasna, Ghimeș etc.) as a result of the alteration of sulphides. The water is often characterized by a white-colored precipitate marking the dissolved sulphur (Bányai, 1929).

Mixed waters

The circulation of CO_2 as it was previously mentioned while discussing the sparkling mineral waters is highly influenced by the tectonic setting of the area. The presence of tectonic lineaments, faults and fractures facilitate the circulation of CO_2 to distances up to 50 km from the volcanic edifices.

Where the CO_2 emissions meet the sedimentary deposits of the Transylvanian Basin the mineral waters have high salinity and dissolved CO_2 content. Such ground waters are found typically in the western slope of the volcanic chain, where the volcanogenic sediments have covered the older sedimentary deposits of the Transylvanian Basin (Bányai, 1929, Kis et al., 2012). This geological setting gives a

particular chemistry of the mineral waters, which gain their dissolved constituents from multiple sources. Circulating through volcanic rocks water is able dissolve Fe, Ca, Mg, while from sedimentary deposits they get enriched in Na, Cl and SO_4. The large amount of CO_2 dissolved in the water increases the HCO_3 content of the water.

In the Eastern Carpathians-Transylvanian Basin boundary mixed waters are considered some mineral waters from Sângeorz Băi, Parva, Anieș, Praid, Corund, Odorheiu Secuiesc, Lueta, Vlăhița.

Geothermal waters

Due to frequent geothermal anomalies in several areas among the volcanic chain, the temperature of some mineral waters may reach up to 20ºC. Such mineral waters can be found at Băile Tușnad, Toplița, Baraolt Basin, Ciuc Basin, Vlăhița, considered the remnants of geysers (Bányai, 1929) and recently the result of the still cooling magma chambers of the youngest volcanic edifices (Vaselli et al., 2002).

From the 20th Century the hydrogeological research focused on the prospecting of thermal and mineral waters for establishing new balneological treatment centers of for bottling industry. They began to study the origin of groundwater and to identify possible water resources suitable for economic capitalization (Crăciun et al., 1989).

CONCLUSIONS

The Eastern Carpathians represent the most important area from the point of view of the abundance of mineral water springs and spas. Four major types were defined according to the physical and chemical features of the waters: sparkling (CO_2-rich), NaCl-rich, sulfate/H_2S-rich, and geothermal waters. The different types of waters often overlap giving birt to the mixed type of waters, present especially in the southern part of the Eastern Carpathians.

The chemical features are mostly influenced by the geological structure of the area. In the Eastern Carpathians the mineral waters are influenced by the Neogene to Quaternary volcanic chain of the Călimani-Gurghiu-Harghita Mountains, the sedimentary deposits of the neighboring Transylvanian Basin and the Carpathian flysch.

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