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Vulnerability to pollution of thermomineral sources in Băile Tușnad area

¹Iulian Popa, ¹Mihaela Scrădeanu, ²Augustin Jenu, ²Daniel Scrădeanu

¹ University of Bucharest, Faculty of Geology and Geophysics, Bucharest, Romania; ² Romanian Association of Hydrogeologists, Bucharest, Romania. Corresponding author: I. Popa, julip_2006@yahoo.co.uk

Abstract. The assessment of vulnerability to pollution of thermomineral sources exploited in Baile Tusnad area was initiated by a bypass road project crossing the hydrogeological protection area of these sources. The investigations conducted in hydrographic and hydrogeological basin of thermomineral sources had the following specific objectives: land use, terrain slope and run-off, spatial and parametric model of the vadose zone, conceptual model of the hydrostructure with a special attention paid to the age of thermomineral water. In order to assess the age of thermomineral waters was compiled an isotopic sampling program with a total of 10 samples for: hypothermal groundwater, fresh mixed with mineral groundwater (Olt meadow area) and surface waters (Olt River and Ciucas Lake). The environmental isotopes used were deuterium, oxygen 18 and tritium. The vulnerability of Tusnad hydrostructure is differentiated in four distinct areas. The most thermomineral sources are placed in areas with medium and high vulnerability.

Key Words: thermomineral sources, bypass road, environmental isotopes, vulnerability map.

Introduction. The main objective of the present study is to assess the vulnerability and the potential impact related to the construction of a bypass road for DN 12 which will cross the hydrogeologic protection perimeter of the thermomineral sources exploited in the Băile Tuşnad area.

The purpose of the bypass road is to improve the traffic flow by diverting the cars outside the localities Băile Tuşnad, Tuşnadul Nou and Tuşnad.

The proposed bypass road variant has a total length of 11,45 km, with starting point km 44+160 and ending point km 55+860 on DN12 (Figure 1).

Based on the field observations and consulting the available documentations (Search Corporation, Prospecțiuni S.A.) the following preliminary observations were drawn:

- the area to be crossed by the bypass road, in the vicinity of the thermomineral sources, is constituted by highly permeable deposits (volcanogenic-sedimentary formations);

- the slope stability is weak, the actual equilibrium being assured by the vegetation; a strip of land of 30-40 m wide is going to be deforested along the bypass road.

This study used the information obtained during several stages of investigation:

- mapping of recharge area for volcanogenic-sedimentary formations, as they represent the main deposits within the protection zones of the thermomineral sources;

- sampling of the sources from the area, for two major purposes:

- chemical characterization of mineral and thermomineral waters,

- isotopic study of the groundwater, in order to get an overall image concerning origin, age, time of residence and eventually recharge pattern from phreatic or surface water.



Figure 1. Hydrographic basin (catchment) in Băile Tuşnad area crossed by bypass road variant.

Material and Method

Location and climatic elements. The Ciuc Depression consists of a line of Neogene-Quaternary intermountain basins, generally oriented NNW-SSE, and placed between Harghita Mts to the west, Hăghimaş, Ciuc and Bodoc Mts to the north, east and south (Geamănu & Popescu 1998).

The studied area is located around Băile Tuşnad, on the left side of the Olt Valley, in the southern part of the Lower Ciuc Depression. The area can be accessed by DN 12 road or by CF 400 Bucureşti-Deda railway. The distances to the main localities in the area are: 32 km to Miercurea Ciuc, 37 km to Sfântu Gheorghe, 35 km to Baraolt and 67 km to Braşov.

The geographic position, elevation (622 m) and wooded steep slopes have determined the existence of a mountain subalpine climate. Thus, the precipitations

recorded in the area are about 800–850 mm, with uneven distribution throughout the year. The most are recorded in April–June period while the less fall in half August to October interval. The snowpack remains, in average, 80-100 days, being more persistent on the north-western slopes. The annual average temperature is around $+5^{\circ}C$ (July average value $+15^{\circ}C$, January average value $-7^{\circ}C$) and duration of sunshine around 1800 hours/year (Geamănu & Popescu 1998).

The geomorphology and hydrology of the area. The Băile Tuşnad hydrostructure is located in the southernmost part of Harghita volcanic massif, between Harghita and Bodoc Mts. In this region the Olt River erosion generates a narrow gorge, with 550-750 m differences from the adjacent areas (Figure 2).



Figure 2. Geomorphologic framework of the study area (Geamănu & Popescu 1998): 1. volcanic mts; 2. flysch formations; 3. depressions; 4. harghita mts. boundary; 5. Ciomadul massif boundary; 6. crater; 7. secondary volcanic doms; 8. interfleuve; 9. defile; 10. pass.

The studied area is developing on the east from the Olt Valley, in the lower bassin of the affluents Vârghiş, Tiszas, Benevize and Cetaţii Valley. The hydrographic basin of these valleys follows to the east the volcanic peaks of Ciomadul Massif: Ciumatul Mic (1238 m) – Ciumatul Mare (1300,8 m) – Mohoş (1182 m) – Haromul Mare (1140,7 m).

The Olt afluents from west side (Pilişca Massif) and east side (Ciumatul Massif) has different lenght. Thus, those from the west (right) side are a litle bit longer (1.0 - 2.5 km), has a large catchment area and a strongly widened lower stream. Although the flow is permanent, the discharges present high variations, due to precipitations. From the western (right) affluents the most important are Minelor Valley and Corbului brook. The eastern (left) affluents have under 1 km lenght, high longitudinal slope, less depth and characterised by a temporary flow. The most important surface water stream is Cetății brook, with the bassin spreaded on both the Cetății and Surduc volcanic doms.

In these hydrographic conditions, the Olt River has an multiannual mean value of the discharge of 9 m³/s. Comparing with that value, the discharge varies from $4.5 - 5 \text{ m}^3$ /s during the dry periods to almost double in the raining ones.

Geologic and tectonic regional settings. The geologic researches performed in the area are synthesized in the geological map scale 1:50.000 – sheet Sânmartin (IGR 1973) and geological map scale 1:200.000 - sheet Odorhei (IGR 1968) - (Figure 3).

The geological formations of Ciomadul volcanic structure lays directly on the bedrock constituted by Cretaceous folded flysch of Carpathian nappes (Seghedi et al 2001).

The volcanic activity in the area started with ash and andesitic lava fragments from Mohoş and Sf. Ana area, which were deposited in sublacustrian environment. Then follows andesitic lava flows and in the final eruptions which produced lava accumulations in some secondary volcanic doms on the Ciomatul Mic – Ciomatul Mare-Vârful Cetății – Harom ligne.

Nowadays the volcanic activity continues only with gas emissions: cold or warm gases rises from the depth on fractures and emerge at the surface as dry CO_2 , H_2S or dissolved in water ("borviz") (Seghedi et al 2001).



Figure 3. Geologic map of Tuşnad Băi – Miercurea Ciuc area (from sheet L 35 XIV- Odorhei, scale 1:200.000, IGR 1968).

Regional geology. The opening of Ciuc Depression basins started in medium Miocene and their evolution continue till Pleistocene, with a general direction from north to south.

The bedrock is constituted by Dacidic units (Middle Dacide units from basement and Outer Dacide units composed by cretaceous flysch nappes). From lithologic point of view there are sedimentary formations: marly limestones and sandstones with diaclase limestone - Sinaia beds facies (Figure 4).

The Neogene-Cuaternary volcanism has been predominant explosive but also with long effusive periods. Within the geological structure two zones have been separated:

- the lower zone, made by volcanic products of the first stage, developed in Pannonian period, is included in volcanogenic-sedimentary formation; represents the base of the volcanic structure;

- the upper zone, resulted from the last volcanic stages developed in Upper Pannonian – Lower Quaternary period.



Figure 4. Geological cross-section between Tuşnad Băi and Sf. Ana Lake (by Geamănu & Popescu 1998).

In the Băile Tuşnad area there are two volcanic structures very well described:

- Pilişca structure, on the right (west) side of the Olt valley, being active during Upper Romanian – Lower Pleistocene (2.4-1.5 Ma);

- Ciomadul structure, on the left (east) side of Olt valley, being active from middle Pleistocene till less than 100000 years ago.

The Pleistocene and Holocene deposits outcrop on the largest surface within the Lower Ciuc Depression. From lithologic point of view there are: gravels, sand and clay as terrace deposits from Middle - Upper Pleistocene, proluvial deposits (Middle-Upper Pleistocene to Holocene), slope deluvial deposits (Upper Pleistocene - Holocene), meadow and swamp deposits (both of Holocene age).

Regional scale tectonics. From tectonic point of view the Lower Ciuc Depression have been installed on a sinked basement sector, between two rised basement sectors: Harghitei Mts to the west and Ciuc Mts to the east. The geological and geophysical data prove that the basement fragmentation is due to a fractures system generally oriented NW-SE or NNE-SSW (Figure 5).

This tectonic pattern of the basement represents the major factor of thermomineral sources genesis in the area. According to Airinei & Pricăjan (1972), Tuşnad area belongs to 3rd alignment of sparkling mineral waters and dry mofette. The group of sources with carbonate mineral waters from Lower Ciuc Depression is located on two alignments oriented north to south from which one is along the Olt valley and the second cut the tributary valleys from the eastern slope of Harghita Mts.



Figure 5. Tectonic features correlated with mofette occurrences in Ciuc Depression (Airinei & Pricajan 1972).

Geologic and tectonic local settings. At the local scale the geology of the zone is mainly represented by magmatic and eruptive rocks (andesites, microdiorites, andesitic agglomerates and pyroclastites), laid on flysch type Cretaceous deposits (Sinaia beds). The later ones are highly folded and tectonized being able to store groundwater and to generate sources of different chemical types. All these are laid on blocks divided bedrock.

The eruptive formations are represented by andesites and andesitic agglomerates, on the west (right) side and pyroclastic type volcanogenic-sedimentary deposits on east (left) side of the Olt valley (Figure 6). Pyroclastites in the Băile Tuşnad area consist of alternation of andesitic sand and gravel, resulted from aquatic sedimentation. Generally the thickness of pyroclastic formation increase west to east, from meters to more than 100 m.

The exploration studies performed in Băile Tuşnad area proved that, beneath the volcanic deposits there is a stack of Neocomian marly and clayey sandstone similar to Sinaia beds facies. Close to the Olt valley these deposits are near the surface while laterally, to the east and west, they are found at increasingly higher depth.

The sedimentary formations are represented by Quaternary alluvial and delluvial deposits.

A chronologic sequence of the rocks in Băile Tuşnad area comprise:

- the oldest eruptive rocks = andesites and andesitic agglomerates with amphibole and pyroxene, from the lower part of Olt right side;

- andesites with amphibole and biotite, to the west;

- pyroclastites followed by andesites and andesitic agglomerates with amphibole and biotite.

The synthetic sequence is: basic andesites, pyroclastites, acid andesites.

The complex lithology and tectonics are the main factors that controls the hydrogeologic and hydrochimic features of thermomineral sources in the area.



Figure 6. Volcanological and hydrothermal setting in Băile Tuşnad area (Mitrofan 2000; geology after Pecskay et al 1992). 1. Volcanogen-sedimentary formation; 2. Pilişca volcanic structure: a-lava flows, b–lava domes; 3. Ciomadul volcanic structure: a-lava flows and domes, b-pyroclastics; 4. Quaternary: a-aluvium, b-swamp; 5. Fault; 6. Remnant of crater rim; 7. Thermal spring; 8.

Geothermal well; 9. Shallow thermal gradient (°C/km)

Water chemistry in Băile Tuşnad area. Cold mineral and thermomineral waters in Băile Tuşnad are distinct from the chemical point of view:

- calcium-bicarbonate water – the cold mineral waters;

- chlorosodium water - the thermomineral waters.

Cold mineral waters come from phreatic aquifer located in pyroclastic rocks or in the alteration and cracked area of magmatic rocks. They have variable flows and are dependent on the rainfall regime.

Thermomineral waters originate in deep aquifer located in the upper horizon (clayey-marl, sometimes gritted) of Cretaceous flysch. The Cretaceous flysh is strongly fissured and covered by a stack of pyroclastites of few meters in Olt Valley and up to 100 - 150 meters at the eastern limit of the river basin (left slope) (Elek 1984).

The thermomineral waters coming from great depths dissolves an important quantity of mineral elements in sedimentary and magmatic rocks through they are moving. The process is accelerated by the presence of carbon dioxide originated from a mantle-derived magma reservoir (Vaselli et al 2002) which, being dissolved in water, it gives a higher aggressiveness. This is why the mineralization of those water is greater than 5000 mg L^{-1} (Elek 1984).

The exploited mineral water sources. Băile Tuşnad hidromineral structure is currently exploited through six mineral water sources, situated on the left bank of the Olt River, separated into two categories:

- mineral water sources for external cure:
 - Ileana Spring and Ana Springs (7 and 8),
- mineral water sources for internal cure:
 - Apor Spring, Mikes Spring, Stănescu Spring.

The current characteristics of the sampled sources. The current chemical status of the waters in the area was evaluated on the basis of a total of 6 samples collected (during September 2012) from Stănescu Spring, Ileana Spring, Rudi Spring, Mikes Spring, OJT well and Marton Spring.

The samples have been analysed at ECOIND Bucharest laboratory. The results are presented synthetically in the ternary diagram (Figure 7), Piper diagram (Figure 8) and Scholler diagram (Figure 9).



Figure 7. Ternary Diagram – sampling from 15.09.2012.





To view the relative concentrations of the main ions from the sampled springs, a standardization of the concentration values expressed in milligrams per litre has been made, on the maximum interval [0; 1] (Table 1). Also has been made a representation of all the analysed samples (Figure 10) and it results the preservation of general chemical characteristics as in 2001, the argument of the stability of the hydrodynamic conditions.



Results and Discussion. The analysis of current chemical characteristics of mineral waters from Băile Tuşnad area, in terms of impact assessment completion of the bypass road, leads to the following results: comparison of the current chemical characteristics (September 2012) of mineral and thermomineral springs in the area with those from Elek (1984) and those performed in 2001 by S.C.Tuşnad S.A. indicate a relative chemical stability.

Chemical stability of mineral and thermomineral waters, in spite of the variability of flows (there were periods of declining flows – Ileana spring or their complete disappearance - **Stănescu** spring), indicates a slow motion groundwater system, with an important deep component. The deep component is argued by the high mineralization and chloro-sodium character of thermomineral springs.

There are two types of mineral waters in Băile Tuşnad area: calcium-bicarbonate water - the mineral cold ones and chlorosodium water - the thermomineral waters (Crăciun & Bandrabur 1993). Existence and stability of the two types of mineral waters over a lengthy period of time argues:

- the existence of two hydrostructures (the deep one, located in the flysch formations and the shallow one located in the unconsolidated deposits of the pyroclastites);

- the descending dominant direction of hydrodynamic links between the two hydrostructures.

Communication between the two hydrostructures is made on intricate paths by percolation of precipitation water through the stack of permeable unsaturated pyroclastites, poorly consolidated. For these reasons, the vertical movement of water is

very slow and it ends up in deep hydrostructure of flysch after about 30 years, isotopic age determined.

The share of surface water in chlorosodium water springs is reduced and for its accurate quantitative assessment are required detailed studies over long periods of time.

Ions concentrations [mg L ⁻¹]							Standardised relative concentrations on [0,1]						
Sprina	RUDI	STANESCU	FOIT	MARTON	II FANA	MIKES	Snrina	RUDI	STANESCU	F OIT	MARTON	II FANA	MIKES
Na	1032	264	380	57.2	37	835	Na	1	0.23	0.34	0.02	0	0.8
CL	2183	532.5	390.5	2059	62.13	92.3	CI	1	0.22	0.15	0.94	0	0.01
HCO3	826.5	377.3	983.9	1098	118.1	3640	нсоз	0.2	0.07	0.25	0.28	0	1
Rez.sec	4301	1171	1595	2864	243	3640	Rez.sec	1	0.23	0.33	0.65	0	0.84
SO4	23.14	19.31	27.12	34.02	45.51	519.4	SO4	0.01	0	0.02	0.03	0.05	1
Са	300	76.8	147	88.1	24.1	252	Ca	1	0.19	0.45	0.23	0	0.83
Ма	73.4	29.6	71.1	10.7	9.39	189	Mq	0.36	0.11	0.34	0.01	0	1
ĸ	270	65.7	68.3	8.64	9.5	112	ĸ	1	0.22	0.23	0	0	0.4
Fe	10.33	6.15	8.77	8.22	0.03	0.35	Fe	1	0.59	0.85	0.8	0	0.03
NH4	14.4	7.48	14.4	9.27	1.87	1.25	NH4	1	0.47	1	0.61	0.05	0
CCOMn	7.19	1.92	4	11.03	2.87	2.07	CCOMn	0.58	0	0.23	1	0.1	0.02
NO3	25.78	20.26	20.87	20.26	45.12	25.78	NO3	0.22	0	0.02	0	1	0.22
02	1.02	0.73	0.61	0.42	3.84	1.8	02	0.18	0.09	0.06	0	1	0.4
В	45.9	13.2	9.8	0.46	1.01	8.23	В	1	0.28	0.21	0	0.01	0.17
NO2	0.024	0.031	0.049	0.018	0.006	0.12	NO2	0.16	0.22	0.38	0.11	0	1
FI	0.66	0.99	1.36	1.19	0.04	0.07	FI	0.47	0.72	1	0.87	0	0.02

Table 1 Ions concentrations [mg L⁻¹] and standardised relative concentrations on [0,1] interval

Vulnerability of exploited mineral waters. Generally, the vulnerability concept refers to the sensitivity of an aquifer system to be polluted by contaminants released at the surface or shallow depth. The key elements for the assessment of aquifer vulnerability to contamination are:

- variations of recharge mechanisms of groundwater;

- natural attenuation capacity of soil and subsoil.

Vulnerability assessment and mapping represent an important component of groundwater protection policy. The vulnerability map has three main goals:

- zonation of the groundwater contamination likelihood;

- establishing a protective scheme not to much restrictive concerning the anthropic activities;

- to choose the prevention measures related to potentially hazardous activities located in low vulnerability areas.

The vulnerability zoning of the left side hydrographic basin in the Tusnad Bai area is based on the following factors:

- slope of the land – shows minimum values near the Olt valley and in the northern part of the area, and maximum values on the western side of the Ciomadul Massif, Cetatii and Surduc Peaks; has direct influence on surface runoff and infiltration rate;

- hydrographic network – reduced development in the area, comprise 4 valleys characterized by small length (less than 1 km long), high slope, relatively small depth and temporary total runoff; it represent preferential pathway for infiltration.

- unsaturated zone – described both by previous geological studies and more recent geotechnical and exploration boreholes; on the left side of the Olt valley are prevalent the pyroclastic deposits with thickness increasing west to east from meters to around 100 m. The geotechnical boreholes drilled for this new bypass road have 25 m depth and are completed in the pyroclastic rocks. The groundwater presence is local and as infiltrations which confirm the good permeability of these pyroclastic deposits.

Depending on the characteristics of this zone (thickness, texture, organic matter and clay minerals content, moisture and hydraulic conductivity), the aquifer is more or less vulnerable. The thickness, lithology and the hydraulic conductivity influence the travel time of water particle from surface to the aquifer (saturated zone). As unsaturated zone is greater, the travel time will be higher and vulnerability will decrease. The organic matter and clay minerals content influence the natural attenuation capacity through sorption and degradation processes.

Land use allows the separation of several categories: residential areas, industrial or agricultural areas, transport infrastructure (roads, railways), forestry; each of them could represent a potential source of pollution or could influence the water infiltration rate in the subsurface.

By synthesizing the multiple factors information, four zones of groundwater vulnerability were delineated (Figure 11):

- zone of very low vulnerability (Z1) – cover the slopes of volcanic domes, at the altitudes between 800 and 1000-1100 m, with large slopes (> 40°) and generally wooded lands, all these leading to a very low infiltration rate;

- zone of low vulnerability (Z2) - it is the zone with highest altitudes in the area (1000-1100 m to 1300 m), with lower slopes and less wooded lands; infiltration rate is higher but larger unsaturated area lead to a low vulnerability;

- zone of medium vulnerability (Z3) – developed between 700 and 800 m altitude, with slopes of 20-30°, relatively well wooded lands, except the northern part of the area;

- zone of high vulnerability (Z4) – mostly superposed on the resort area, between Olt valley and 700 m elevation; the lesser protection is due to small slopes which allow a good infiltration together with a reduced thickness of pyroclastic deposits.



Figure 11. Zones of vulnerability to pollution in the hydrographic catchment (left side of the Olt valley) – Băile Tuşnad area.

The actual road (DN12) that crosses the Băile Tuşnad area is located within the high vulnerability zone (Z4) while the bypass road (BR) is placed in the medium vulnerability zone (Z3).

Conclusions. In Băile Tuşnad area the groundwater accumulations are related to flysch formations, almost exclusively on faults and fissures systems and to pyroclastic unconsolidated deposits. Consequently there are springs with different temperatures and chemical pattern: calcium-bicarbonate cold water and chlorosodium thermomineral water. Carbon dioxide plays an important role both for groundwater dynamics and chemistry.

The vulnerability to pollution is controlled by recharge mechanism, natural attenuation capacity of subsurface environment, terrain slope, hydrographic network, land use.

For the natural mineral water occurrences the geomorphological, geological and structural conditions are favourable for a very good natural protection. The recharge areas located at high elevation and the absence of polluting anthropogenic activities represent additional safety elements for chemical and microbiological qualities preservation.

The recharge of the groundwater in Băile Tuşnad area has a surface component from the meteoric waters and a deep (juvenile) component, especially for thermomineral springs.

Even our assessment indicate a medium vulnerability to pollution for the bypass road path, its realization involves risks that can affect the springs exploited downstream in the area. Thus:

- deforestation along the bypass road will modify the infiltration rate (reduce) by increasing the surface runoff;

- the excavations to 5-8 m depth could affect the slope stability;

- potential leakage of contaminants generated by the traffic (dust, oil and gasoline spill, anti-ice or de-icing materials).

The mineral/thermomineral springs from the Băile Tuşnad area belong to a hydrostructure that exist in a fragile equilibrium, both from qualitative and quantitative point of view.

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Iulian Popa, University of Bucharest, Faculty of Geology and Geophysics, Department of Geological Engineering, 6 Traian Vuia Street, 020956 Bucharest, Romania, e-mail: julip_2006@yahoo.co.uk

Mihaela Scrădeanu, University of Bucharest, Faculty of Geology and Geophysics, Department of Geological Engineering, 6 Traian Vuia Street, 020956 Bucharest, Romania, e-mail: mihaelapag@yahoo.com Augustin Jenu, Romanian Association of Hydrogeologists, 6 Traian Vuia Street, 020956 Bucharest, Romania, e-mail: augtenu@yahoo.com

Daniel Scrådeanu, Romanian Association of Hydrogeologists, 6 Traian Vuia Street, 020956 Bucharest, Romania, e-mail: daniel.scradeanu@gg.unibuc.ro

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