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Natural background levels and threshold values for phreatic waters from the Quaternary deposits of the Bahlui River basin

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Abstract

In the Moldavian Plain, known as one of the poorest regions of Romania when it comes to its water supplies, the evaluation of the groundwater (which is used for different purposes) becomes highly necessary. According to the Water Framework Directive 2000/60/EC and Groundwater Directive 2000/118/EC, as part of their effort to achieve the *good* chemical and quantitative status for groundwater, EU member states must assess the latter using threshold values as quality standards for pollutants in groundwater. The present paper presents a case study based on the methodology for the derivation of natural background levels and groundwater threshold values suggested through the EU research project BRIDGE. Given that the groundwater bodies from the study area are exposed to anthropic stress, the 90th percentile is used as natural background level for the phreatic waters from the Quaternary deposits of the Bahlui River. The threshold values were established using drinking water standards as reference It was, thus, discovered, that, when the 90th percentile is used as natural background level, the threshold values obtained in the present study for the parameters NH_4^+, Na^+, Mg^{2+}, Fe and SO_4^{2-} are higher than the reference values.

Keywords: phreatic water quality, natural background levels, threshold values, chemical status.

Introduction

By joining the European Union, Romania adopted a set of principles regarding environmental protection, and the monitoring and interpretation of groundwater quality under conditions of anthropogenic stress, in particular. The new regulations regarding groundwater were set forth through the Water Framework Directive 60/2000/EC.

Thus, Article 17 contains development strategies aimed at both preventing and con-

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trolling the pollution of groundwater, through the implementation of benchmarks meant to ensure the reaching of the *good* chemical status, in agreement with Article 4 (1) (b). The assessment of the quality of groundwater bodies is carried out by comparing the chemical parameters obtained with groundwater quality standards or the threshold values established for pollutants, groups of pollutants or indicators. A groundwater body achieves the *good* chemical status when indicators of pollution concentrations do not exceed the quality standards stipulated by law, or when the effects of saline seepage do not diminish its ecological or chemical quality. A water body is regarded as being at risk when at least 20% of the monitoring points indicate that it is polluted.

In order for EU member states to fulfill the provisions of the Water Framework Directive 2000/60/EC, and reach the common objective of the *good* (qualitative and quantitative) status for their groundwater bodies by 2015, a methodology for the determination of threshold values for substances present in water was imposed through the BRIDGE (Background Criteria for the Identification of Groundwater Thresholds / WFD-BRIDGE.net) project.

The "natural background level" was defined as *the concentration of a substance or the value of an indicator present in solution of groundwater which is derived by not significant anthropogenic influenced processes from geological, biological or atmospheric sources* (Directive 2006/118/EC; Blum et al., 2006; Wendland et al., 2008).

Both the concentration and the content of a groundwater solution are dependent upon a variety of natural factors (the mineralogy of the rocks, the arrangement of the strata, the chemical interaction of the infiltrated water with soils and rocks, the biological processes taking place in the soil, the residence time) and anthropogenic factors (water regulation, industrial pollution, diffuse intakes from agriculture).

The result lies in a variety of methodologies for the assessment of natural background levels of compounds present in groundwater bodies. For example, natural background levels can be estimated by comparing the current chemical status of the groundwater with drinking water standards (Swedish EPA, 2000), or by using the statistical approach for the pre-selection of data (Blum et al., 2006; Wendland et al., 2008). When it comes to providing data-driven estimates of natural background levels for groundwater, Moriani et al. (2012) compare the pre-selection method with the component separation method.

In the present study, the background levels and threshold values have been estimated using techniques described in the BRIDGE methodology (Müler et al., 2006; Hart et al., 2006). Given that the hydrogeochemical characterization of the water bodies corresponds to a *mediocre* level, and that this particular method is preferable in such a case (Hart et al., 2006; Molinari et al., 2012), we have applied the pre-selection of data so as to provide estimates for the natural background levels and threshold values of the chemical compounds present in the groundwater from the Quaternary deposits of the Bahlui River basin.

Natural framework

The Bahlui River basin is a medium-sized catchment area (1950 km^2) located in the central part of the Moldavian Plateau. The Bahlui is the right tributary of the Jijia, and several smaller rivers, such as Bahlueţ and Nicolina, join it in draining this hydrographical basin, which is one of the areas in Romania where the anthropic influence is most visible, stretching (from a hydrotechnical point of view) across 71% of its surface (Savin, 1998).

The Moldavian Plateau is known as one of the poorest regions of the country when it comes to its water supplies, either surface or underground, and this has strong implications regarding the economic and social development of the area.

The lithological characteristics of the central part of the Moldavian Platform allowed the accumulation of groundwater at different depths. The phreatic water bodies are not under pressure and may connect to surface water bodies or infiltration water from precipitation. For this reason, these groundwater bodies display vulnerability to human activities (agriculture, industry or waste water). In the Bahlui River basin, phreatic waters are found in several types of hydro-geological structures, which were separated by Dragomir (1998) as follows:

a). the geological hydro-structures of the floodplain deposits (alluvial deposits);

b). the geological hydro-structure of the terraces deposits;

c). the geological hydro-structure of the interfluve deposits;

d). the geological hydro-structure of the carbonate deposits.

The present paper focuses on the aquifers from alluvial and terraces deposits, which are of Quaternary age (Fig. 1). The shallow groundwater is replenished through rainfall, snowmelt and the surface water bodies.

The alluvial deposits from the floodplain are Holocene in age, and they are the result of transportation and accumulation by the Bahlui River. These deposits occur a few meters above the riverbed and are composed of sands and loessy silts (Brânzilă, 1999). The phreatic aquifer is located in gravels and sands that contain fossil fragments and, rarely, boulders. The aquifer is covered by an alternation of clays and clayey silts, sometimes with lenticular intercalations of sands or rare calcareous concretions, especially upstream (Panaitescu, 2008). This assemblage is waterproof. The confined bed of the aquifer is composed of clays, marls and marly clays of Sarmatian age.

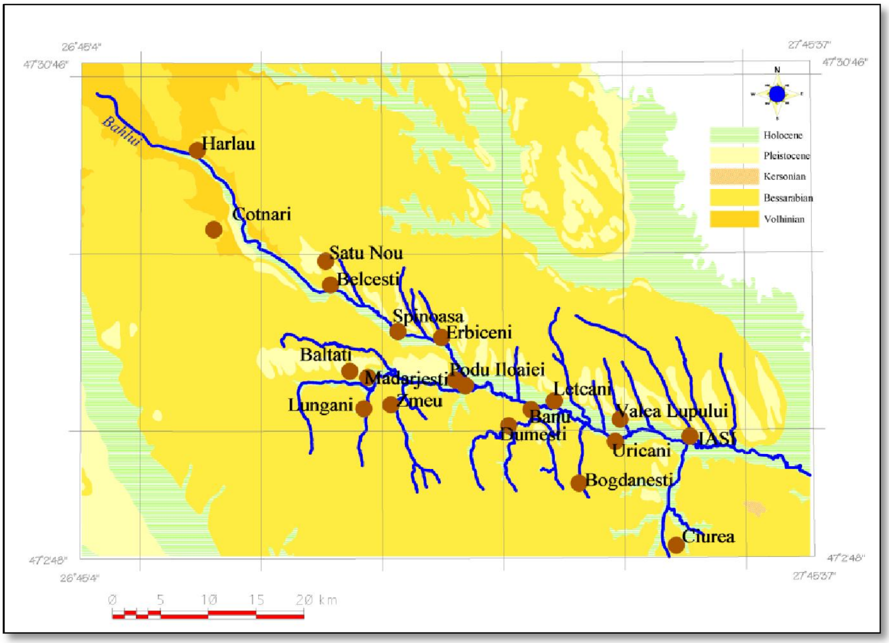

Fig. 1 Groundwater monitoring locations within the Quaternary deposits of the Bahlui River basin.

On both sides of the Bahlui Valley, there are terrace deposits of Pleistocene age. These terraces have been studied by Obreja (1957, 1961), Martiniuc (1966), Băcăuanu (1968), Băcăuanu et al. (1980), and Brânzilă (1999). Brânzilă (1999) noticed a granulometric difference in the terrace deposits: the infill of the upper terraces consists of gravel, as opposed to that of the lower terraces, which display sand and silt. The phreatic waters have accumulated in layers of gravel and sand. The confining cover of the aquifer is composed of silty clay with calcareous concretions and fine sands. The confined bed of the aquifer displays marly sandstones and marly clays of Bessarabian age.

Materials and methods

1. The data set available

We have based our analysis on a set of time series concentration data recorded between 1980 and 2012, from 18 locations (Fig. 1) – each with at least one monitoring well – included in the network of observation wells managed by the Prut-Bârlad Water Basins Administration. To this data, we added data from the literature (Dragomir, 1998; Catrina, 2008). For each sample, we have determined the chemical parameters (NO_3^- and NO_2^-) and the indicator parameters (Na^+, K^+, NH_4^+, Mg^{2+}, Ca^{2+}, HCO_3^-, SO_4^{2-}, pH, total hardness, and oxidability). Most of the hydrogeological monitoring wells from the Bahlui River basin are part of the Ciurea experimental station (20 wells).

2. The pre-selection method

The pre-selection method for the derivation of natural background levels has been suggested by Wendland et al. (2005) within the framework of the BRIDGE project. This method has been revised by Wendland et al. (2008), Hinsby et al. (2008), and Coetsiers et al. (2009). The main criteria suggested for the pre-selection of data are the following:

a). samples with incorrect ion balance or unknown hydrological characteristics are removed;

b). samples from saline aquifers ($[Na^+] + [Cl^-] > 1$ g/L) or with nitrate concentrations that exceed 10 mg/L are also excluded;

c). for each monitoring point, the time series data is converted to median values.

The remaining data is used for the determination of the natural background levels, which are defined, for all groundwater parameters, as the 90th and 97.7th percentiles of the concentration (depending on the amount of data).

The following are two cases in which the threshold values (TV) are determined upon comparing the estimated natural background levels (NBL) to the reference standards (REF) (Müler et al., 2006; Hinsby et al., 2008):

- NBL / REF < 1 => TV= (NBL + REF) / 2;
- NBL / REF ≥ 1 => TV = NBL.

Usually, drinking water standards are considered appropriate reference values.

Results and discussion

1. Water chemistry

The chemistry of the phreatic groundwa-ter from the study area depends on the litho-logical composition of the alluvial and terrace deposits, the connection with surface water bodies, the rainfall regime and infiltration wa-ters, the water – soil – atmosphere interaction, as well as on anthropogenic factors. Ground-water quality is influenced by reactions such as silicate weathering, redox reactions, calcite, gypsum and chloride dissolution, and cation exchange.

The data was statistically analyzed so that the hydrogeochemical characterization of the waters could be formulated. Among the quality indicators that were taken into account, one could mention the following: major ions (HCO_3^-, SO_4^{2-}, Cl^-, Ca^{2+}, Mg^{2+}, Na^+, K^+), oxygen regime indicators (CCOMn), nutrients (NH_4^+, NO_3^-, NO_2^-), pH, and TDS (total dissolved solids) (Tab. 1). Based on the criteria suggested by Murray et al. (1996), all samples with incorrect ion balance are excluded.

The groundwater from the Quaternary deposits of the Bahlui River basin exhibits higher values for the sodium found in the plains, compared to that found in the terraces. The presence of sodium is due to the fact that, under present-day climatic conditions, in the process of ionic exchange with the water, the siliceous clays from the Sarmatian strata retain the calcium and eliminate the sodium (Schram, 1971). The alkaline pH values associated with high HCO_3^- concentrations could indicate calcite dissolution, but the values of Ca^{2+} need to be higher. On the contrary, in the groundwater bodies studied, the variability range of Ca^{2+} is subordinated to that of Na^+. The lower Ca^{2+} concentration in water can be justified by the occurrence of cation exchange in the aquifer (Coetsiers, 2008). The dominant anions are bicarbonate, followed by sulfate. The source of the sulfate is represented by deliquescent salts (gypsum and/or anhydrite, glauberite, thenardite, mirabilite) from the Quaternary deposits. Some samples, collected from Banu and Uricani, display higher chloride contents, associated with higher Na^+ values. This indicates a pollution source, or the presence of chloride minerals in upper strata or soils.

Table 1 Groundwater composition

Parameter	N	Mean	Std. Dev.	Min.	Max.	P 10	P 90
NH_4^+ (mg/L)	61	1.43	3.51	0.00	26.55	0.09	2.12
Na^+ (mg/L)	61	409.41	549.25	28.00	3620.00	66.66	847.50
K^+ (mg/L)	61	0.54	1.73	0.00	9.38	0.00	2.34
Ca^{2+} (mg/L)	61	109.46	63.41	8.02	359.43	44.00	180.00
Mg^{2+} (mg/L)	61	71.01	40.85	10.00	272.00	36.00	109.00
Fe (mg/L)	61	3.17	22.43	0.00	168.00	0.00	0.70
Cl^- (mg/L)	61	206.01	679.02	10.00	4850.00	16.00	300.00
SO_4^{2-} (mg/L)	61	488.29	438.63	48.98	2470.00	98.00	942.00
HCO_3^-	61	829.99	362.69	317.00	2205.00	515.00	1220.00
NO_3^- (mg/L)	61	6.42	9.07	0.00	45.00	0.00	16.25
NO_2^- (mg/L)	61	0.36	1.42	0.00	10.40	0.00	0.30
CCOMn	61	10.48	8.84	0.00	48.00	5.10	17.90
pH	61	7.63	0.29	7.00	8.50	7.30	8.00
Total hardness (°dH)	61	32.75	15.14	5.60	86.60	20.10	47.00
TDS	61	1748.74	1474.99	610.00	10045.00	713.50	2830.00

The variation in TDS values allowed us to include the shallow groundwater samples in either the fresh (TDS < 1000 mg/L) or the brackish (1000 < TDS < 10000 mg/L) categories. One sample (collected from Banu) is of moderate saltwater, with a TDS value of 10045 mg/L.

A Sulin diagram was used for the determination of the origin of the water in the study area. All the phreatic groundwater is located in the lower quadrant of the diagram (Fig. 2), which indicates that it was, originally, meteoric water of the Na_2SO_4 and $NaHCO_3$ type. The derived sulfates and bicarbonates are, however, associated with continental conditions.

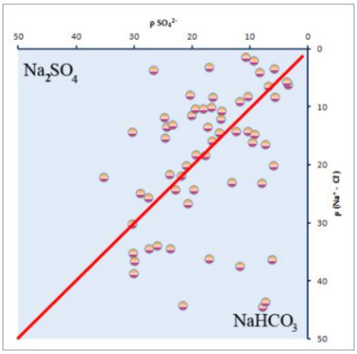

Fig. 2 Sulin graph for the genetic classification of the phreatic groundwater in the study area.

The distribution pattern highlighted by the Piper diagram indicates that, when located in the Na or the mixed fields, the groundwater from the study area varies from bicarbonate to sulfate waters in the anion triangle. The diamond-shaped field indicates that the groundwater is of one of the following types:

a). the Ca^{2+} + Mg^{2+} and HCO_3^- water type – 31.14% of samples;

b). the Na^+ + K^+ and HCO_3^- water type – 11.47% of samples;

c). the Na^+ + K^+ and Cl^- – SO_4^{2-} water type, indicating secondary salinity properties attributed to the dissolution of salts in the water-bearing deposits – 42.62% of samples;

d). the Ca^{2+} + Mg^{2+} and SO_4^{2-} – Cl^- water type – 14.75% of samples.

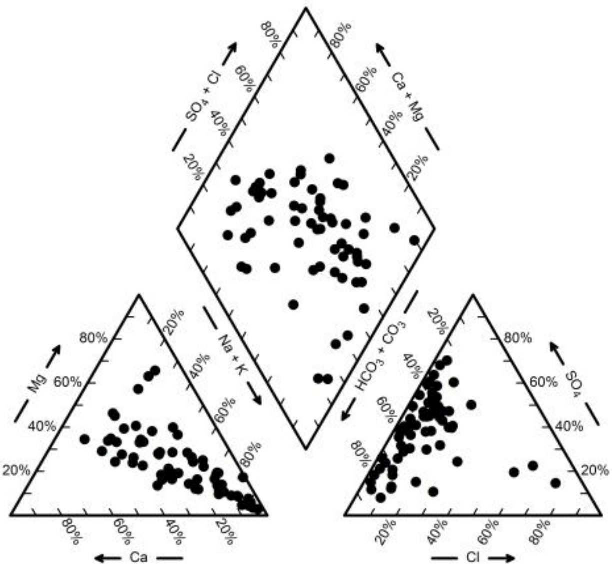

Fig. 3 Hydrogeochemical classification of groundwater samples collected from the Quaternary deposits of the Bahlui River basin, achieved using the Piper diagram.

Table 2 Correlation matrix of the major elements in the groundwater

	Na	Ca	Mg	Cl	SO_4	HCO_3	pH	TDS
Na	1.000							
Ca	-0.098	1.000						
Mg	-0.015	0.300	1.000					
Cl	**0.857**	-0.023	0.002	1.000				
SO_4	**0.446**	0.272	**0.404**	0.073	1.000			
HCO_3	**0.571**	-0.130	-0.113	0.190	0.396	1.000		
pH	0.272	-0.250	0.018	0.130	0.128	0.378	1.000	
TDS	**0.974**	0.051	0.158	**0.838**	**0.560**	**0.520**	0.239	1.000

The correlation coefficients among major ions and pH and TDS parameters were calculated for correlation analysis. According to Table 2, the Na^+ – Cl^-, Cl^- – TDS and Na^+ – TDS pairs show a strong positive correlation, which confirms the common source of these cations (either the dissolution of chloride salts, or pollution), and their major participation to the mineralization of the waters. The good positive correlation between Na^+ and both SO_4^{2-} and HCO_3^- indicates a recharging of the Quaternary aquifer with meteoric water. The SO_4^{2-} – TDS and HCO_3^- – TDS pairs, respectively, exhibit good positive correlation, indicating that, along with the Cl^- – TDS pair, they reflect the mixing of waters with different origins. The weak positive correlation between HCO_3^- and SO_4^{2-} points to the role of deeply percolated meteoric water in the composition of the groundwater.

2. Natural background level and threshold values

In order to assess the natural background levels for the groundwater from the Quaternary deposits of the Bahlui River basin, the pre-selection method described above was applied to the database.

Table 3 Number of sampling points eliminated from the estimation of natural background levels

Locality	No. of sampling points	No. of sampling points eliminated	
		Elimination criterion Na^+ + Cl^- > 1000 mg/l	Elimination criterion NO_3^- > 10 mg/l
Hârlău	1		
Cotnari	3	1	
Satu Nou	1		
Belcești	6	2	1
Spinoasa	2		1
Erbiceni	1		
Podu Iloaiei	1		1
Lețcani	1		
Banu	4	2	
Valea Lupului	1		1
Uricani	2	2	
Iași	10	1	
Aroneanu	1		1
Bălțați	1		
Mădârjești	2	1	1
Lungani	1		1
Zmeu	1		1
Dumești	1		1
Bogdănești	1		
Ciurea	20		1

The criteria suggested for the samples affected by human influence (chloride and nitrate concentration) have led to the exclusion of 32 % of the initial samples (Tab. 3). Since the unconfined and relatively shallow nature of the aquifers in question makes them highly vulnerable to pollution, the 90^{th} percentile is suggested for natural background level values.

Table 4 shows a comparison between the results of the determination of natural background levels for the groundwater from the alluvial and terrace deposits of the Bahlui River basin, derived at the 10^{th}, 50^{th}, 90^{th} and 97.7^{th} percentiles, respectively, for the same parameters.

Given that, in Romania, the legal quality standards for groundwater are, largely, equal to those stipulated in the European Drinking Water Directive (98/83/EC), the use of the drinking water standards as reference for the estimation of threshold values (table 5) is appropriate, especially when the aquifers studied are being used as drinking water supplies.

In the case of the 90^{th} percentile used as natural background level, the threshold values obtained are higher than the drinking water standards for the parameters NH_4^+, Na^+, Mg^{2+}, Fe and SO_4^{2-}. The high concentrations of these parameters could indicate natural origins.

Table 4 Estimation of natural background levels for the groundwater bodies from the study area

Parameter	Estimation of natural background levels			
	P 90	P 97.7	P 10	P 50
NH_4^+ (mg/L)	**2.13**	5.44	0.15	0.65
Na^+ (mg/L)	**560.00**	852.60	84.26	237.75
K^+ (mg/L)	**2.46**	6.99	0.00	0.00
Ca^{2+} (mg/L)	**180.00**	206.80	59.06	112.00
Mg^{2+} (mg/L)	**95.00**	120.57	37.20	59.00
Fe (mg/L)	**0.72**	33.57	0.00	0.15
Cl^- (mg/L)	**157.00**	200.62	17.60	48.10
SO_4^{2-} (mg/L)	**778.00**	942.00	117.50	360.50
HCO_3^-	**1010.00**	1931.42	544.20	715.50
NO_3^- (mg/L)	**4.00**	7.24	0.00	2.00
NO_2^- (mg/L)	**0.20**	0.32	0.02	0.08
pH	**8.00**	8.14	7.30	7.50
Total Hardness (°dH)	**44.80**	50.52	20.54	30.90
TDS (mg/L)	**2040.00**	2332.36	768.00	1290.00

Table 5 Derivation of threshold values for the groundwater bodies from the study area

Parameter	Reference values	Threshold value (TV)	
	Directive 98/83/EC	TV estimation	Estimation criterion
NH_4^+ (mg/L)	0.5	**2.13**	NBL / REF ≥ 1; TV = NBL
Na^+ (mg/L)	200	**560.00**	NBL / REF ≥ 1; TV = NBL
K^+ (mg/L)	10	**6.23**	NBL / REF < 1; TV = $\frac{(NBL+REF)}{2}$
Mg^{2+} (mg/L)	50	**95**	NBL / REF ≥ 1; TV = NBL
Fe (mg/L)	0.200	**0.72**	NBL / REF ≥ 1; TV = NBL
Cl^- (mg/L)	250	**203.5**	NBL / REF < 1; TV = $\frac{(NBL+REF)}{2}$
SO_4^{2-} (mg/L)	250	**778.00**	NBL / REF ≥ 1; TV = NBL
NO_3^- (mg/L)	50	**27**	NBL / REF < 1; TV = $\frac{(NBL+REF)}{2}$
NO_2^- (mg/L)	0.5	**0.35**	NBL / REF < 1; TV = $\frac{(NBL+REF)}{2}$

Conclusions

The methodology suggested through the BRIDGE European project for the assessment of natural background level and threshold values, and the definition of the chemical status of groundwater, has been tested on the groundwater bodies from the alluvial and terrace aquifers of the Bahlui River basin. Due to the nature of these Quaternary groundwater bodies, they are very vulnerable to pollution. Consequently, the natural background levels were determined using the 90th percentile on a pre-selected set of data. The methodology used in the estimation of threshold values is strongly dependent upon the choice of reference values. In the present paper, Drinking Water Standards are used as reference values. For the groundwater bodies on which the present study focuses, the methodology applied leads to very high threshold values for NH_4^+, Na^+, Fe, Mg^{2+} and SO_4^{2-}.

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