

**HYDROGEOCHEMICAL ASPECTS LINKED TO THE SHALLOW  
GROUNDWATER QUALITY AT CRISTEȘTI (IAȘI COUNTY)**

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**Abstract**

The hydrogeochemistry of the shallow groundwaters from Cristești has been described by means of hydrogeochemical diagrams (Piper, Durov) and through the analysis of the hydrogeochemical changes that occur from the terraces area, at the foot of which the first drilling was carried out towards the aquifers conduit discharge area (Prut river). Thus, the  $\text{HCO}_3^-$  -  $\text{Na}^+$  waters, are projected within the CAD range (calcite – anhydrite – dolomite), whereas the  $\text{SO}_4^{2-}$  -  $\text{Na}^+$  waters ”migrate” towards gypsum/anhydrite and dolomite, as the calculated saturation indexes prove. The indicator that presents exceeding values of MAC, in all the drillings, is CCOMn.

**Key words:** shallow groundwater, water rock interaction, hydrogeochemistry, nitrates

**Introduction**

The drillings at the Cristești hydrological station investigate, from a qualitative and quantitative viewpoint, the phreatic aquifer quartered in the common field Jijia - Prut.

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Knowledge about this aquifer and its hydrogeological and hydrochemical potential is important in the present-day context of encouragement of investments regarding water supply in rural regions from this area (Cristești, Ungheni, Bosia – Iași county). The Cristești section also monitors the shallow groundwater resource from the alluvium plain at the Eastern extremity of the European Union.

The area in which the drillings were carried out spans quite a large strip of land in the united alluvial plains of the rivers Jijia and Prut. The drillings were performed on both banks of the river Jijia, following the direction WSW – ENE, between the localities Cristești and Ungheni.

The objectives of this work are the study of the evolution of main ions and the assessment of the quality of the waters, as a result of the implementation of EU Directives. The main polluting agents from the researched area that lead to the degradation of the water quality and make it impossible for it to be used as drinking water are fertilisers used in agriculture, improperly sealed septic tanks and the waste materials resulted from the zootechnical centres. Groundwater contamination has major implications on health and environment (Navaro, 2007). In order for water to be drinkable, it has to comply with a series of norms imposed by the World Health Organisation (WHO) and the European Union (EU).

### **Studied area**

From a geological viewpoint, the studied area belongs to the Moldavian Platform, that represents, in fact, a socle from Precambrian, made up from metamorphic rocks and a sedimentary cover (with Ordovician, Silurian, Cretacic, Neogen and Quaternary deposits). The studied perimeter is characterised by the appearance of Sarmatian and Quaternary deposits (fig. 1).

Basarabian is characterised by the presence of compact calcareous clays and of grey clays with sand lenses. On top of the basarabian deposits there are alluvial deposits from the river Prut alluvial plain, made up of coarse sands, gravel, clays, clayey-sandy silts and fine sands. Alluvial deposits are arranged vertically as follows:

- at the basis, a level of coarse sands with gravel, with a thickness between 5 and 15 metres, belonging to the lower Holocene.
- fine granulation alluvium deposits, made up of clayey-sandy silts, clays, fine powdery sands that belong to the late Holocene and are arranged in parallel, lense-like layers over the lower Holocene, with widths ranging from 3 to 10 metres.

On the terrace, there are loess deposits, sands and sands with rare gravels of Quaternary age.

Mineralogically, Quaternary deposits are mainly made up of clayey minerals (illites and smectites). Subordinatingly, there are soluble salts, plagioclase and potassium feldspars, micas (muscovite) and quartz.

It can be noticed that, in general, the thickness of the coarse sands with gravel deposits is greater at the centre of the alluvial plain, getting smaller as it advances towards the area of contact with the hilly region at the western border. A common feature of the superficial structure from the river Prut alluvial plain is the lack of uniformity and the great vertical variation of the alluvial deposits, both as facies and as width.

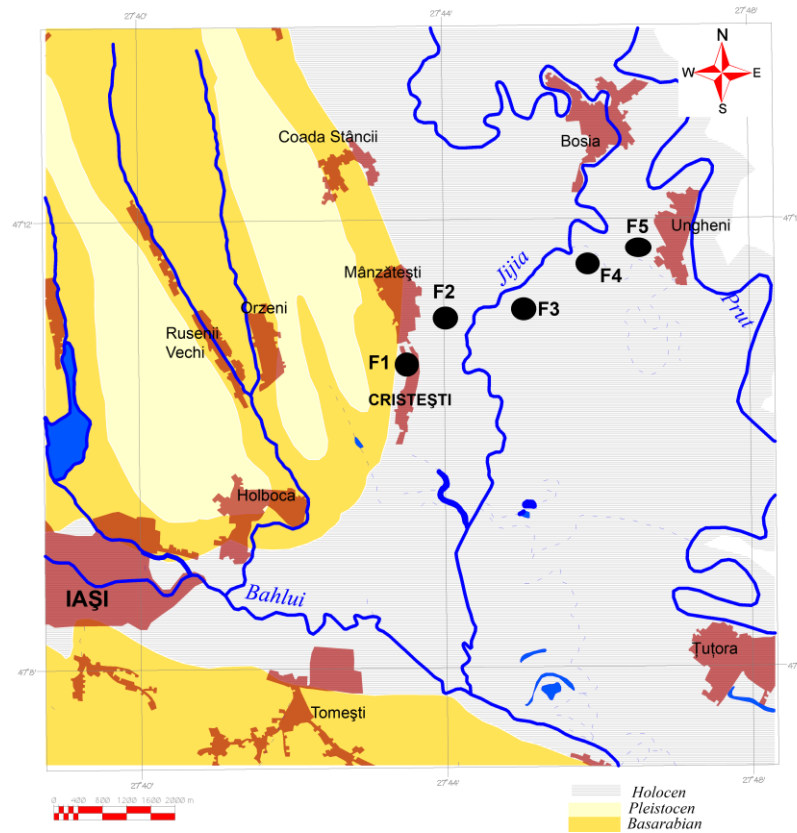


Fig. 1 The Geology of the studied area

From a geomorphological viewpoint, the studied area belongs to the hilly Jijia Plain, spanning to the east of the Moldavian Plateau and lies between the front of the terraces to the west and the minor riverbed of the Prut river to the east. The altitude of the common alluvial plain of the rivers Jijia and Prut is between 30 to 40 metres, slightly sloping from

north to south and presenting more prominent points where there are strips of land. Because the landscape slope is very small and the alluvial plain only slightly slanting, the flows of the rivers Prutului and Jijia are strongly meandering.

From climatic viewpoint, the hilly Jijia Plain is under the influence of the temperate continental climate. According to Ungureanu (1993), the average rainfall is rather poor (450 – 550 mm yearly), especially compared to the high temperatures in summer.

Hydrogeology of the Area. The phreatic aquifer is quartered in the horizon of sands and gravels, its hydrostatic level having a slightly ascending character in those parts of the alluvial plain where clayey and powdery strata from the upper lithological complex are better developed.

The upper layer of the aquiferous stratum is made up of clays, a fact demonstrated by the presence of numerous areas of stagnant water from the alluvial plain.

The shallow groundwater quartered in the alluvial plain deposits are dynamically linked to the water level in the rivers. The flow and quality of the alluvial plain shallow groundwater are dependent on the granulometric composition of the alluvial deposits. With the increase of the permeability of rocks that make up alluvial plains, the water exchange between them and rivers decreases and sometimes the mineralization of the shallow groundwater increases, such as the case is with phreatic waters from the Prut – Jijia alluvial plain. The flowing direction of the underground waters is N – S and NW – SE, approximately parallel with the flowing direction of the river Prut. The levels of the underground water vary naturally, depending on the water supply sources and the aquiferous layer: the inflow from the terrace, atmospheric rainfall, ground water of the rivers Jijia and Prut which are connected to the aquiferous layer.

## Materials and Methods

For the present study, we have used, elaborated and interpreted data gathered from the water samples collected from 1969 until 2007, from the drillings of the state-owned hydrological network, which is administered by the Prut Waters Directorate (Direcția Apelor Prut - DAP), Iași and the meteorological data supplied by the Moldova Regional Meteorological Center. Work on the drillings, water samples collection and the physico – chemical analyses were done by the Prut Waters Directorate. The data was statistically analysed so as to get the hydrogeochemical characterisation of the waters, but also in order to be able to compare it against the norms and standards framework in force. In order to get the hydrogeochemical analyses, the findings were validated, by checking the ionic balance of the admitted errors. Among the quality indicators that were taken into account, mention can be made of: major ions ( $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ), oxygen regime indicators (CCOMn) and nutrients ( $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ ).

## Results and Discussion

### *Water Chemistry*

The drillings from the Cristești section belong to the ROPR02 body of water "the alluvial plain and the terraces of the lower and middle Prut river and of its tributaries", which was separated into alluvial areas and terraces of the rivers Prut, Bahlui and Jijia. The identified underground body of water belongs to the porous type, accumulated into Quaternary and Sarmatian deposits and, being situated close to the surface, has a free level. Initially, the Cristești section encompassed five drillings, out of which F1 and F5 are still functional at present. The monitoring interval and the depth of the drillwork for each particular drilling are presented in table 1.

Tab. 1 Drillings of the Cristești section

Drilling number	Monitoring period (years)	Depth (metres)
F1	1969 – 2007	25
F2	1969 – 1982	18
F3	1969 – 1981	17
F4	1969 – 1995	17
F5	1969 – 2007	26

From a lithological viewpoint, aquifers present the following characteristics:

F1 – the aquiferous layer is made up, on the upper part, of sand, rarely gravel and pebbles on the lower part and was tapped into between 10 to 19 metres.

F2 – the aquiferous layer is made up of sand on the upper part and sand, gravel and pebbles on the lower part. The aquifer was tapped into between 9 to 12 metres.

F3 – the aquiferous layer was tapped into between 11.9 to 13.4 metres, being made up of sand and gravel.

F4 – the aquiferous layer was tapped into between 12.5 to 14.2 metres and is made up of sand and gravel.

F5 – the aquiferous layer was tapped into between 11.5 to 14.7 metres depth from the groundlevel and is made up of sand and rarely gravel.

The studied waters present pH values that range from 7.0 to 8.5, whereas TDS (total solids dissolved) varies greatly from 120 to 6460 mg/L. From the point of view of major ions, the studied waters present a remarkable heterogeneity. This fact is thoroughly demonstrated through the analyses of ternary diagrams (fig. 2), which show compositions that vary from sodium bicarbonate to sodium sulphate. This fact clearly shows the different geochemical processes that take place at the interface of the water with the rock, as well as the short residence times of these waters.

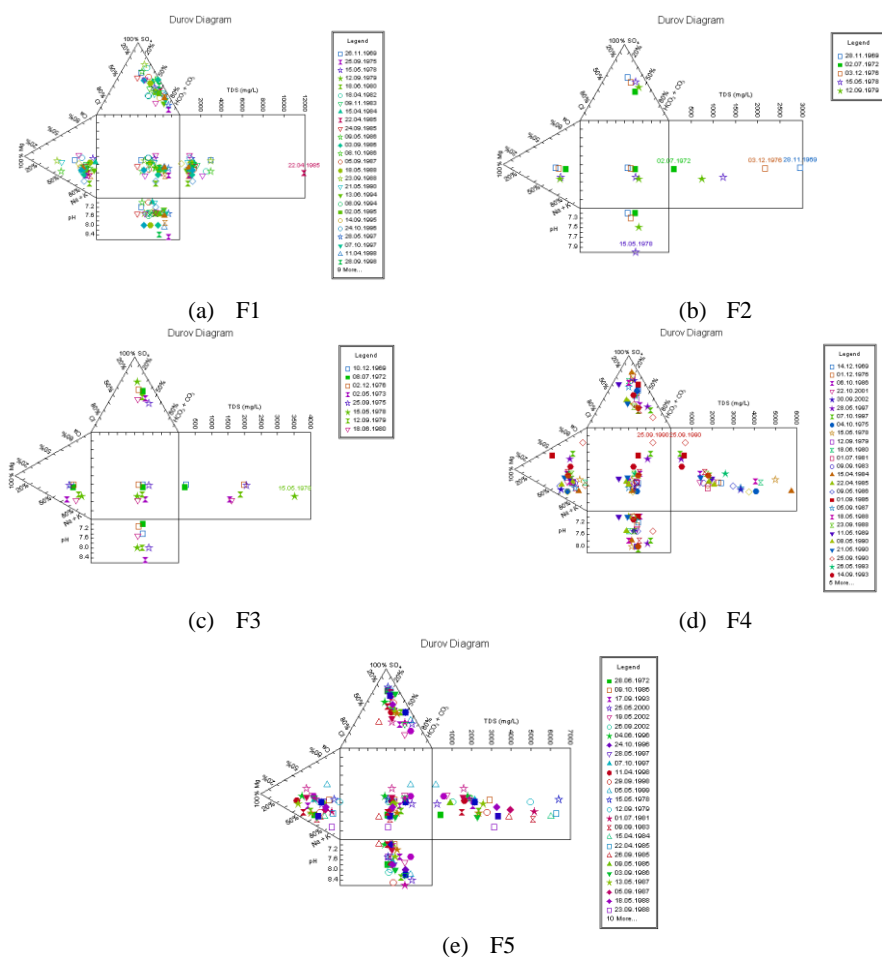


Fig. 2 Durov's diagram showing the chemical composition of the shallow groundwater in the Cristești section

Most of the water samples belong to the type  $\text{Na}^+ - \text{SO}_4^{2-} - \text{HCO}_3^+$  (tab. 2), in conformity to the lithology of the area. The dominant anions are bicarbonate (for F1) and sulphate (for the other drillings), and the most well-represented cation is  $\text{Na}^+$ . The presence of sodium is due to the fact that, under the present-day climate conditions, siliceous clays from the Sarmatian strata, in the process of the ion changes with the water, retain the calcium and oust the sodium (Schram, 1971).

The high content of  $\text{HCO}_3^-$  can be explained by the dilution of  $\text{CaCO}_3$  by waters high on  $\text{CO}_2$ , but, from the matrix of the correlation coefficients, one can notice that there is no correlation between  $\text{Ca}^{2+}$  and  $\text{HCO}_3^-$ . According to Popa et al. (1993) waters with a high content of  $\text{HCO}_3^-$  have two possible important sources of  $\text{CO}_2$ : atmospheric  $\text{CO}_2$  and  $\text{CO}_2$  from soils, which can reach up to 1.6 % in areas with vegetation.

Tab. 2 Hydrogeochemical facies of shallow groundwater

Well no.	Hydrogeochemical facies
F1	$\text{HCO}_3^- - \text{SO}_4^{2-} - \text{Na}^+ - \text{Ca}^{2+}$
F2	$\text{SO}_4^{2-} - \text{HCO}_3^- - \text{Na}^+ - \text{Ca}^{2+}$
F3	$\text{SO}_4^{2-} - \text{HCO}_3^- - \text{Na}^+ - \text{Ca}^{2+}$
F4	$\text{SO}_4^{2-} - \text{HCO}_3^- - \text{Na}^+ - \text{Ca}^{2+}$
F5	$\text{SO}_4^{2-} - \text{HCO}_3^- - \text{Na}^+ - \text{Mg}^{2+}$

Tab. 3 Matrix of the correlation coefficients for major ions from the sampled waters

	$\text{Na}^+$	$\text{K}^+$	$\text{Ca}^{2+}$	$\text{Mg}^{2+}$
F1				
$\text{Cl}^-$	0.533	-0.271	0.564	0.544
$\text{SO}_4^{2-}$	0.791	-0.400	0.539	0.824
$\text{HCO}_3^-$	0.285	0.192	0.069	0.309
F4				
$\text{Cl}^-$	0.575	0.096	0.500	0.378
$\text{SO}_4^{2-}$	0.958	-0.271	0.648	0.405
$\text{HCO}_3^-$	0.561	0.237	0.330	0.550
F5				
$\text{Cl}^-$	0.565	-0.187	0.286	0.465
$\text{SO}_4^{2-}$	0.948	-0.352	0.701	0.227
$\text{HCO}_3^-$	0.549	0.046	0.470	0.420

The correlation coefficients (tab. 3) determined for the major ions present in water are also in conformity with the Durov diagrams presented above. One can notice very good correlations, for all waters, between  $\text{SO}_4^{2-}$  and  $\text{Na}^+$ , as well as between  $\text{SO}_4^{2-}$  and  $\text{Mg}^{2+}$ , at drilling F1. The source of the sulphate is represented by deliquescent salts (gypsum and/or anhydrite, glauberite, thenardite, mirabilite) from the Quaternary deposits.

The water type is directly influenced by the climatic factor, more precisely by the rainfall. One can notice that, during wet periods, the character of the water tends to be of sulphate type, whereas, during dry periods,  $\text{HCO}_3^-$  is dominant. In order to exemplify

this, we have considered the following situations: variation of chemical composition of the water sampled in September 1975 and 1988, out of the drilling F1.

a. In 1975, the average rainfall was 572.5 mm/year, and the water had the geochemical features of a  $\text{Na}^+ - \text{HCO}_3^- - \text{SO}_4^{2-}$  type;

b. In 1988, the average rainfall was 635.8 mm/year, and the water had the geochemical features of a  $\text{Na}^+ - \text{SO}_4^{2-} - \text{HCO}_3^-$  type.

The same situation is relevant for other drillings, for instance, at drilling F2, in 1984 (with a rainfall of 672.1 mm/year), the water belonged to  $\text{Na}^+ - \text{SO}_4^{2-} - \text{HCO}_3^-$  type, whereas in 1990 (when there were rainfalls of 397.1 mm/year), the water had the geochemical features of  $\text{Na}^+ - \text{HCO}_3^- - \text{SO}_4^{2-}$  type.

Average pH values and standard pH deviations, amounts of major ions and the saturation index in calcite, aragonite, dolomite and gypsum for the studied shallow groundwater are presented in table 4, where the similarities and differences among these points of study are indicated.

Tab. 4 Statistical parameters for the major ions, the pollution and the saturation indices

	F1		F2		F3		F4		F5	
	Mean	Std. Dev.	Mean	Std. Dev.	Mean	Std. Dev.	Mean	Std. Dev.	Mean	Std. Dev.
pH	7.54	0.36	7.44	0.34	7.69	0.52	7.45	0.38	7.63	0.49
$\text{Na}^+$	283.80	103.89	290.60	104.18	458.63	171.94	471.42	327.12	565.32	412.82
$\text{K}^+$	4.91	6.93	17.00	41.64	0.00	0.00	6.00	13.01	14.11	25.20
$\text{Ca}^{2+}$	122.06	52.09	167.60	98.53	146.25	68.55	186.19	98.02	196.57	161.83
$\text{Mg}^{2+}$	58.22	71.58	91.30	60.35	88.53	23.71	91.84	57.85	132.69	75.71
$\text{HCO}_3^-$	701.06	128.61	628.80	197.17	710.88	146.29	754.71	277.05	801.89	335.28
$\text{SO}_4^{2-}$	441.70	388.00	642.40	387.50	848.25	405.70	862.81	739.17	1190.92	970.20
Cl	97.59	67.37	160.00	98.89	188.00	37.76	242.16	118.69	217.62	157.10
TDS	1360.71	660.99	1454.00	1013.15	1645.50	1001.66	2265.33	1320.28	3628.44	1659.75
$\text{NH}_4^+$	3.56	17.15	1.65	1.24	0.60	0.75	1.57	2.46	0.77	0.92
$\text{NO}_3^-$	7.70	10.14	6.00	11.22	3.64	5.34	14.51	21.61	24.76	32.06
$\text{NO}_2^-$	0.18	0.34	0.01	0.02	0.00	0.00	0.23	0.30	54.23	325.41
CCOMn	14.45	31.78	8.76	4.96	27.10	48.83	18.34	22.72	10.79	10.80
$\text{P}_{\text{CO}_2}$ (bar)	$10^{-1.76}$	0.34	$10^{-1.73}$	0.37	$10^{-1.92}$	0.46	$10^{-1.71}$	0.34	$10^{-1.84}$	0.54
SI cal	1.14	0.37	1.08	0.32	1.34	0.45	1.23	0.59	1.40	0.63
SI dol	2.16	0.82	2.26	0.64	2.87	0.92	2.42	1.21	2.97	1.17
SI gyp	-0.44	0.46	-0.11	0.50	-0.01	0.41	-0.01	0.52	0.09	0.69
SI arag	1.00	0.37	0.94	0.32	1.20	0.45	1.09	0.59	1.26	0.63

In the first drilling, the dominant anion is the bicarbonate, whereas in the other drillings the sulphate anion dominates quantitatively; its value increases towards east. The pH of the waters from the five drillings has an average value that ranges between 7.4 and 7.7. The concentration of  $\text{HCO}_3^-$  is higher in the shallow groundwater from F1, a fact which indicates that more calcite or dolomite was diluted in the water from this aquifer



than in the others. The average value of the mol ratio  $\text{Ca}^{2+}/\text{Mg}^{2+}$  is larger than the unity. There can be noticed an increase of the total mineralisation as one gets closer to the aquifer conduit discharge area. Compared to the TDS value, shallow groundwater from the researched area are brackish. The quite high mineralisation of shallow groundwater from this area is determined by the chemistry of the rocks, by the small discharge slope and by the existence of the waterproof superior horizon.

The calculation of the partial pressure of  $\text{CO}_2$  ( $P_{\text{CO}_2}$ ) using the AqQA 1.1 programme got higher values than the atmospheric pressure of  $10^{-3.5}$  bar, which indicates that the shallow groundwater from the Cristești section get loaded with  $\text{CO}_2$  as it infiltrates in soil.

Likewise, following Fitts's model (2002), we calculated the saturation indexes for calcite, aragonite, dolomite and gypsum and concluded that the waters are supersaturated in carbonates. As far as sulphate is concerned, it is noticed that, along the direction WSW – ENE the saturation index increases. Drillings F1 and F2 are unsaturated, drillings F3 and F4 are approximately saturated, and the drilling near the Prut river, F5, is supersaturated in gypsum.

Starting from the chemical principle of the differentiated degree of attraction between ions in a solution (the Paneth – Fajans Hahn rule) and in conformity with Scholler's ion exchange series (Dragomir, 1998) we have established the amount of dissolved salts (fig. 3). One notices that, as one gets closer to the river Prut, the sulphates concentration increases from 37 % (in F1) to 52% (in F5) and the chlorides concentration from 3% to 11 %, whereas bicarbonates decrease.

A general framework of the processes that affect the studied underground waters can be described through the diagram  $\text{Ca}^{2+}/(\text{Ca}^{2+} + \text{Mg}^{2+})$  vs  $\text{SO}_4^{2-}/(\text{SO}_4^{2-} + \text{HCO}_3^-)$  (fig. 4), which shows that, in the case of bicarbonated waters from F1, most of the water samples plot within the CAD range (calcite – anhydrite – dolomite) and at the other drillings (where sulphate is the dominant anion) one notices a transition towards the gypsum or anhydrite range. The composition of waters plotted in the CAD range is controlled by the dissolution of calcite, dolomite and gypsum, whereas the percolation of gypsum is widespread in the other waters. This corresponds to the variation of the saturation indexes mentioned above. From the research carried out by Schiavo (2006), reduced values of  $\text{SO}_4^{2-}/(\text{SO}_4^{2-} + \text{HCO}_3^-)$  ratio suggest that the interaction of the water with the sulphates is negligible.

#### *Quality of the Shallow Groundwater*

In order to be able to use the shallow groundwater from the Cristești section as drinking water, we have followed the norms imposed by the Romanian legislation, which was made in agreement with the European legislation, more precisely the Law 311/2004, in agreement with Directive 98/83/EC.

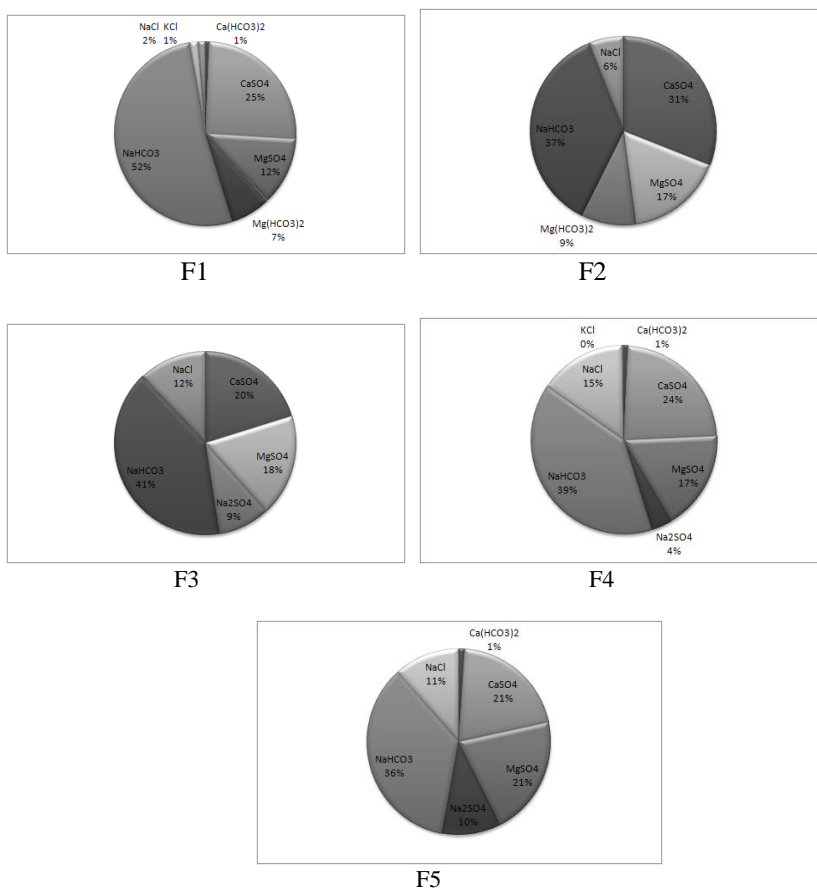


Fig. 3 Possible dissolved salts in the shallow groundwater

From the evolution of the indicators that we have studied, we can state that:

1. There are no exceeding pH values, the pH ranging within the imposed limits of 6.5 to 9.5 units.
2. In the major ions group, one can notice amounts a couple of times higher (even 5 times) for the sulphate anion, compared to the maximum allowed concentration (MAC).
3. In the nutrients group, out of the nitrogen forms present in the water ( $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ ) one can identify frequent overruns of MAC of 0.5 mg/L for  $\text{NH}_4^+$  anion.

$\text{NO}_3^-$  and  $\text{NO}_2^-$  are generally within the allowed range, but there have also been noticed occasional exceeding values of the MAC.

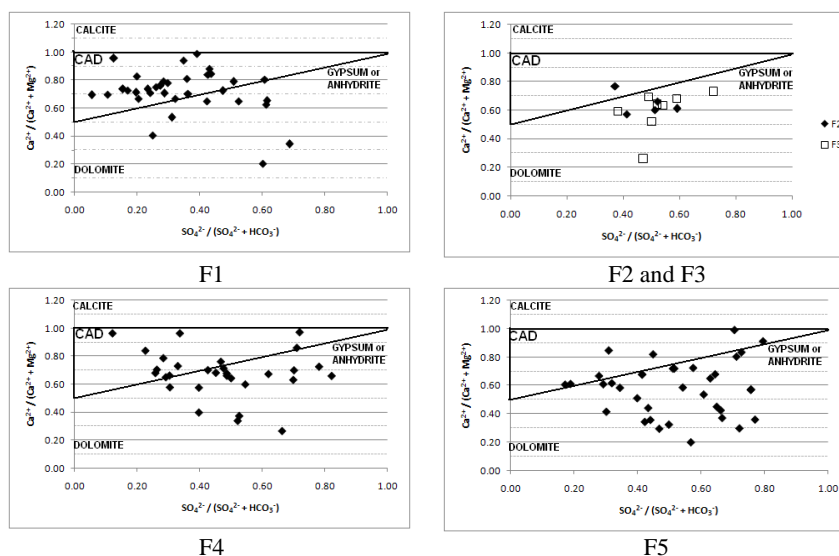


Fig. 4 Diagram of  $\text{Ca}^{2+}/(\text{Ca}^{2+} + \text{Mg}^{2+})$  vs.  $\text{SO}_4^{2-}/(\text{SO}_4^{2-} + \text{HCO}_3^-)$  concentrations in sampled water

4. Out of the indicators for the oxygen regime, we have studied the CCOMn, which presents extremely high values in all the collected samples, registering overruns of MAC, which ranges averagely between 3.50 and 10.83, no matter the time of the year when the determination was carried out.

Although the area is favourable for agriculture, the presence of the nitrate cannot be due solely to fertilisers used in order to increase productivity. A possible source is represented by the septic tanks that are not properly sealed or the oxidation of the ion  $\text{NH}_4^+$  (nitrification). This latter process normally takes place on the soil surface, where organic matter and oxygen can be found in abundance (Drever, 1979).

Once the  $\text{NO}_3^-$  gets to the unsaturated aerobic area, it will behave as a conservative anion, its geochemistry will resemble that of  $\text{Cl}^-$ , and its evolution can be followed using the ratio  $\text{NO}_3^-/\text{Cl}^-$  or the diagram  $\text{NO}_3^-$  versus  $\text{Cl}^-$  (Schiavo et al., 2006; Majumder et al., 2008). Majumder et al. (2008) states that, if shallow groundwater have a high concentration of  $\text{Cl}^-$  and a lower concentration of nitrate, the source of  $\text{Cl}^-$  is not represented by fertilisers. This aspect corresponds to the values resulted from the present research for the ratio  $\text{NO}_3^-/\text{Cl}^-$ , which is smaller than the unity. In figure 5 one can notice

that, no matter what the concentration of the chlorines is, the nitrate amounts smaller than 20 mg/l represent 70% of all data.

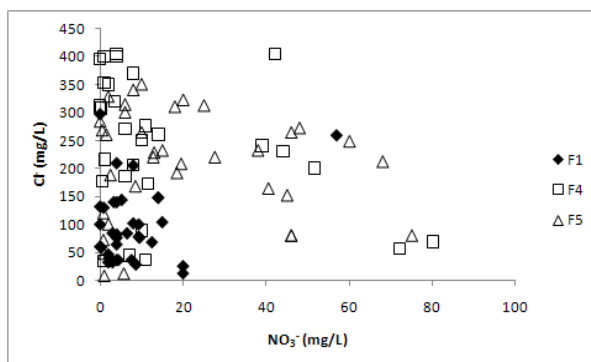


Fig. 5 Relationship between  $\text{Cl}^-$  and  $\text{NO}_3^-$  concentration for shallow groundwater

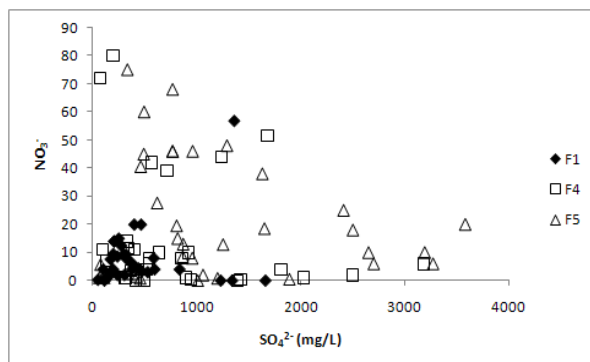


Fig. 6 Relationship between  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentration for shallow groundwater

High amounts of nitrate are systematically associated to waters very high on  $\text{SO}_4^{2-}$  (Schiavo et al., 2006). One of the fertilisers used in agriculture is the ammonium sulphate. If we associate the pollution of underground waters with the agricultural activity, it would be expected to have correlations between high concentrations of nitrate and high sulphate values. From figure 6 one can notice that there is no such a link, the values of the correlation coefficient varying between -0.18 (in F4) and 0.21 (in F1). This

conclusion confirms that this type of fertiliser is not widely used in our country, its registration and certification in Romania taking place in 2007.

## Conclusions

The shallow groundwater from the Cristești section are divided into two major categories:  $\text{HCO}_3^- - \text{Na}^+$  waters, situated at the foot of the terraces and  $\text{SO}_4^{2-} - \text{Na}^+$  waters from the common alluvial plain of the rivers Jijia – Prut. The chemical composition of the waters depends on the nature and structure of the lithologic substratum, on the rainfalls regime, on the water – soil – atmosphere interaction and on the anthropogenic factor. Along the WSW – ENE direction of drilling, one notices an increase of the TDS and of the saturation index in gypsum.

The studied waters do not comply with the STAS 1342/91 requirements, harmonised with the European legislation by the Law 311/04, and are not drinkable. For this reason, they are part of the risk underground water category, as well as the whole ROPR02 body of water. Among the causes that lead to the degradation of the water quality there are hydrogeochemical processes that favour the dissolution of different ions, the presence of fertilisers in soil and the mineralisation of the organic matter, as well as the effect of the conservation of the former zootechnical centres.

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