


Hydrochemical evaluation of spring's water in Mila Wilaya, North-East Algeria

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ABSTRACT

Springs are natural outflows of groundwater to the surface, enabling to understand the processes of their mineralization; contact with rocks modifies their chemical composition, a combination that is expressed in hydrochemical facies, which are classified into several types. Knowledge of the spatial distribution and chemical composition of spring waters is essential for a good understanding of the hydrodynamisms and hydrogeology of a given region. The mountains bordering the town of Mila are home to a number of groundwater springs, 25 of which are the subject of this study. They were sampled, then analysed at the ADE Mila (Algerian Water Company) laboratory in 2021, and their hydrochemical properties were determined. Geologically, the area hosting these springs belongs to the Tellian domain, the eastern segment of the Maghreb chain, which represents the alpine range of North Africa. Comparison of the water quality of these springs with Algerian and WHO (World Health Organization) drinkable standards has enabled most samples to be classified as soft or moderately hard water, with the dominant facies HCO_3^- - Ca^{2+} - Mg^{2+} and Cl^- - SO_4^{2-} - Ca^{2+} - Mg^{2+} . In these waters, the main factors controlling hydrochemical composition are water–rock interactions, and rainwater contribution.

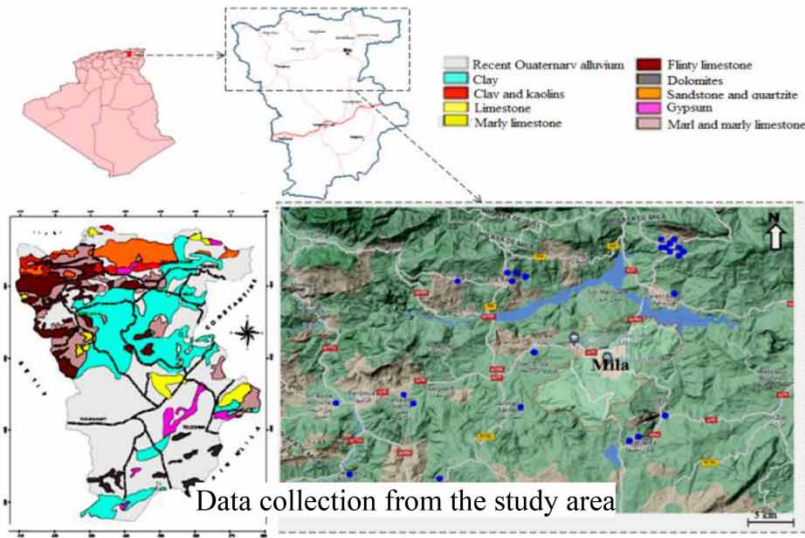
Key words: drinkability, facies, hydrochemical properties, Mila, spring's water

HIGHLIGHTS

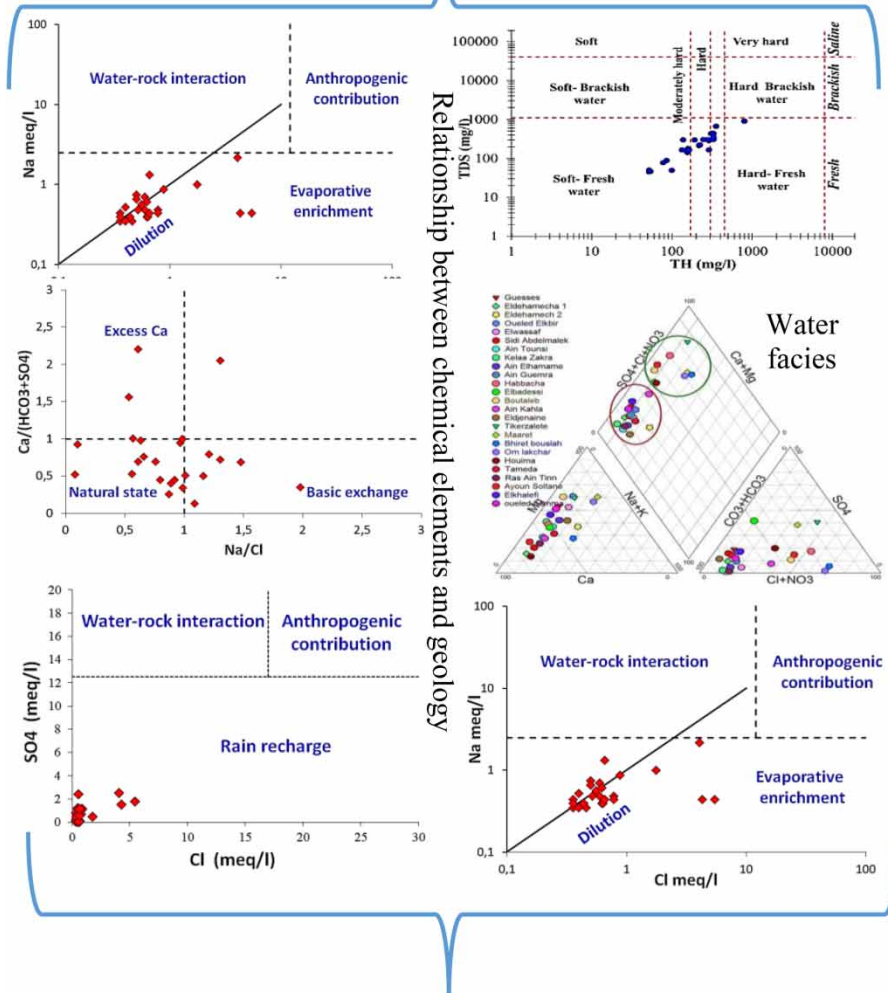
- The source water is geographically repertory, sampled and undergoes a well-determined physico-chemical analysis protocol, to identify the elements that form their mineralization.
- Assessment and classification of source water quality against local and international standards.
- Determination of the origin of the chemical elements that constitute the mineral charge of these waters.
- The chemical characteristics of spring's water have been shown to be influenced by the leaching process, which shows the influence of local geology.
- Spring water bears the signature of the flow of precipitation that seeps into the soil and changes its chemical quality.

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GRAPHICAL ABSTRACT



R E S U L T S



1. INTRODUCTION

Groundwater provides an estimated 25–40% of all the world's drinking water. It can interact with the different minerals in the aquifer and become enriched in several elements, some of which are good for health, others less so and some of which can even be toxic if critical concentrations are exceeded (Ferreira *et al.* 2017). Geological processes are the main influences on water chemistry, due to infiltration and circulation in deeper layers, water changes its chemical composition while interacting with different geological layers (Jasik *et al.* 2020).

Springs result from a concentration of groundwater flow paths, so that water flows out of a single location (or a small number of nearby locations) rather than diffusely into surface watercourses. As such, spring water chemistry reflects the interaction of groundwater with the host rock of the aquifer as well as any chemical constituents that may be introduced from surface sources. Spring water chemistry is therefore not intrinsically different from groundwater chemistry and the same principles apply (White 2010).

The quality of spring waters depends not only on natural factors such as aquifer lithology, groundwater velocity, recharge water quality and interaction with other types of water or aquifers, but also on anthropogenic activities, which can alter these fragile systems, either by polluting them or by changing the hydrological cycle (Helena *et al.* 2000).

In Mila Wilaya, there are 415 springs identified with a mobilized flow of 11,633,315 m³/year (Soukehal & Cherrad 2011). In the rural areas of Mila, springs are the most important source of water for the people living in this region. They are found almost everywhere and play a vital role in the life of the inhabitants. Most of the Communes are located in hilly and isolated areas, where spring water has been used to satisfy domestic needs (washing, drinking), livestock watering and irrigation of agricultural fields by the villagers of these Communes.

The main objective of this article is to determine the hydrochemical waters properties of 25 springs, located in the Mila region, to evaluate their drinkability, and understand the composition and origin of the chemical elements that make up the mineral content of these waters, as well as, how they are acquired?

The hydrochemical properties of these waters springs samples were analysed during the year 2021 at the ADE laboratory of Mila (Algerian Water Company).

2. MATERIALS AND METHODS

Mila Wilaya is located in northeastern Algeria, about 50 km north-west of Constantine (Figure 1). It is characterized by a cool sub-humid climate characteristic of the mountainous relief and influenced by the presence of the large Beni Haroun dam (Rullan-Perchirin 1985; Côte 1998; Chebbah & Kabour 2018). The climate is marked by large temperature differences; temperatures in summer are quite high and around 40 °C, in winter they are generally low and can sometimes fall below zero degrees. Rainfall is quite high with an average of over 600 mm/year (standard deviation SD = 88, 96 mm, for the period of 1998–2003) (Chebbah & Kabour 2018).

The hydrographic network is extremely dense and hierarchical, the shape of the region as a basin with an East-West corridor shows a general orientation of the hydrographic network South-North or North-South depending on the case (Mebarki 1982).

Mila is located at the limit between two geological provinces, and seems to present a less important seismicity than the neighbouring regions such as the Constantine or Babors area. It belongs to the Tellian domain, the eastern segment of the Maghrebic chain, which represents the alpine chain of North Africa. This chain, made up of aquifers put in place during the Miocene, was structured by the Alpine Orogen, their base could be dated to the terminal Burdigalian and their top to the Quaternary (Zouaoui 2008; Chettah 2009).

The formations in the Mila region are primarily transgression deposits from the continental and marine realms (Mio-Pliocene detrital and evaporite deposits). One of the geological characteristics of the Mila basin is the predominance of clay deposits leading to numerous ground instabilities. The crushing zones of the major tectonic accidents generally constitute favourable zones for the infiltration, circulation and storage of water. Indeed, these zones constitute good aquifers and often show spring resurgence (Athmania *et al.* 2010). The 25 springs studied are located in the northern communes of Mila Wilaya (Figure 1).

The methods described by Rodier (Rodier *et al.* 2009) were followed during field and laboratory work. The goal was to ensure that the water samples truly represented the hydrochemical properties and conditions of their natural environment.

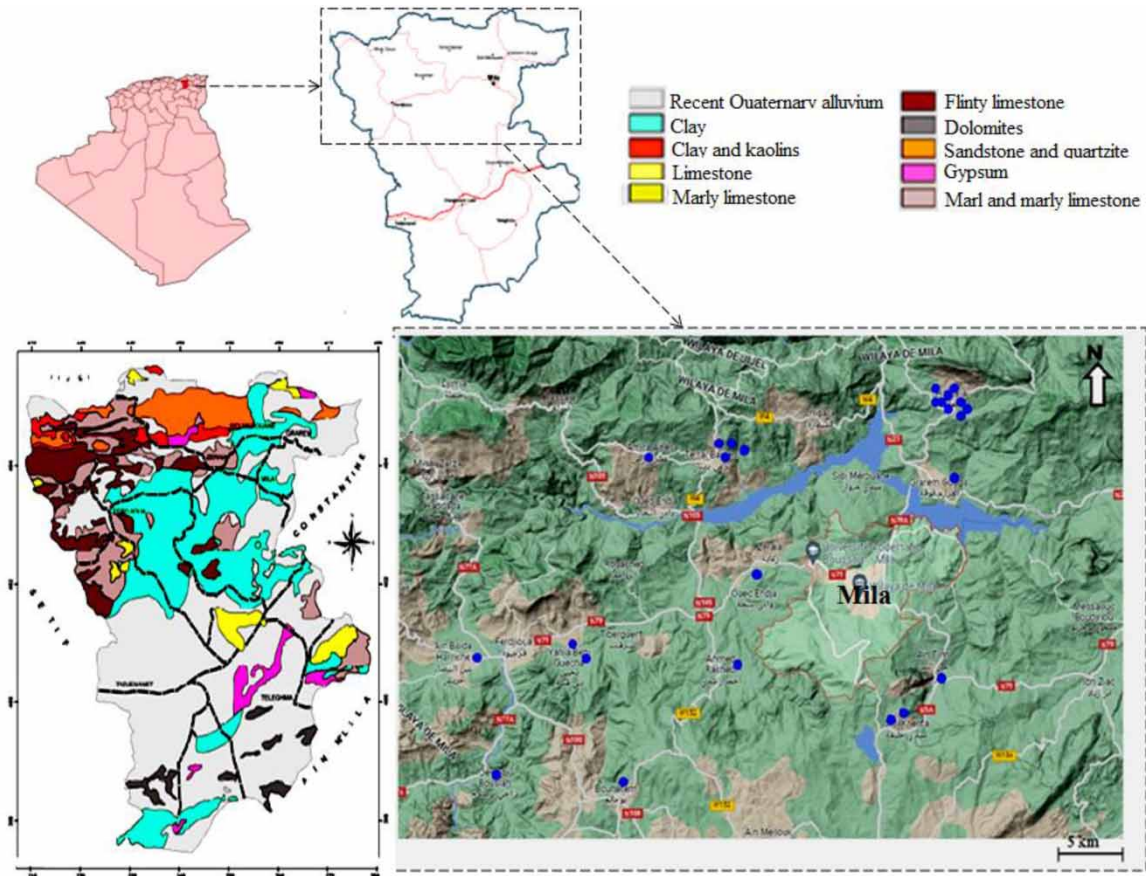


Figure 1 | Geographical and geological location of the study area and location of the studied springs.

Water was taken directly from the springs, where electrical conductivity (EC), salinity (SAL), TDS (with a Hach multiparameter, sensION156) and pH (Adwa, AD 1000) were measured on-site. Other elements are measured in the laboratory, such as turbidity (Hach, 2100N). Concentrations of potassium (K^+), sodium (Na^+) and calcium (Ca^{2+}) were analysed using a flame photometer (JENWAY, PFP7), while those of nitrate (NO_3^-), nitrite (NO_2^-) and total hardness (TH) were titrated using the volumetric method; sulphate (SO_4^{2+}), magnesium (Mg^{2+}) and chlorine (Cl^-) by spectrophotometry (Hach, LANGE, DR3900). Samples were analysed for the main cations and anions used for chemical characterization.

The application of graphical methods to raw data enables results to be visualized, compared and interpreted more quickly and easily. The graph of total dissolved solids (TDS) versus TH (in Excel) enables samples to be positioned within their water quality ranges. The Piper diagram enables the facies of each sample to be determined (created by Diagram V6.77) based on its anion and cation composition. The role of the Gibbs diagram (in Excel) is to visualize the origin of certain chemical elements present in the analysed water. The graphical representation of the Ca/Mg ratio (in Excel) shows the origin of calcium, as a function of its content in the water.

3. RESULTS

3.1. Major ion chemistry

The use of statistical analysis of major ions (Mg^{2+} , Na^+ , K^+ , Ca^{2+} , SO_4^{2-} , NO_3^- and Cl^-) as a basic tool to interpret the hydrogeochemical characteristics of groundwater (Gao *et al.* 2019). Table 1 presents the statistical description of the main ions in Mila spring waters.

Dissolution of carbonates is an important origin of Ca^{2+} and Mg^{2+} ; the dominant cation present in the collected water samples is Ca^{2+} . The concentration of Ca^{2+} varies from 4.01 to 272.54 mg/l. The increase in Ca^{2+} concentration in the samples is caused by the dissolution of gypsum and limestone, this ion may also be due to cation exchange (Li *et al.* 2016). Mg^{2+} ranges from 4.84 to 55.89 mg/l. The average values of Na^+ concentrations vary from 8 to 50 mg/l, with a maximum concentration of Na^+ observed at the level of the El

Table 1 | Descriptive statistics of the main parameters of Mila spring’s water, 2021

Statistics	Unit	Average	Max	Min	Standard deviation
EC	μS/cm	517.18	1,808.00	78.70	291.60
TDS	mg/l	255.98	904.00	45.00	199.20
Sal	%	0.23	0.90	0.00	0.21
Turb	NTU	4.70	36.00	0.62	7.66
TH	mg/l	223.52	800.00	52.00	154.88
Ca ²⁺	mg/l	53.38	272.54	4.01	53.64
Mg ²⁺	mg/l	23.19	55.89	4.84	12.67
Na ⁺	mg/l	14.12	50.00	8.00	9.13
K ⁺	mg/l	1.2	7.00	0.00	1.58
HCO ₃ ⁻	mg/l	190.87	378.20	9.76	113.89
Cl ⁻	mg/l	38.80	193.00	12.76	48.46
SO ₄ ²⁻	mg/l	31.55	121.33	0.70	30.99
NO ₃ ⁻	mg/l	5.71	20.42	0.00	7.06
NO ₂ ⁻	mg/l	0.03	0.10	0.00	0.03
OM (Organic matter)	mg/l	1.11	2.33	0.00	0.61
DR (dry residue)	mg/l	333.92	1,314.00	4.00	279.95

Dehamecha 2 spring, in the Commune of Sidi Khelifa (south of the study area). The average concentration of cations in the spring waters are observed as follows: Ca²⁺ > Mg²⁺ > Na⁺ > K⁺.

The dominant anion in the spring water samples is HCO₃⁻, with average concentrations in the following order: HCO₃⁻ > Cl⁻ > SO₄²⁻ > NO₃⁻. HCO₃⁻ varies between 9.76 and 378.20 mg/l in, with an average of 190.87 mg/l. The average concentrations of Cl⁻ and SO₄²⁻ are 38.8 and 31.55 mg/l, respectively. Anthropogenic inputs, such as industrial and agricultural activities, are a major source of NO₃⁻ in groundwater (Kabour 2017). The concentration of NO₃⁻ in the study area found varies from 0 to 20.42 mg/l.

Among the indices for assessing water quality, two are important: TDS (total dissolved solid) and TH (hydro-metric titre) (Gao *et al.* 2019). TDS concentrations vary from 45 to 904 mg/l and TH from 52 to 800 mg/l. The graph in Figure 2 shows that 48% of the samples belong to the soft fresh water domain, 28% to the moderately hard fresh domain, 24% are hard fresh and 4% are hard fresh water. This characteristic probably results from the enrichment of the groundwater in dissolved salts from the carbonate formations.

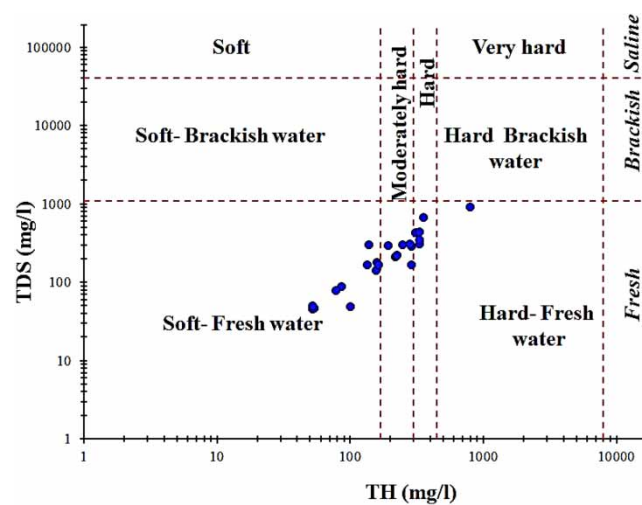


Figure 2 | Graph of total dissolved solids (TDS) versus total hardness (TH) of Mila spring’s water, 2021.

3.2. Water facies

The Piper diagram is considered one of the simplest and widely used methods for determining groundwater facies (Gao *et al.* 2019). In this study, the Piper diagram is obtained using 'Diagram v 6.77' software.

The hydrochemical composition (major cations and anions) of the Mila spring waters was plotted on a Piper diagram (Figure 3), in order to highlight the main hydrogeochemical characteristics, their volume and the processes that affect the spring waters (Piper 1944). Two main hydrochemical facies were identified: HCO_3^- - Ca^{2+} - Mg^{2+} and Cl^- - SO_4^{2-} - Ca^{2+} - Mg^{2+} . These two facies are formed from the sequence of processes that led to the formation of the spring water, which modify the chemistry of the water along the flow path, and especially the lithology of the subsurface geological formation. The HCO_3^- - Ca^{2+} - Mg^{2+} facies may be due to dissolution of carbonaceous rock and water-rock interaction in shallow aquifers. HCO_3^- is the main anion and Ca^{2+} and Mg^{2+} are the main cations in this facies. The facies Cl^- - SO_4^{2-} - Ca^{2+} - Mg^{2+} may result from increased cation exchange. The main anion is Cl^- and the next most frequently present anion on this facies is SO_4^{2-} .

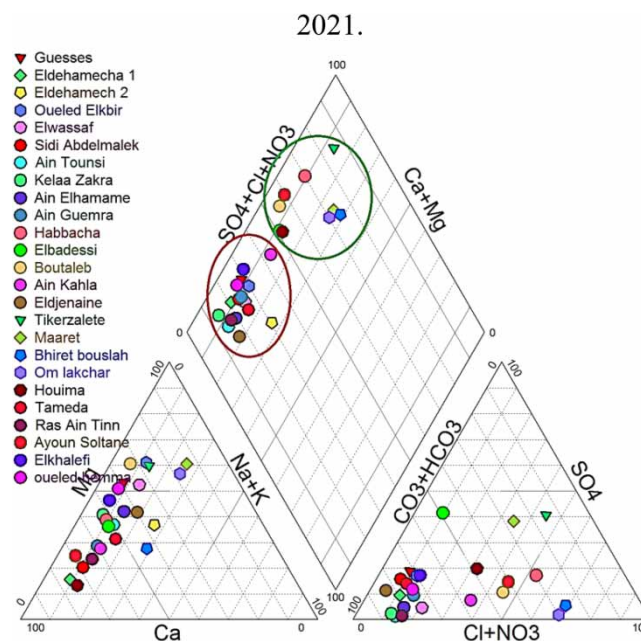


Figure 3 | Piper diagram of the Mila spring's water, 2021.

4. DISCUSSION

4.1. Drinkability of water

It can be noted that most of the samples taken from Mila springs are considered to be of freshwater type. The bicarbonate concentration in the majority of these springs is in the range that reflects the calcareous origin, except for the springs in the South, which reflect the Cl^- - SO_4^{2-} - Ca^{2+} type (Houima, Ayoun soltane and Ain Kahla). The results show that a high concentration of Mg^{2+} generally accompanies a high concentration of Ca^{2+} , which may be an indication of dolomitization, especially for these sources. Low concentrations of Mg^{2+} reflect the carbonate origin.

Table 2 gives the thresholds set by Algerian regulations for spring water (JORADP 2000, 2011; Hazzab 2011; Kabour 2017) and the standards of drinkability according to the WHO (World Health Organization) regulations (W.H.O. 2017). Referring to these standards, we note that all sampled sources are within Algerian and WHO standards, except for Tikerzalet (0.347 mg/l) and Maaret (0.486 mg/l), which slightly exceed the iron concentration threshold. The Houima and Ouled Belkhir springs have a concentration of 272.54 mg/l in Ca^{2+} and 55.89 mg/l Mg^{2+} respectively resulting from the dissolution of the carbonate formations of Jebel Msid Aicha. These concentrations slightly exceeds both standards. The three springs are located in the north of the study area.

Table 2 | Comparison of water quality indicators for Mila springs, 2021

Features	Symbol	Unit	Maximum permissible value according to the standards		
			Algerian (JORADP 2000, 2011)	WHO (W.H.O. 2017)	Mila sources maximum concentration
Electrical conductivity	EC	μS/cm	2,800	2,000	1,808
Chlorides	Cl ⁻	mg/l	200–500	250	193
Sulphates	SO ₄ ²⁻		400	250	121.33
Calcium	Ca ²⁺		75–200	100	272.54
Magnesium	Mg ²⁺		150	50	55.89
Sodium	Na ⁺		200	150	50
Potassium	K ⁺		20	12	7
Nitrates	NO ₃ ⁻		50	50	20.42
Nitrites	NO ₂ ⁻		0.1	3	0.1
Iron	Fe		0.3	0.3	0.486

Values that exceed the maximum permitted are shown in bold.

4.2. Mechanism for acquiring spring water chemistry

The use of the Gibbs diagram (Figure 4) indicates that the chemical composition of most of the Mila spring water samples was controlled mainly by the weathering reaction of the rocks (84%). The interaction between rocks and water leads to the leaching of ions into the groundwater system, thus influencing the water chemistry (Ferreira *et al.* 2017). Only four points (16%) may have been influenced by rainfall dominance, which are located in the north of the study area (Terai Beinen).

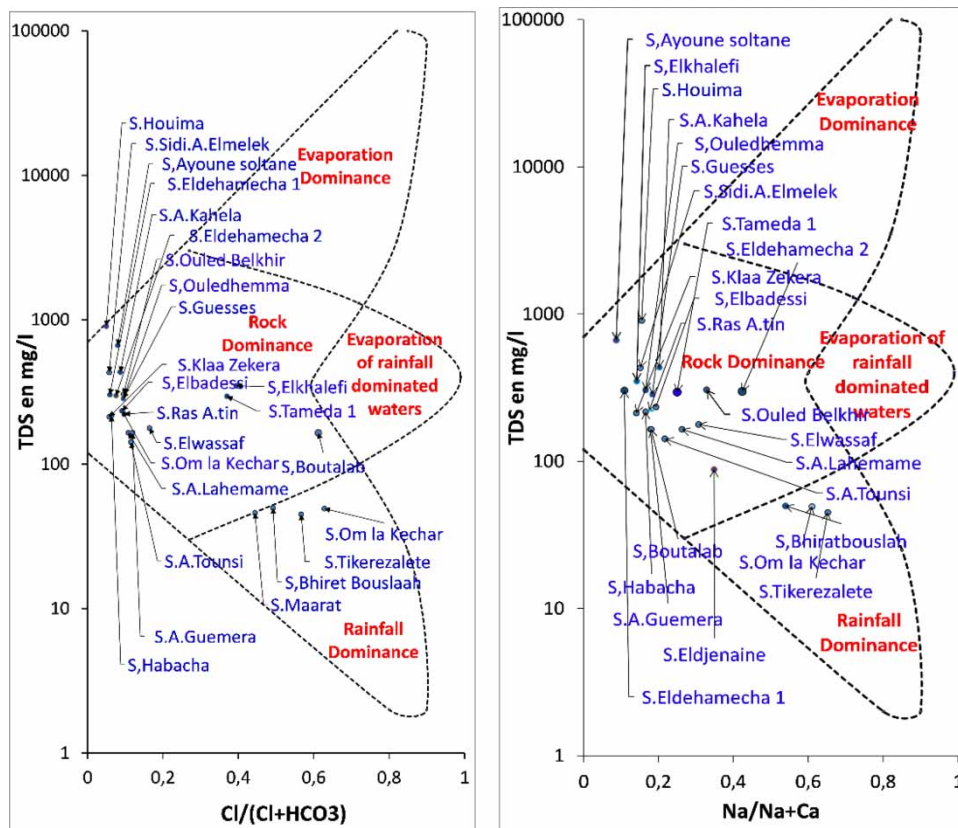


Figure 4 | Gibbs diagram, Mila spring's water quality control mechanisms, 2021.

4.3. Origin of chemical elements

The molar ratio Ca^{2+}/Mg^{2+} gives information on the origin of these elements from the dissolution of calcite and dolomite. If this ratio $Ca^{2+}/Mg^{2+} \leq 1$, it is a dissolution of dolomite, on the other hand, a higher ratio >1 indicates the dissolution of calcite (Mayo & Loucks 1995), when it is higher than 2, it indicates the dissolution of silicate minerals (Katz *et al.* 1997; Nair *et al.* 2015).

The spring waters of the Mila region are characterized for the majority of cases (52%), by ratios higher than 2 due to dissolution of silicate minerals (clay and phyllosilicate), 28% expressing dissolution of calcite and 20% expressing a dissolution of dolomites (Figure 5).

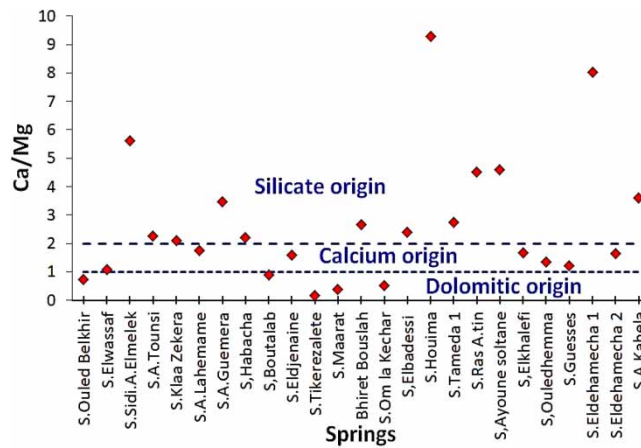


Figure 5 | Origin of calcium in spring water in the Mila spring's water.

The use of the ratio SO_4^{2-}/Cl^- indicates that all the analysed samples are in the 'rain recharge' range. The ratio is lower than 1 for 64% of the analysed samples, which determines the predominance of chlorides over sulphates, explained by the leaching of the gypsum and marl soils located upstream on the path of the recharge waters (Kabour 2017). Figure 6 shows that 100% of the spring water is of rain-fed origin.

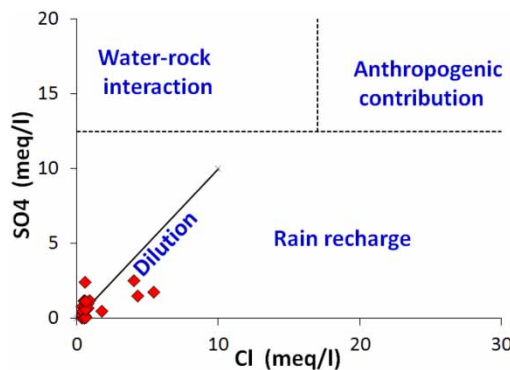


Figure 6 | Origin of SO_4^{2-} .

Na^+ and Cl^- are two chemical elements present in natural waters and are often linked to the dissolution of halite (NaCl). The evolution of Na^+ is studied as a function of the chloride content, because chloride is considered to be a stable and conservative tracer of evaporites, very soluble, rarely entering into saline precipitations, not involved in redox phenomena and little influenced by the intervention of bacteria. In addition, its movement is not delayed in water (Ouandaogo-Yameogo *et al.* 2013; Kabour 2017).

The graphical representation of the evolution of sodium contents as a function of chloride concentrations on a logarithmic scale (Figure 7) makes it possible to observe four acquisition processes of Na^+ and Cl^- ions: dilution, anthropogenic input, water/rock interaction and enrichment by evaporation (Debieche 2002; Kabour 2017). In

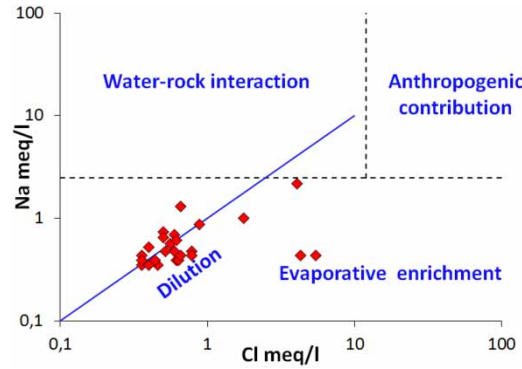


Figure 7 | Origin of Na⁺ and Cl⁻.

our case, the dilution constitutes 100% of the spring water (Figure 7), this ratio is lower than 1 for 72% of the analysed water samples representing spring water in the Mila region, which can be explained by the leaching of clayey soils by the recharge water (Figure 7).

The projection of the different points on the Mg²⁺/Cl⁻ diagram makes it possible to determine the different origins of the Mg ion (Chery 2006; Kabour 2017). We observe that the ratio is greater than 1 for the majority of the points, which indicates a dominance of Mg²⁺ over Cl⁻; in our case, their origin results mainly from dilution (Figure 8).

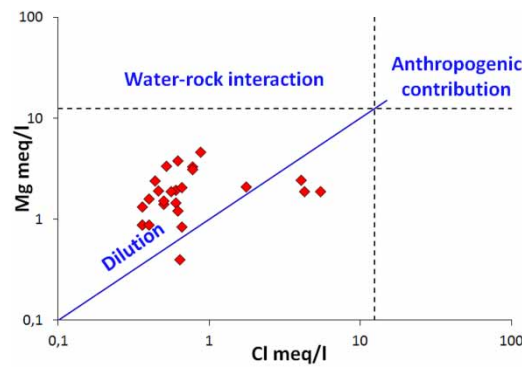


Figure 8 | Origin of Mg²⁺.

The examination of the correlation between Ca²⁺/(HCO₃⁻ + SO₄²⁻) and Na⁺/Cl⁻ allows to understand the origin of calcium according to two chemical elements bicarbonates and sulphates (Chery 2006; Kabour 2017), and indicates the main basic exchanges that take place in the water/rock environment, and also the two main origins of Ca²⁺, namely carbonates and gypsum (Figure 9).

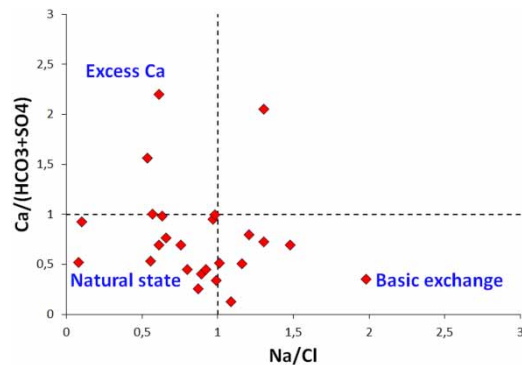


Figure 9 | Basic exchange.

The exchange with the rock matrix also controls the calcium content in the spring waters of the Mila region, in case the residence time is relatively long.

There is a decrease in the calcium content in relation to $\text{HCO}_3^- + \text{SO}_4^{2-}$, and a decrease in Na^+ in relation to Cl^- , which implies a high content of HCO_3^- and SO_4^{2-} , which is frequently found in the solution of evaporite sedimentary rocks, particularly gypsum (CaSO_4).

The projection of the different points on the $\text{Ca}^{2+}/(\text{HCO}_3^- + \text{SO}_4^{2-}) - \text{Na}^+/\text{Cl}^-$ diagram (Figure 9) shows that 24% of the water points undergo a base exchange with a long residence time. 64% of the points present the natural state and 12% express an excess of Ca^{2+} compared to HCO_3^- and SO_4^{2-} ions, probably due to the gypsum formations, which provide more Ca^{2+} ions than can be consumed by the effect of chemical precipitation or the effect of calcium fixation by the clays (Debieche 2002).

5. CONCLUSIONS

This work presents the hydrogeochemical characteristics of the waters of 25 springs located in the region of Mila, as well as the evaluation of their suitability for drinking and domestic use. This characterization allowed to determine the order of classification of the concentrations of the main cations in these waters, which is $\text{Ca}^{2+} > \text{Mg}^{2+} > \text{Na}^+ > \text{K}^+$, while for the anions, it is: $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^-$.

Referring to the graph of TDS versus TH, most of the water samples belong to the fresh–fresh and soft–moderately hard water domain, and the dominant water types are of mixed type.

Rock leaching and precipitation inputs are the predominant processes in the formation of hydrochemical components. The dissolution of silicate, calcite, dolomite and gypsum are the main processes that contribute to define the chemistry of Mila's spring waters; there is also another important hydrogeochemical process that contributes to the elaboration of this quality, which is the exchange of cations, such as Ca^{2+} and Mg^{2+} with Na^+ and K^+ .

According to Algerian and WHO standards, the quality of spring water in the Mila region is of excellent class for most of the samples analysed.

In order to preserve and protect the quality of these springs against the accelerating process of urbanization and industrialization, it is necessary to establish a strategy of control, surveillance and reinforcement of the monitoring of the physicochemical quality, in order to achieve a sustainable development and management that respects the environment and allows the rational use of spring water.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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