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Assessment of Physico-Chemical Quality of Fresh Water Springs in Village Pepaj, Rugova Region, Kosova

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Abstract: Springs are the most important source of fresh water for drinking and other household utilization in the Rugova Mountain Region. Spring is a natural source of groundwater. The purpose of this study was to assess the quality of spring waters in the village Pepaj, Rugova Region. The spring water samples were taken from the main water sources where maximum peoples were using them for drinking purpose. The samples collected from these springs have been analysed for physico-chemical parameters such as temperature, pH, dissolved oxygen, electrical conductivity, total dissolved solids, total hardness, HCO_3^-, NH_4^+, NO_3^-, PO_4^-, SO_4^{2-}, Cl^- and F^-. The values of those parameters have been evaluated with respect to guidelines provided by World Health Organization (WHO) and European Union (EU) standards. This study indicated that the values of each parameter were found to be within the permissible limit values, which shows the spring waters is safe for drinking and other domestic uses. The concentrations of all parameters varying according to the nature of the terrain, through which they flow, geological structure, soil and species composition of tree stands. Several differences were identified between the spring waters temperature, pH, electrical conductivity, relatively high concentration of bicarbonate ions, low concentrations of biogenic compounds such as NO_3^- and NH_4^+, while PO_4^{3-} were not found to be present, F^- and Cl^- concentrations were also determined to be low.

Keywords: *Rugova Mountain Region, spring water, drinking water quality*

Introduction

Water is essential for life on earth. Human existence mainly depends on fresh water supply which is less than 1% of the water available on earth. Water quality is important for health and economic development, chemical composition of water varies with seasons, anthropogenic influences and natural processes (Reagen & Bookins, 1997). Drinking water is generally obtained from surface waters, such as lakes, rivers and streams, and groundwater, such as spring and well waters. A spring is a place where groundwater flows naturally from rock or soil onto the land surface or into a body of water. Some springs discharge where the water table intersects the land surface, but also they occur where water flows out from caverns or along fractures, fault or rock contacts that come to the surface. Spring may result from karst topography where surface water has infiltrated the earth's surface (recharge area), becoming part of the area's groundwater that travels through a network of cracks and fissures/openings ranging from intergranular spaces to large caves. The forcing of the spring to the surface can be the result of a confined aquifer in which the recharge area of the spring water table rests at a higher elevation than that of the outlet (Bell, 1998).

Springs are the most important source of water for the people living in the Rugova, mountain terrain. They are found almost everywhere but people who lives there, even in ancient times, have sought the best quality spring waters as drinking and other domestic purposes. Most of the settlements are located in remote at the top of the ridge, where the springs have higher quality. In this context, springs were largely responsible for determining the sites of ancient settlements (Meuli & Wehrle, 2001). Nowadays, modern techniques allow the construction of deep drilled wells. Yet, springs are still an important groundwater source since they are easily accessible and usually provide clean drinking water (Smet & Wijk, 2002). Occurrence of these springs depends on recharge area characteristics such as surface cover, geology of the area, permeability of top soil, soil structure and slope of ground surface. Generally springs occur where ground surface and the impermeable subsurface strata intersect with the groundwater table. The discharge of the spring water fluctuates

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seasonally and mainly depends on rainfall pattern in the recharge area and variation in the amount of rainwater that is able to infiltrate the ground.

Spring water is usually safe from contaminants (i.e. pathogens, chemicals, metals); since groundwater is naturally filtered as it flows through the soil. Hence, spring water is generally safe for human consumption (Hawley, 2003). Though spring water has been considered to be pure because of its filtration through layers of soil, it has its own health and acceptability problems based on concentration level of certain physico-chemical parameters (APHA, 2005). The studies related to quality of water from spring in village Pepaj have not been conducted. The objective of this research was to evaluate the quality of some springs water at the village Pepaj, Rugova Region by assessing the levels of physico-chemical parameters, which justifies the quality of a drinking water.

Materials and Methods

General description of sampling site

Spring water samples were collected from village Pepaj, Rugova Region, Kosova. Rugova Mountains are in the north-western part of Kosova. It is a region within “Bjeshkët e Nemuna”, also known as the Albanian Alps. It was proclaimed a national park in 2013. The region is considered to be the most inaccessible mountain range in Europe and the wildest range on the Balkan Peninsula which is best described in their name. They encompass areas of unique landscape with high craggy peaks, alpine lakes, caves, waterfalls and deep river canyons such as the Rugova Canyon 1,000 m deep and 25 km long. Regarding biodiversity, the area represents one of the most important regions in Europe. In terms of Flora the regions is very rich and diverse with alpine and sub-alpine mountain habitats, alpine meadows and pastures with species rich vegetation, and large forest complexes including primeval tree stands of the endemic Balkan pine (UNEP, 2010).

Rugova is at latitude of 42°44’ N and a longitude of 20°3’ E, and it is 93 km from Prishtina, the capital of Kosova. It covers a territory of 20,330 hectares, and from east to west it extends for 23 km. Rugova has a wet, continental climate that is influenced by the mountains, short and hot summers, and long and harsh winters. Spring is late, and the seasons change quickly. The high level of precipitation is a result of clashes between the tropical and continental climates. The annual minimum and maximum precipitation are 540.6 millimetres and 1,336 millimetres, respectively (Natyra e Rugovës, 2001).

Geology of the Study Area

The springs surveyed are chosen according to the following criteria: accessibility and frequency of use by residents. They are located at geographical coordinates and elevation determined by using a GPS type (GARMIN GPSmap 62). Table 1 shows the names, geographical coordinate and elevation of the springs studied. At those elevations the woodland consists mostly of spruce and pine.

Table 1. Name and geographical coordinates of springs explored

Code	Source name	Geographical coordinate	Elevation (m)
S1	Pusi ne Pllana	X-7428394 : Y-4732916	1691
S2	Gurra e leprit	X-7428549 : Y-4732703	1636
S3	Gurra e kryse	X-7428434 : Y-4732476	1574

The first research on geological composition of the region was conducted in the early twentieth century. These were mainly the research work of the Dinarides tectonic, lithological composition and stratigraphic. The area where the study was conducted spread on the palaeozoic, triassic and quaternary formations. Palaeozoic formations are spread in the southern part, on Sharri Mountains in the west, and in the north in the shape of a band that is few km long. These formations are composed of schist, quartzite, sandstone, limestone and conglomerates. Triassic formations are represented mainly by limestone and dolomite which are over 600 m thick, which are formed in quiet conditions of sedimentation. Limestones are tectonically destroyed, with lot of cracks and seams. The network density of these cracks sometimes favours erosion and karst processes which in their turn serve as a way for the passage of groundwater (Antonjevic *et.al*., 1978). Quaternary sediments are widespread in this region. Pleistocene and Holocene sediments are found in this region as well represented by glacial and fluvioglacial while proluvium formations, deluvium and alluvium have greater spread along the valley. In Figure 1 is shown the topography map and location of study area, village Pepaj, Rugova.

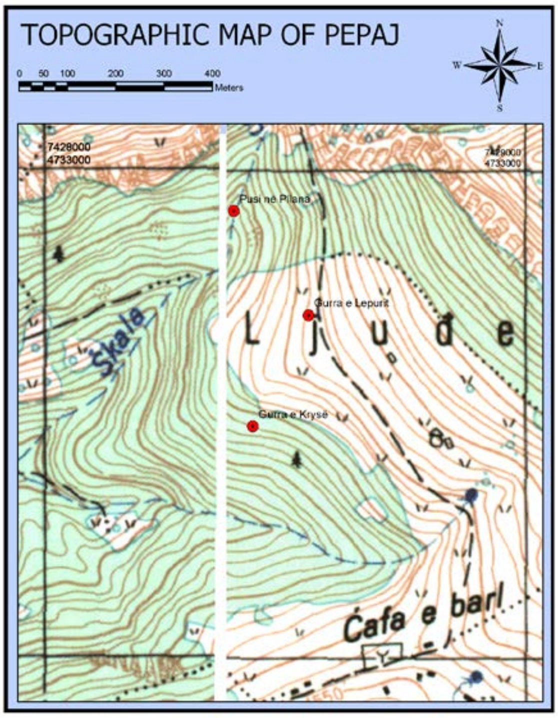


Figure 1. Map depicts topography and location of the springs studied, village Pepaj, Rugova

Sampling and physic chemical analyses

The samples were collected from springs at 10.00 to 12.00 am in the month of October 2016 for physico-chemical examinations. The samples were collected in plastic bottles of two litres capacity without any air bubbles. Prior to collection, the bottles were washed with distilled water and subsequently rinsed thoroughly with the sample water. Samples from the springs S2 and S3 were taken direct form spring outlets while the samples from S1 were taken from waterhole collected direct from spring. For validation of results, three water samples were collected at every site of sampling. The physio-chemical parameter such as pH, temperature and electrical conductivity (EC) were measured in situ in the field using Hanna model multipara meter probe (HI 991301). Dissolved Oxygen (DO) of spring water samples were also measured in situ using portable Dissolved Oxygen Meter model HI9147. All the instruments were calibrated with standard solutions before measurements (Standard Method ISO5814). In meantime the collected samples were stored in cool box and transported to N.SH. Agrovet Laboratory, Prishtina and analysed immediately following the protocol of standard methods (APHA, 2005). Total Hardness (TH) was determined by colorimetric titration using ethylene diamine tetra acetic acid (ETDA) as a complexing agent to bind free multivalent cations (Standard Method 2340C). Bicarbonate (HCO_3^-) was carried out using acid titration, with methyl orange as indicator. Ammonia (NH_4^+), nitrate (NO_3^-), phosphate ($PO4_3^-$), sulphate (SO_4^{2-}), chloride (Cl^-) and fluoride (F^-) were measured using spectrophotometer model HACH DR 3900 (Standard Methods USEPA: 8155, 8192, 8048, 8051, 8113, 10225). Tests were undertaken in duplicate and the results averaged to reduce experimental errors.

Results and Discussion

The results of the physico-chemical parameters analysis of water samples collected from springs in village Pepaj, Rugova Region are summarized in table 2. Water samples collected from the three spring sources have crystal clear appearances, a good taste and were odourless. Physico-chemical quality of spring waters is important not only in the assessment of the degree of pollution but also in the choice of the best source and the treatment needed. The water quality results obtained from these springs were compared to the maximum values recommended by EU (98/83/EC Directive, 1998) and

WHO (2008) standards. The mean values of the studied physico-chemical parameters were found to be within the limits given in the EU (1998) and WHO (2008) standards.

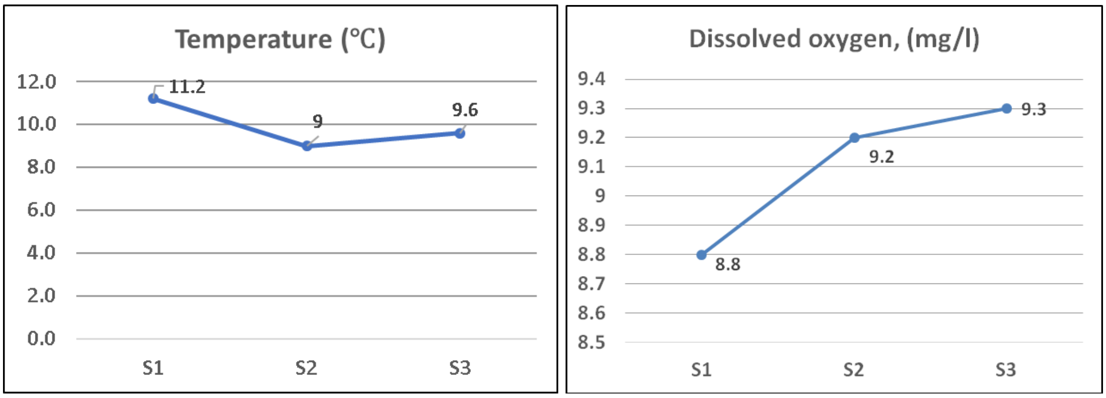
Table 2. Result of the physico-chemical parameters of spring water samples in the village Pepaj, Rugova

Parameters	S1	S2	S3	EU (1998)	WHO (2008)
Temperature, (^{o}C)	11.2	9	9.6	25	
pH	6.7	7.06	6.6	6.5-9.5	6.5-8.5
Dissolved oxygen, mg/l	8.8	9.2	9.3		≥6
Conductivity, µS/cm	480	360	390	2.500	1500
Total dissolved solids, mg/l	307	230	250		600
Total hardness, ^{o}dH	17.08	15.344	15.96		
Bicarbonate, mg/l	305	274	285		500
Ammonia, mg/l	0.152	0.244	0.206	0.5	1.5
Nitrate, mg/l,	1.85	3.12	1.96	50	50
Phosphate, mg/l	<0.02	<0.02	<0.02		0.1
Sulphate, mg/l	5	8	5	250	250
Chloride, mg/l	0.2	0.12	0.1	250	250
Fluoride, mg/l	0.106	0.091	0.082	1.5	1.5

One of the most important characteristics of an aquatic system is temperature because it controls the amount of dissolved oxygen that is present in water and many other chemical reactions. Cool water is generally more palatable than warm water, and temperature will impact on the acceptability of a number of other inorganic constituents, dissolved oxygen in the water and chemical contaminants that may affect taste. High water temperature enhances the growth of microorganisms and may increase taste, odour, colour and corrosion problems. Humans tend to prefer cold water rather than lukewarm, as cold water is likely not containing microbes. The pleasant taste associated with spring waters or mineral waters is derived from the minerals dissolved in it, as pure water is tasteless. The temperature of spring water samples ranged from 9.0°C to 11.2 °C during the study period. Lowest water temperature was observed in spring S2 and highest temperature value was observed in spring S1 (Figure 2a). Higher temperatures of water in spring S1 resulted from the effect of air temperature on the standing water where samples were taken from waterhole collected direct from spring.

The pH of water is a measure of the acid-base equilibrium and, in most natural waters, is controlled by the carbon dioxide-bicarbonate-carbonate equilibrium system. An increased carbon dioxide concentration will therefore lower pH, whereas a decrease will cause it to rise. pH of springs water is influenced by geology of catchment area. Carbonate materials and limestone are two elements that can buffer pH changes in water. Calcium carbonate ($CaCO_3$) and other bicarbonates can combine with either hydrogen or hydroxyl ions to neutralize pH (Weber & Stun). When carbonate minerals are present in the soil, the buffering capacity (alkalinity) of water is increased, keeping the pH of water close to neutral. Additional carbonate materials beyond this can make neutral water slightly basic. Guidance for pH is based on both taste and also corrosion control. When the pH is acidic and below pH 6.5, water can have a bitter, metallic taste. When the pH is more basic than pH 8.5, then water can have a slippery feel and soda taste (Burlingame *et al*., 2007). The pH in the water of the springs analysed ranged from 6.6 (S1) to 7.06 (S2), it was slightly acidic at S1 and S3 (table 2). All the water samples analysed have pH within the safe limit of 6.5 to 8.5 standards set by the EU (1998) and WHO (2008).

Dissolved oxygen (DO) is a measure of the amount of oxygen freely available in water. DO is the most important water quality variable determining the health status of an aquatic ecosystem (Badran, 2001). Oxygen enters the water by photosynthesis of aquatic biota and by the transfer of oxygen across the air-water interface. The oxygen content of natural waters varies with temperature, salinity, turbulence, the photosynthetic activity of algae and plants, and atmospheric pressure. The solubility of oxygen decreases as temperature and salinity increase. Water low in dissolved oxygen has an unpleasant smell while waters high in dissolved oxygen are good for drinking purposes. The permissible limit of the DO in drinking water should be ≥6 mg/L (WHO, 2008), whereas the dissolved oxygen is found to be higher in all the spring samples. The value for the DO of each source analysed is shown in figure 2b. DO of samples S1 (8.8 mg/l), S2 (9.2 mg/l) and S3 (9.3 mg/l) were within the EU (1998) and WHO (2008) acceptable limits.



(a) (b)

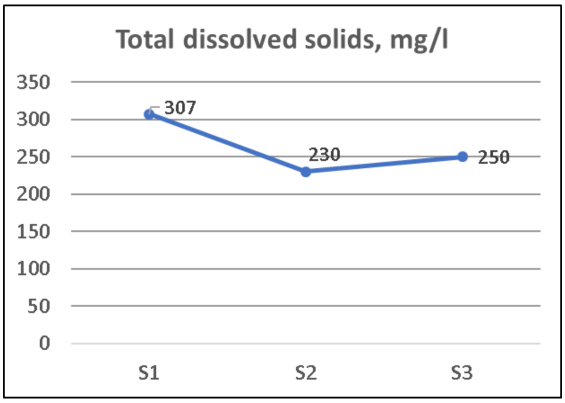
Figure 2. Variation of temperature (a) and dissolved oxygen (b) of spring waters for comparison of their quality

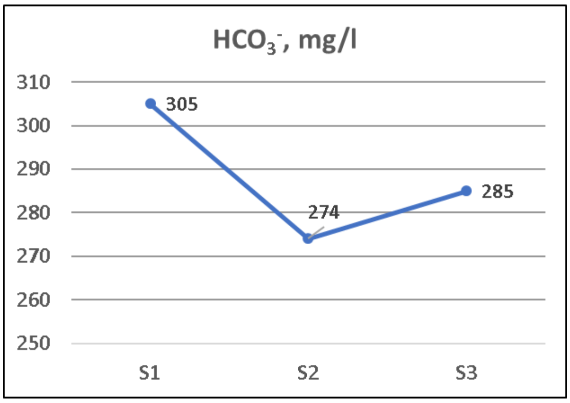
Electrical conductivity (EC) reflects the ability of an aqueous to conduct electrical current that depends on the presence and total concentration of ions, their mobility and valance and on the temperature (Clesceri *at al*., 1998). High values of EC in groundwater are due to high dissolved solids. In present study, the values of conductivity in all sampling points ranged from 360 to 480 µS/cm. Spring (S1) had the highest electrical conductivity (table 2). The values of conductivity obtained in all sampling sites were within the standards value of EU (1998) and WHO (2008). The difference in conductivity can be attributed to the geology of underground water system such as the type of rocks and minerals through which water flows. For example, limestone leads to higher conductivity because of the dissolution of carbonate minerals. Contact time between water and rock, the flow rate, and the flow path could also affect conductivity.

Total dissolved solids (TDS) are an aggregate measure of minerals in water. TDS can also include dissolved organic matter, although this constituent is minor compared to inorganic minerals. The amount and type of minerals in drinking water are the major determinants of taste (Burlingame *et al*., 2007). Concentrations of TDS from natural sources have been found to vary widely. While the level of TDS is an important factor for drinking water preference, TDS levels are also important in the ability to discriminate between different waters (Dietrich & Gallagher, 2013). The concentrations of TDS in all sampling sites ranged from 230 to 307 mg/l. These values were within the standard limits of drinking water quality set by EU (1998) and WHO (2008). Results from analysis, also shows a similarity in trend between values of electrical conductivity in the area under the study. The trend decreases in order S1>S3>S2 (figure 3a). Thus a low level of TDS contents of the spring waters allows the water for drinking and other domestic uses.

Bicarbonate, HCO_3^- is an ion that is common to all natural waters because all bicarbonates are water soluble. Bicarbonate in combination with calcium and magnesium forms carbonate hardness. Bicarbonate concentrations in natural waters varied from less than 25 mg/l in non-carbonate rocks areas to over 400 mg/l where carbonate rocks are distribute. A high concentration of bicarbonates in drinking water is deviated by weathering of calcium bearing rocks. The dissolved carbon dioxide derived from rain is the primary source of bicarbonate ions in spring water. As it enters the soil and rocks, it dissolves more carbon dioxide in water. Carbonates such as limestone, dolomite and calcite dissolve to form bicarbonates by the action of CO_2 on these basic materials (Ritzi *et al*., 1993). Bicarbonate ions concentrations in the water of the springs were between 274 and 305 mg/l, the lowest value was recorded at spring S2 and the highest value was recorded at spring S1 (figure 3b). All the samples were found to be within safe limits of EU (1998) and WHO (2008) standards.

Total hardness is the property of water which prevents the lather formation with soap and increases the boiling points of water. Hardness, which occurs naturally in water, is an aggregate parameter that is the sum of aqueous divalent cations. Calcium and magnesium are the major divalent cations in natural fresh waters, and hence the major ions in hardness. Both calcium and magnesium are essential minerals and beneficial to human health in several respects. Inadequate intake of either nutrient can result in adverse health consequences. Occasionally, ferrous ion or the manganese ion can contribute to hardness, but these metals are minor. Hardness varies substantially with the presence of limestone and dolomite in the local geology (Combs, 2005). High levels of total hardness are not considered a health concern. On the contrary, calcium is an important component of cell walls of aquatic plants, and of the bones or shells of aquatic organisms. Magnesium is an essential nutrient for plants, and is a component of chlorophyll. Total hardness values for the three springs studied show no major difference from one spring to the other. They vary between 15.344 ^{0}dH and 17.08 ^{0}dH, respectively recorded at the springs S1 and S2. Classification of water hardness in German degrees is: soft (less than 8.4 °dH), medium (8.4 - 14 °dH), hard (14 - 20 °dH) and very hard (more than 20 °dH). Based on these results, all spring waters are generally hard. This hardness is the result of limestone terrain crossed by water. The water quality analysis showed the hardness values of the spring water samples were within permissible limit and is safe for drinking and other domestic uses.





(a) **(b)**

Figure 3. Variations of TDS (a) and HCO_3^- (b) of spring waters for comparison of their quality

Ammonia, NH_4^+ in the environment originates from metabolic, agricultural and industrial processes and from disinfection with chloramines. Natural levels in groundwater and surface water are usually below 0.2 mg/l. Intensive rearing of farm animals can give rise to much higher levels in surface water. Ammonia in water is an indicator of possible bacterial, sewage and animal waste pollution. Ammonia in drinking-water is not of immediate health relevance, and therefore no health-based guideline value is proposed (ATSDR, 2004). Low levels of ammonia are naturally abundant in the environment. Ammonia is essential for life as it provides a source of nitrogen for proteins and other essential biomolecules. In the present study the lowest concentration of ammonia was 0.152 mg/l at S1 while the highest concentration was 0.244 mg/l at S2. The concentrations of ammonia of all the samples from the different sources were within the EU (1998) and WHO (2008) acceptable limits. The very low concentrations of ammonia indicate that springs water have not been subject to pollution.

Nitrates, NO_3^- are naturally occurring. All rainfall and groundwater aquifers contain some nitrate-nitrogen. Follett *et al*. 1991 stated that low levels of nitrogen (in the form of nitrate) are normal in groundwater and surface water. However, elevated nitrate caused by human activity is a pollutant in the water. Nitrate enters ground or spring water from many sources, including nitrogen-rich geologic deposits, wild-animal wastes, precipitation, septic system drainage and fertilizer. High nitrate levels in waters to be used for drinking will render them hazardous to infants as they induce the “blue baby” syndrome (*methaemoglobinaemia*). In the present study the levels of nitrate in all the sampling sites S1, S2 and S3 were 1.85 mg/l, 3.12 mg/l and 1.96 mg/l respectively, and all these values are far below the recommended permissible limit set by EU (1998) and WHO (2008). Lower nitrate concentrations indicate that the spring waters were not affected by anthropogenic activities.

The sources of phosphates, PO_4^{3-} include natural decomposition of rocks and minerals, storm water runoff, agricultural runoff, erosion and sedimentation, atmospheric deposition, and direct input by animals/wildlife. Higher concentration of phosphate in water than 0.1 mg/L is an indication of pollution (McCutheon *et al*., 1983). Phosphates are not toxic to people or animals unless they are present in very high levels. Digestive problems could occur from extremely high levels of phosphate.

Values of phosphates for the study area were less than the detection limit of 0.02 mg/l. This is indicates that springs water are not polluted.Sulphates, SO_4^{2-} exists in nearly all natural waters, the concentrations varying according to the nature of the terrain through which they flow. Sulphates occur naturally in numerous minerals, including gypsum, barite and epsomite (Queneau & Hubert). For the present study the levels of SO_4^{2-} in all sampling sites were S1 (5. 8 mg/l), S2 (5 mg/l) and S3 (5 mg/l) respectively. Sulphate concentrations in all of the analysed samples were within the permissible limit of the EU (1998) and WHO (2008) drinking water standards. Sulphate is positively associated with consumer taste preference for drinking waters. Overall, sulphate has less perceived taste than chloride. Sulphate at high concentrations can cause diarrhea when first encountered, although eventually individuals acclimate to high sulphate levels (USEPA, 2012).

Chloride, Cl^- occurs naturally in fresh, estuarine or salt water from dissolution of rocks and minerals. In freshwater, its concentration is commonly less than 10 mg/l (van der Leeden *et al*., 1990). Chloride is normally the most dominant anion in water and it imparts salty taste to the water. The permissible limit of chloride in drinking water is 250 mg/L as given by EU (1998) and WHO (2008). In present study, the results of chlorides in all sampling sites, S1, S2 and S3 were 0.2, 0.12 and 0.1 mg/l respectively. The higher concentration of chloride was recoded at S1 (figure 4.a). The chloride level recorded in the entire sampling points of the spring water was lower than the permissible limits set by EU and WHO (2008). According to the USEPA guideline high level of Cl^- results eye/nose irritation; stomach discomfort and increase corrosive character of water. It should be noted that increasing chloride concentration is also undesirable as it can enhance corrosion of metal pipes.

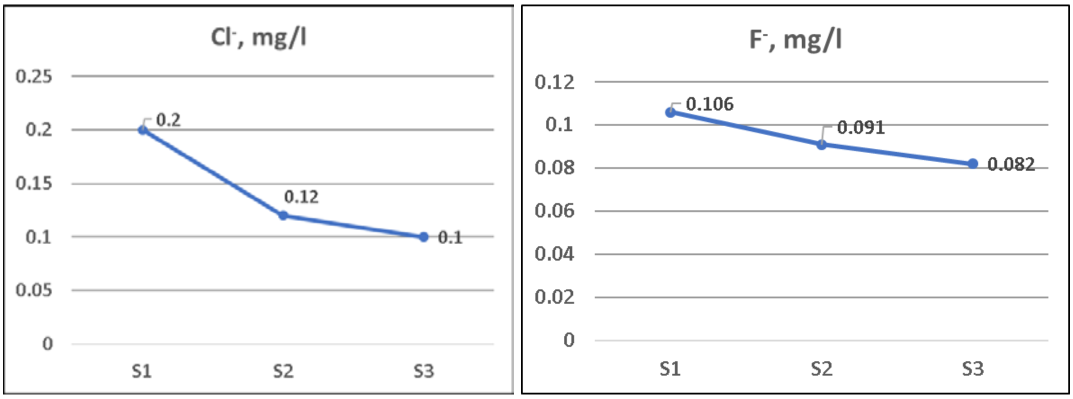


Figure 4. Variations of chloride (a) and fluoride (b) of spring waters for comparison of their quality

Fluoride, F^- is a natural element found in rocks and soil. Fluorides may occur naturally in water and unlike chlorides, they have low solubility. Fluoride is both naturally occurring in water and added to drinking water to prevent tooth decay. The fluoride concentrations in natural groundwater vary with geological composition of rocks with 0.3-0.4 mg/l typically found in water that is in contact with limestone and dolomite (DeZuane, 1997). Fluoride is an essential element of normal growth and development of humans. When fluoride levels in water are at appropriate levels, tooth decay is limited and oral health is improved. Fluoride is recognized as having a benefit effect on the development of children's teeth with 1.0 mg/L being the optimum concentration. However, concentration over 1.5 mg/L may damage children's teeth causing staining, mottling or cavities, the condition known as dental fluorosis. The measured values of F^- ion in all sampling sites S1, S2 and S3 were 0.106, 0.091and 0.082 mg/l respectively (figure 4b). The F^- concentration of all the springs samples were within the EU (1998) and WHO (2008) permissible limit for drinking water. Fluoride concentrations in all sampling sites were desirable for optimal dental health.

Conclusion

In this study the physico-chemical parameters such as temperature, pH, DO, EC, TDS, total hardness, HCO_3^-, NH_4^+, NO_3^-, PO_4^-, SO_4^{2-}, Cl^- and $F^-$of the spring's water were analyzed. The spring waters showed spatial variations among physico-chemical parameters and results were compared with EU and WHO guidelines for drinking water. The pH in the water of the springs was slightly acidic at S1 and S3. The values of temperature, EC, TDS, TH and HCO_3^-, were slightly higher at spring S3 than that at springs S1 and S2. While there were very little spatial variations in physico-chemical parameters among the springs S2 and S3. Anionic concentrations from the water samples reveal that HCO_3^- is dominant and ranges from 274 to 305 mg/l. In all samples, the order of abundance in anionic content was of the order $HCO_3^- > SO_4^{2-} > NO_3^- > Cl^- > F^-$. Low concentrations of SO_4^2 and biogenic compounds such as NO_3^- and NH_4^+ were detected, while PO_4^{3-} was not found to be present. This indicates that the spring waters were not affected by anthropogenic activities. F^- and Cl^- concentrations were also determined to be low. Fluoride concentration in all sampling sites ranged from 0.082 - 0.106 mg/l which is desirable in waters for optimal dental health. From the experimental data it was found that the values of all parameters were within the safe limits of drinking water quality set by EU and WHO standards. According to the results obtained, it can be concluded that the spring waters within the study area is of excellent quality for dinking and domestic use with the respect of physicochemical parameters.

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