WEATHERING PROCESSES EFFECTS ON THE CHEMISTRY OF THE MAIN SPRINGS OF THE YARMOUK BASIN, NORTH JORDAN

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Major cations and anions of main spring waters in the Yarmouk Basin of north Jordan were measured to assess the effects of weathering and geochemical processes in the basin area. The area has a semi-arid type of climate. A total of 36 water samples were collected from springs in 2006 to monitor the water chemistry. Chemical analysis of spring waters show that the concentration of the cations is of order $Ca>Na>Mg>K$ while that for anions is $Cl>HCO_3>NO_3>SO_4$. Statistical analyses indicate positive correlation between the following pairs of parameters: Cl and Mg (r = 0.49), Cl and K (r = 0.46), Cl and Na (r = 0.74), SO_4 and Mg (r = 0.23), and SO_4 and K (r = 0.20). Water shows varying chemical facies (Ca-SO_4, Mg-SO_4, Na-SO_4), which relates to the interaction with the geological formations of the basin (carbonates, dolomite, marl, basalt and various silicates) and evaporation. The dissolution of halite, calcite, dolomite and gypsum explains part of the observed Na, Ca, Mg, Cl, SO_4 and HCO_3, but other processes, such as cation exchange and weathering of aluminosilicates also contribute to the water composition.

INTRODUCTION

The Yarmouk Basin (hereafter denoted YB) in north Jordan is a key area on the hydrological map of the country. Few reports and papers containing geochemical data for the waters and sediments are available for the major basin springs (Hawi, 1990; Rimawi et al., 1992; Swarieh and Sahawneh, 1998; Abderahman and Awad, 2002). Unlike for rivers or open lakes, where the weathering intensity and the rate of sedimentation are significant, for springs fluctuations depend on year-to-year changes in rainfall and other climatic factors. In this investigation a detailed geochemical study of the major springs in the YB in north Jordan was carried out to identify the sources of the major ions in terms of atmospheric supply, chemical weathering of catchment lithology, soils of the drainage basin and groundwater, and to study the clay mineralogy of the sediments to understand the relationship between mineralogy and water chemistry.

GEOMORPHOLOGY AND GEOLOGY OF THE YARMOUK BASIN AND ITS CATCHMENT

The YB is located in the northwestern part of Jordan. Seventy-five percent of this basin lies in Syria. In Jordan, the basin is located between coordinates 32° 20' to 32° 45' N and longitudes 35° 42' to 36° 23' E, covering an area of about 1,426 km^2 (Figure 1).

The north Jordan area between the Zarqa and Yarmouk Rivers (Figure 1) is a key area on the hydrological map of the country. The adjacent mountains and high areas (Ajlun Mountains and Golan Heights), which are at 1,200 m above sea level, are the highest uplands in the region east of the Jordan Rift Valley (JRV). These areas receive high rainfall. In addition, the Yarmouk River flows at the borders of Syria and Jordan and delineates the northern boundary of the study area, and the Jordan River represents the western boundary (Figure 1). The Yarmouk River originates from Jabel Al-Arab (Syria) and drains from Jordanian and Syrian territories. The average discharge of this river is about 400 million cubic meters per year (Salameh and Bannayan, 1993). It contributes about 57% of the flow in the King Abdullah Canal, which supplies the JRV area with irrigation water and, in the summer, with potable water for Amman, the capital. In 2006, a major dam (Al-Wehda Dam) between Jordan and Syria was constructed on the river. It is proposed that this dam will supply Jordan with about 110 MCM/yr of potable water. Water quality of the springs, which discharge into the dam, is of great importance to the eventual usefulness of the waters to be stored.

The study area is part of the semi-arid climate of the Mediterranean Sea that has a limited amount of rainfall and high temperatures. Meteorological records collected by the Jordan Meteorological Department (JMD) at the Irbid station (north Jordan) during the years 1955 to 2007 give the mean annual rainfall and temperatures as 420 mm and 18 °C, respectively. Most of the rain falls between the months of November and April, with a peak in January (JMD 2007, personal communication).

Geologically, the rock formations of the study area are classified as the Ajlun Group, Balqa Group and Jordan Valley Group of Upper Cretaceous to Tertiary age (Makhlouf et al., 1996 and Moh'd, 2000). The oldest is the Wadi Es-Sir Limestone (WSL) formation of Turonian age belonging to the Ajlun Group, essentially composed of limestone and dolomitic limestone. In north Jordan, and locally in the basin area, the upper 100 m of the WSL formation is exposed on the southwestern part of the basin area (Figure 2). The WSL formation of the Ajlun Group is overlain by rocks of the Balqa Group and include, in ascending order: Wadi Umm Ghudran (WG),
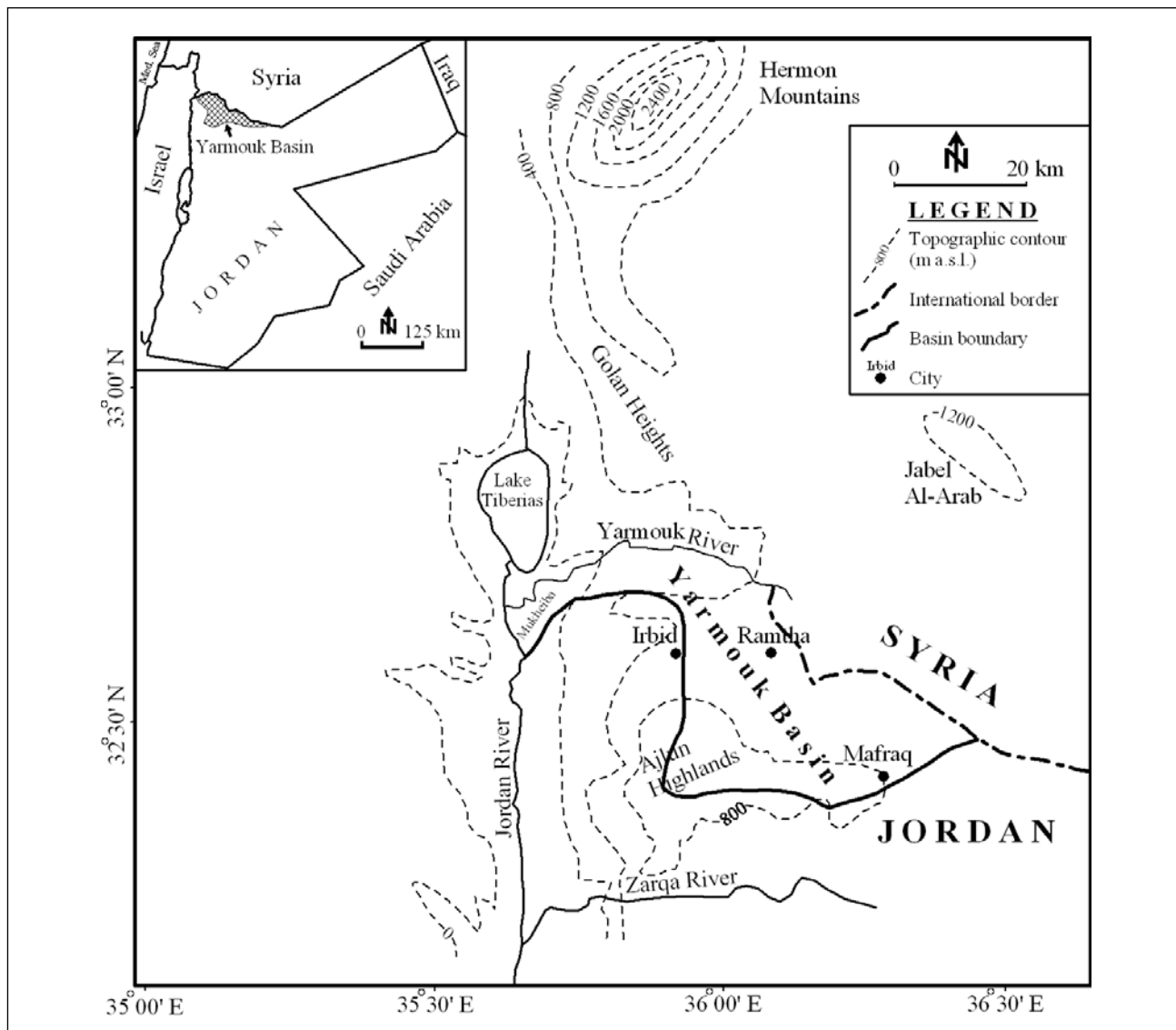
Figure 1. Location map of north Jordan showing principal physiographic features.

Amman Silicified Limestone (ASL), Muwaqqar Chalk-Marl (MCM), Umm Rijam Chert-Limestone (URC) and Wadi Shallala (WS) formations. The base of the Balqa Group, WG formation of Santonian age, comprises marl, marly limestone, chalk and chert. Up to 40 m thickness is exposed in Wadi Umm Ghudran Ed Dibab, south Irbid (Figure 2). The overlying limestone, chert, chalk and phosphorite beds that are exposed in the southern part of the basin area are members of the ASL formation (Campanian age). In the area of study this formation is about 60 m thick. Bituminous marl and clayey marl of the MCM formation of Maastrichtian age overlies the ASL formation and is exposed in the central part of the basin area. These are about 200 m thick (Parker, 1970; Makhlouf et al., 1996; Moh'd, 2000). Alternating beds of limestone, chalk and chert of the URC formation of Paleocene age overlies the MCM formation. In the basin area, the URC formation outcrops in the northern part (Figure 2). The thickness of this formation is 200 m at the outcrop. At the site of Wadi Shallala area (northeast Irbid; Figure 2), about 35 m of chalk and marly limestone with glauconite is present. These belong to the WS formation of Eocene age. In the eastern part of the basin area, basaltic flows (BS formation) of the Jordan Valley Group (Oligocene age) cover rocks of the Balqa Group. In addition, basalts were found as small exposures scattered to the south, north and northwest of Irbid (Figure 2).
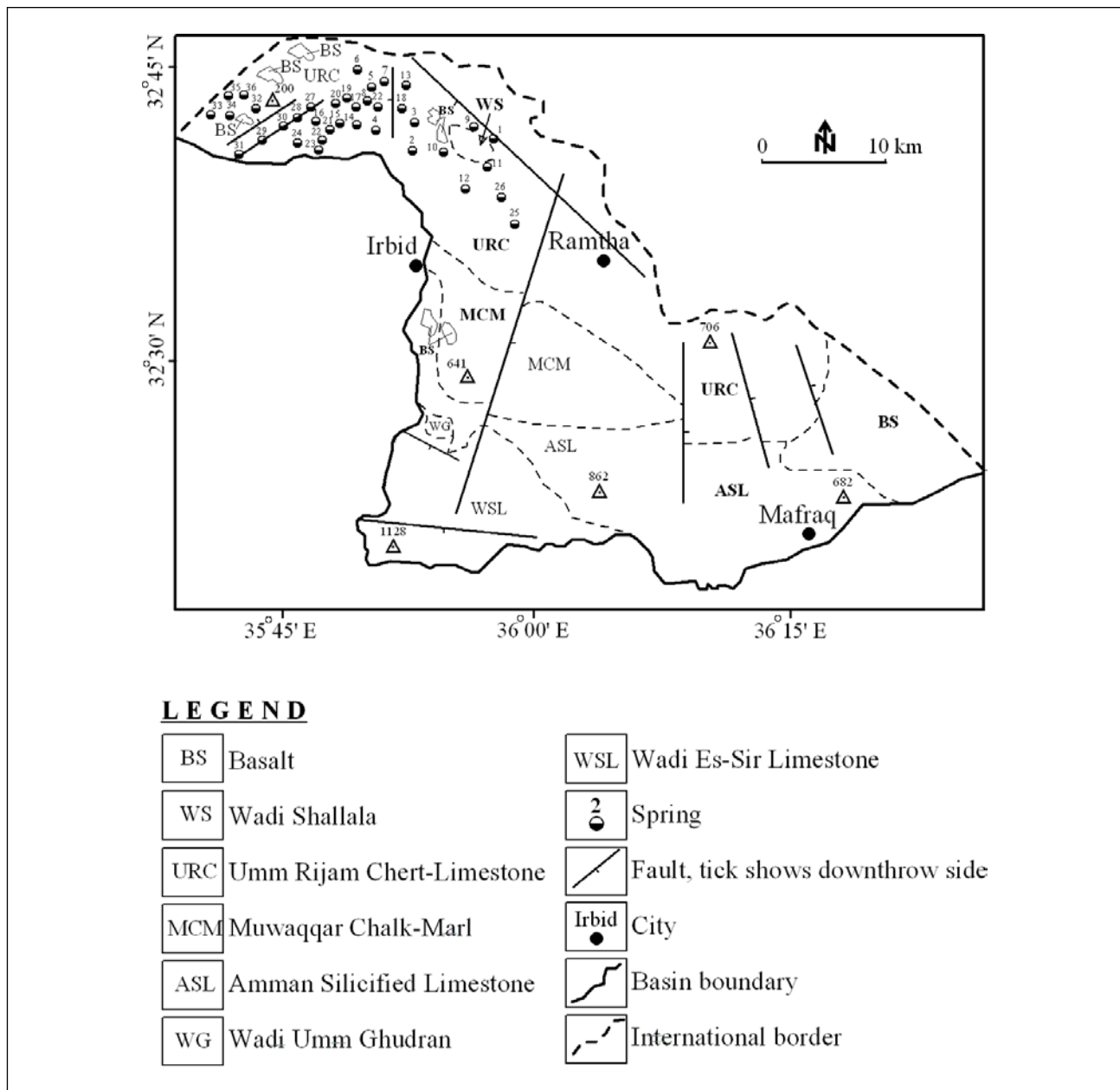
Figure 2. Spring location and generalized geologic map of the Yarmouk Basin, north Jordan.

METHODS

Water samples for chemical analysis were obtained during the year 2006 from the 36 springs shown in Figure 2. The samples were collected in polyethylene bottles; the bottles were previously washed with distilled water and dilute hydrochloric acid. After that, they were rinsed before filling to minimize sample contamination. These samples were then transported to the laboratory with care to prevent possible evaporation effects.

The hydrogen ion concentration (pH), electrical conductivity (EC, mS cm^{-1} at 25 °C) and total dissolved solids (TDS) were measured in the field using a pH-meter, portable EC-meter and TDS-meter, respectively. Chemical analysis for the major cations calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+) and potassium (K^+) was accomplished in the laboratory using atomic absorption spectrophotometry. The anions nitrate (NO_3^-) and sulfate (SO_4^{2-}) were measured by spectrophotometric techniques. Titration methods were used to determine the concentrations of

the chloride (Cl$^-$) and the bicarbonate (HCO$_3^-$); chloride (Cl$^-$) was measured by using potassium chromate (K$_2$CrO$_4$) as an indicator and silver nitrate (AgNO$_3$) as a titration solution; bicarbonate (HCO$_3^-$) was measured by titrating the samples with 0.02 (NH$_4$)$_2$SO$_4$, using methyl orange as an indicator. Sulfate was determined using a spectrophotometric method by measuring turbidity caused by adding BaCl$_2$, resulting in the precipitation of BaSO$_4$. These analyses were performed in facilities at the Department of Chemistry and at the Department of Earth and Environmental Sciences of Yarmouk University, Irbid, Jordan.

RESULTS AND DISCUSSION

Major ion chemistry

The chemical composition of the YB spring waters is given in Table 1. The observed charge balance between cations (TZ$^+$) and anions (TZ$^-$) calculated by the formula: (TZ$^+$ - TZ$^-$/TZ$^+$ + TZ$^-$ x 100) between cations and anions (< 10%) and the ratio of TDS/EC (0.5) are within acceptable limits, confirming the reliability of the analytical results. Statistical analysis of the data shows that the TZ$^+$ and TZ$^-$ are coupled by the relation TZ$^+$ (meq l^{-1}) = 0.69 TZ$^-$ (meq l^{-1}) + 1.04 with a correlation coefficient of 0.69 for 36 data points (Figure 3).

The YB spring waters are neutral to mildly alkaline. The pH for the waters ranges between 7.01 and 8.09. The electrical conductivity (EC) of the spring waters ranges between 347 and 1234 mS cm^{-1} (average 637 mS cm^{-1}). Among major cations, calcium (Ca^{2+}) is the major constituent, ranging between 1.64-2.35 meq l^{-1} with an average of 2.05 meq l^{-1}. It accounts for 54.5% of the total cations. Sodium (Na$^+$) is second in cationic abundance, accounting for 23% (0.54-1.76 meq

Table 1. Chemical composition of spring waters in the Yarmouk Basin, north Jordan.
pH hydrogen ion concentration; EC electrical conductivity; TDS total dissolved solids.
Units: meq l^{-1}, except EC (mS cm^{-1}) and TDS (mg l^{-1}).

| Spring	pH	EC	TDS	HCO$_3$	SO$_4$	Cl	NO$_3$	Ca	K	Mg	Na	TZ$^-$	TZ$^+$
Ghazzal	7.71	999	499.5	0.98	0.59	3.33	0.69	2.15	1.13	1.00	1.52	5.59	5.80
Khureibeh	7.36	630	315	1.15	0.44	1.38	0.70	2.14	0.11	0.57	0.75	3.67	3.57
Qweilbeh	7.50	480	240	1.31	0.28	1.50	0.69	1.97	0.04	0.50	0.60	3.78	3.11
Hubras	7.45	460	230	0.98	0.13	2.56	1.05	1.92	0.04	0.44	0.54	4.72	2.94
Al Rafeed	7.61	520	260	1.31	0.10	0.15	0.70	1.98	0.05	0.51	0.73	2.26	3.27
Aqraba	7.26	856	428	0.98	0.37	3.31	1.10	2.06	0.42	1.30	1.54	5.76	5.32
Um Ershid	7.59	1234	617	0.98	0.56	4.67	0.69	2.18	1.63	1.17	1.73	6.90	6.71
Yubla	7.12	501	250.5	1.31	0.42	1.44	0.70	2.06	0.01	0.50	0.57	3.87	3.14
Barrashta	7.23	530	265	1.15	0.03	1.27	0.84	2.10	0.01	0.49	0.60	3.29	3.2
Abdah	7.88	488	244	0.66	0.28	1.60	0.74	1.87	0.05	0.59	0.95	3.28	3.46
Es-Sukkar	7.59	582	291	1.48	0.41	2.34	0.69	1.99	0.05	0.70	1.12	4.92	3.86
Esh Sheha	7.43	655	327.5	0.82	0.22	2.17	1.19	1.97	0.31	0.68	1.12	4.40	4.08
Al Jamal	7.51	560	280	0.98	0.06	1.60	0.88	2.05	0.12	0.57	0.82	3.52	3.56
El Turab	7.68	456	228	1.15	0.00	1.30	0.75	1.92	0.01	0.48	0.57	3.20	2.98
El Fotaha	7.27	559	279.5	1.31	0.33	1.42	0.94	2.08	0.08	0.53	0.70	4.00	3.39
El Maghara	7.39	495	247.5	1.31	0.05	1.34	1.02	1.97	0.04	0.49	0.62	3.72	3.12
El Kufeir	7.33	469	234.5	0.82	0.12	1.64	0.72	1.95	0.06	0.45	0.59	3.30	3.05
Sama	7.39	482	241	0.82	0.01	1.66	0.93	1.97	0.02	0.47	0.67	3.42	3.13
El Baradah	8.09	467	233.5	0.82	0.32	1.49	1.56	1.82	0.01	0.74	0.81	4.19	3.38
Um Arays	7.18	594	297	1.15	0.02	1.36	1.14	2.09	0.21	0.60	0.83	3.67	3.73
El Minqa	7.33	347	173.5	1.15	0.22	1.18	0.61	1.64	0.02	0.46	0.57	3.16	2.69
El Harathin	7.41	941	470.5	1.15	0.18	2.65	2.03	2.23	1.02	0.82	1.20	6.01	5.27
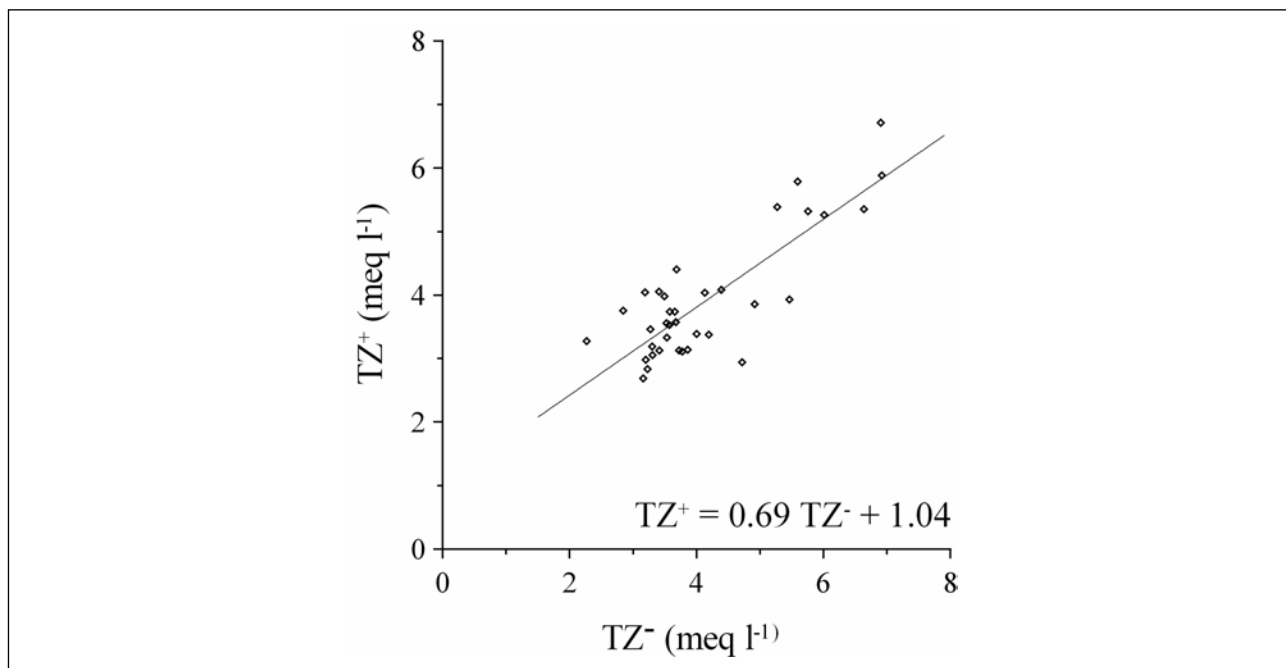
Figure 3. Sum of base cationic charge (TZ^+; meq l^{-1}) versus the sum of anionic charge (TZ^-; meq l^{-1}); TZ^+ = (Ca^{2+}) + (Na^+) + (Mg^{2+}) + (K^+); TZ^- = (Cl^-) + (HCO_3^-) + (NO_3^-) + (SO_4^{2-}).

l^{-1}) of the total cations. Magnesium (Mg^{2+}) 19.2% (0.39-1.91 meq l^{-1}) and potassium (K^+) 3.3% (0.01-1.63 meq l^{-1}) are less predominant ions in the spring waters. Among major anions, chloride (Cl^-) (0.15-4.67 meq l^{-1}) and bicarbonate (HCO_3^-) (0.66-1.48 meq l^{-1}) are generally dominant representing on average 45 and 28% of all the anions, respectively. The other anions, such as nitrate (NO_3^-) and sulfate (SO_4^{2-}) have minor contribution to the total anions. The nitrate (NO_3^-) concentration ranges between 0.14-2.03 meq l^{-1} and it contributes 21% of the total anions. Sulfate (SO_4^{2-}) concentrations in the spring waters are low, ranging between 0.0- 0.59 meq l^{-1} accounting for 5.5% of the total anions.

Source of dissolved ions

The source of dissolved materials in the spring waters seems to be primarily weathering of rocks and precipitation. Total dissolved solids (TDS) plotted on Gibbs (1970) diagram against Na/(Na + Ca) for 36 data points show that the water samples fall in the rock dominance field (Figure 4), suggesting that the weathering of rocks primarily controls the major ion chemistry of water samples.

Compositional relations among dissolved species can reveal the origin of solutes and the processes that generated the observed water composition. Statistical analyses indicate a positive correlation between some pairs of parameters shown in Figure 5 and Table 2.

The Na-Cl relationship has often been used to identify the mechanism for acquiring salinity and saline intrusions in semi-arid regions as is the case in our study area (Magaritz et al., 1981; Sami, 1992; Jalali, 2005). Figure 5b shows the value of Cl^- as a function of Na^+ in the water samples. The dissolution of halite in water releases equal concentrations of sodium and chloride into the solution, but analytical data in Figure 5b deviate from the expected 1:1 relation indicating that a large fraction of sodium is associated with an anion other than chloride. A Na/Cl molar ratio greater than one (value of 4.9 recorded at Al Rafeed spring, serial no. 5, Figure 2) is interpreted as reflecting Na^+ released from silicate weathering reactions (Meybeck, 1987). Silicate dissolution
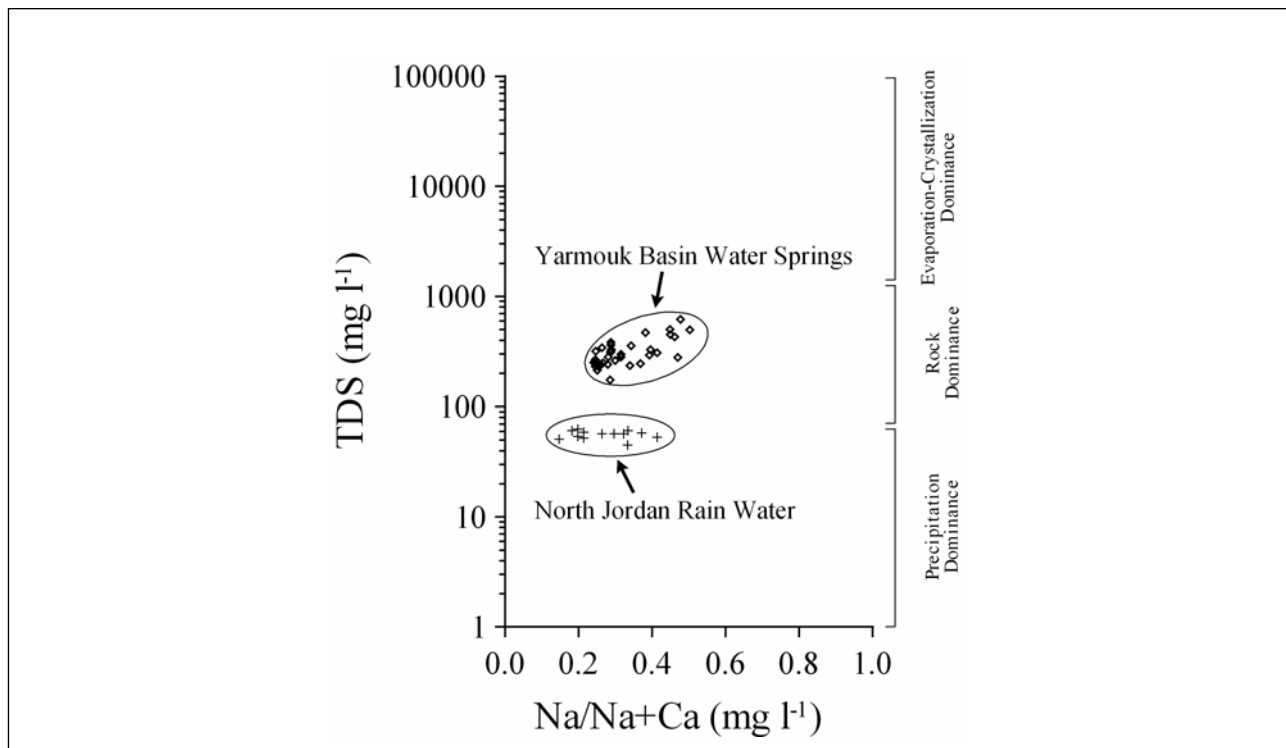
Figure 4. Plot of Yarmouk Basin spring waters data on Gibbs (1970) diagram depicting dominant factors responsible for the ion chemistry.

can be a probable source for Na^+ in the YB spring waters. One potential source of excess Na^+ is weathering of sodium plagioclase in the igneous rocks of the studied area. The excess of Na^+ in this sample may also result from dissolution of sodium sulfate minerals (Magaritz et al., 1981). The contribution of potassium to the spring waters is low (Table 1). The low levels of potassium in natural waters are a consequence of its tendency to be exchanged on to clay minerals and to participate in the formation of secondary minerals (Mathess, 1982).

Figure 5i shows the relationship between Ca^{2+} and HCO_3^- in the water samples. As can be seen, there is no relationship between these ions and the correlation coefficient is not significant (Table 2). This indicates that calcite is not the source of Ca^{2+}. Figure 5k shows that most of the data deviated from the expected 1:1 relation indicating another source of Mg^{2+} must be present, possibly from weathering. The origin of the Mg^{2+} must be associated, at least partially, with the dissolution of aluminosilicates, such as biotite and chlorite. The excess of Mg^{2+} may also result from dissolution of sodium and magnesium sulfate minerals. Figure 5j shows the value of HCO_3^- as a function of Na^+ in water samples. As can be seen, the water samples deviated from the 1:1 relation, indicating another source of Na^+.

A plot of Ca^{2+} and SO_4^{2-} (Figure 5e) shows that most of the water samples deviated from the 1:1 line and the correlation coefficient is not significant (Table 2), indicating another source of Ca^{2+}. There was a weak correlation between Na^+ and SO_4^{2-} (0.34), Mg^{2+} and SO_4^{2-} (0.23) and K^+ and SO_4^{2-} (0.20) suggesting that part of the SO_4^{2-} originates from dissolution of magnesium and potassium minerals (Figure 5f, g and h). Weak correlation between K^+ and other major ions suggests that K^+ is derived mostly from K-feldspars. If Ca^{2+}, Mg^{2+}, SO_4^{2-} and HCO_3^- are derived from simple dissolution of calcite, dolomite and gypsum, then a charge balance should exist between the cations and anions. As indicated in Figure 6, a deficiency of $(HCO_3 + SO_4)$ relative to $(Ca + Mg)$ exists in the water samples. Therefore, Ca^{2+} and Mg^{2+} must be balanced by Cl^-, the only other major anion.
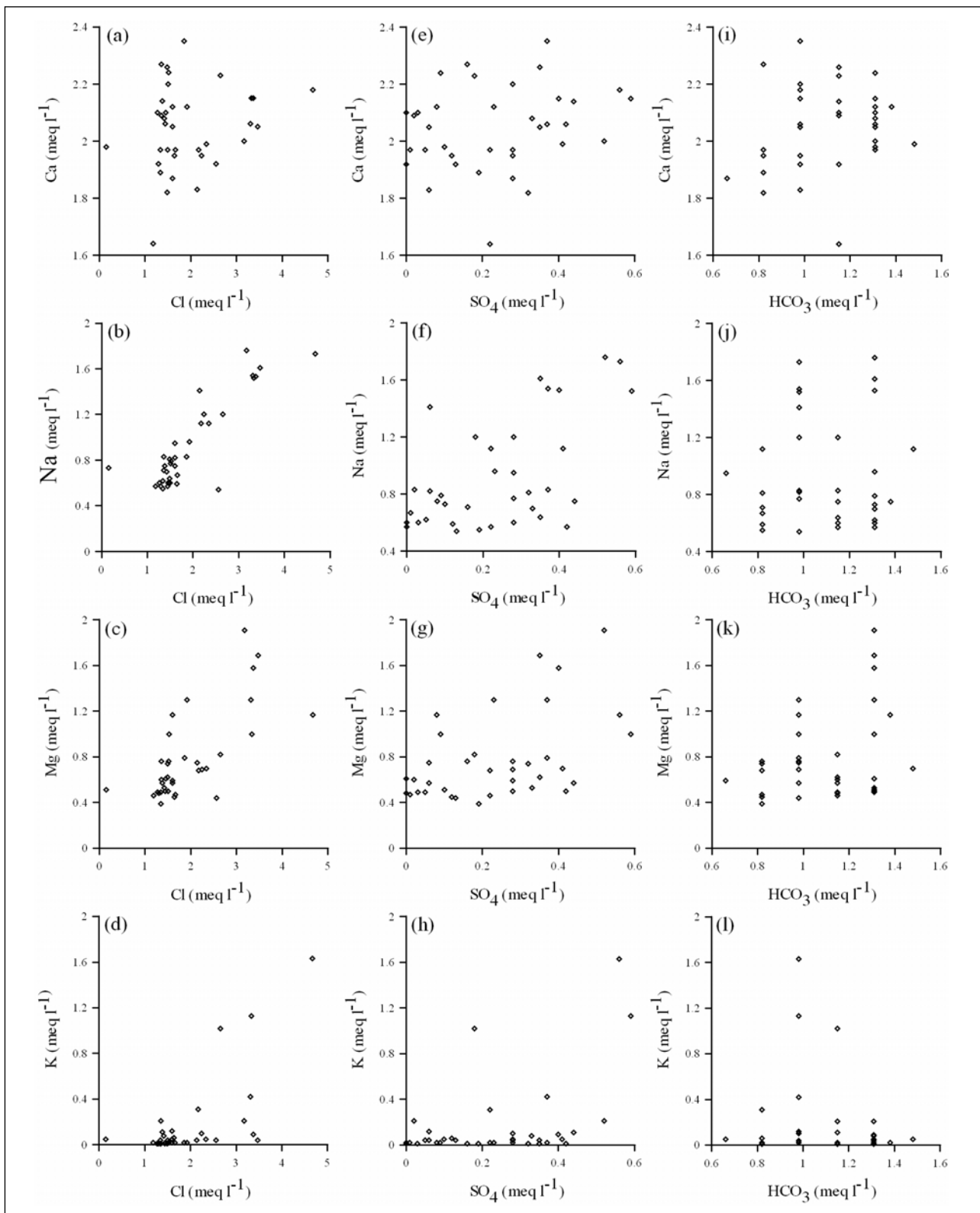
Figure 5. Relationship between different cations and anions in the Yarmouk Basin spring waters.

The evidence of silicate weathering can be derived from the relation between $Ca + Mg - HCO_3 - SO_4$ and $Na + K - Cl$. Analysis of the data shows that the $Ca + Mg - HCO_3 - SO_4$ (meq l^{-1}) and $Na + K - Cl$ (meq l^{-1}) are coupled by the relation $Ca + Mg - HCO_3 - SO_4$ (meq l^{-1}) = -0.29 $Na + K - Cl$ (meq l^{-1}) + 1.40 with a correlation coefficient of 0.12 for 36 data points (Figure 7). $Na + K -$

Table 2. Correlation matrix of the dissolved ions in the Yarmouk Basin spring waters.
Underlined coefficients are significant at the 95% significance level.

	EC	pH	Ca	Mg	Na	K	HCO$_3$	SO$_4$	Cl	TDS
EC	1.00									
pH	0.02	1.00								
Ca	<u>0.33</u>	0.11	1.00							
Mg	<u>0.61</u>	0.02	0.09	1.00						
Na	<u>0.69</u>	0.00	0.02	<u>0.64</u>	1.00					
K	0.55	0.01	0.07	0.07	0.33	1.00				
HCO$_3$	0.01	0.03	0.04	0.08	0.00	0.02	1.00			
SO$_4$	0.37	0.00	0.05	0.23	0.34	0.20	0.00	1.00		
Cl	<u>0.68</u>	0.00	0.04	<u>0.49</u>	<u>0.74</u>	<u>0.46</u>	0.00	<u>0.36</u>	1.00	
TDS	1.00	0.02	0.33	0.61	0.69	0.55	0.01	0.37	0.68	1.00

Cl represents the amount of Na + K gained or lost relative to that provided by chloride salts dissolution (mostly halite dissolution), while Ca + Mg - HCO$_3$ - SO$_4$ represents the amount of Ca^{2+} and Mg^{2+} gained or lost relative to that provided by gypsum, calcite and dolomite dissolution. In the absence of these reactions, all data should plot close to the origin (Jalali, 2005). If these processes are significant in controlling composition, the relation between these two parameters should be linear with a slope of -1. Figure 7 indicates an increase in Na + K related to a decrease in Ca + Mg or an increase in HCO$_3$ + SO$_4$ and that almost all Na$^+$, Ca^{2+} and Mg^{2+} participate in the ion exchange reaction (Garcia et al., 2001). As no water sample is saturated with halite or gypsum, Cl$^-$ and SO$_4$$^{2-}$ ions are provided by the amount of dissolved halite and gypsum, respectively. Bicarbonate ions are provided by the net amount of calcite and dolomite dissolved. As the concentration of divalent cations in solution increases due to the dissolution of gypsum, calcite
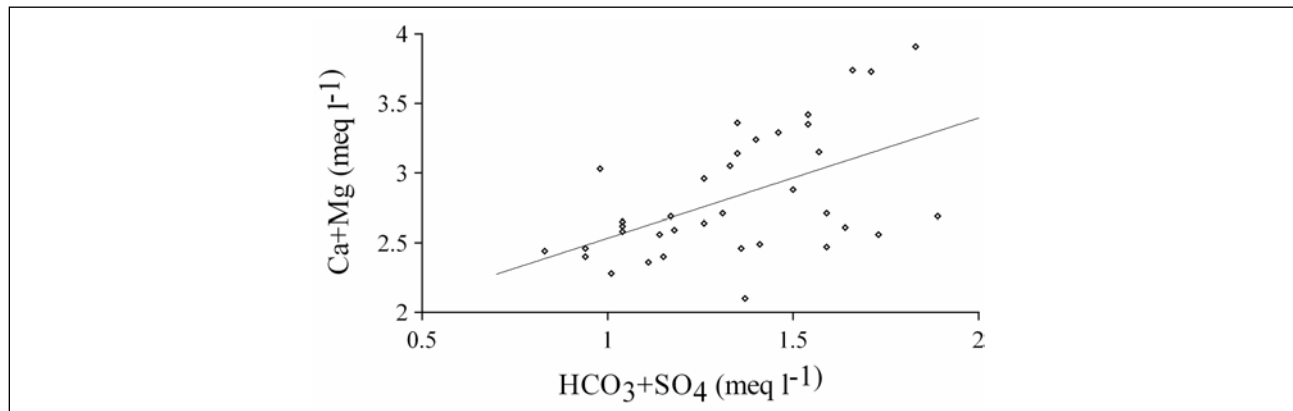

Figure 6. Relationship between Ca + Mg and HCO$_3$ + SO$_4$ in the Yarmouk Basin spring waters.
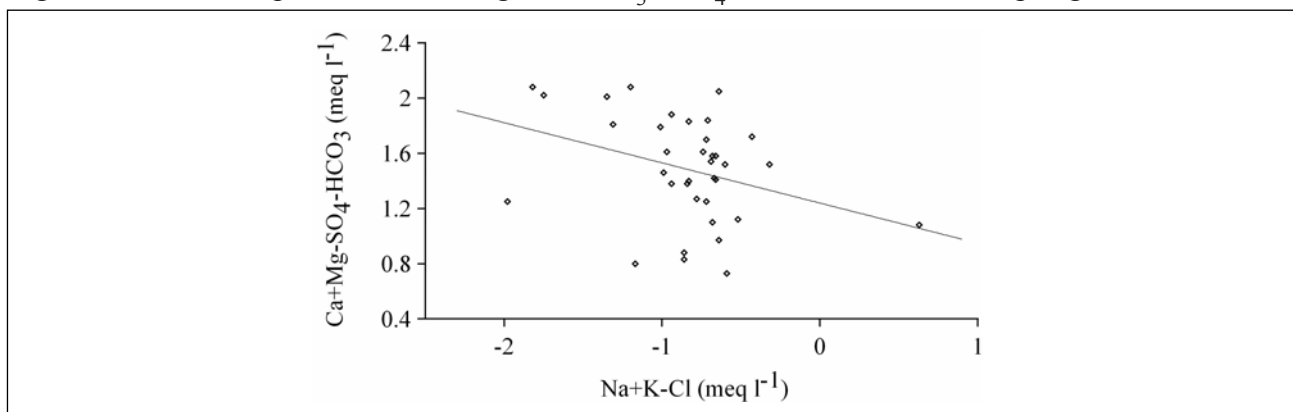

Figure 7. Relationship between Ca + Mg- SO$_4$-HCO$_3$ and Na + K-Cl in the Yarmouk Basin spring waters.
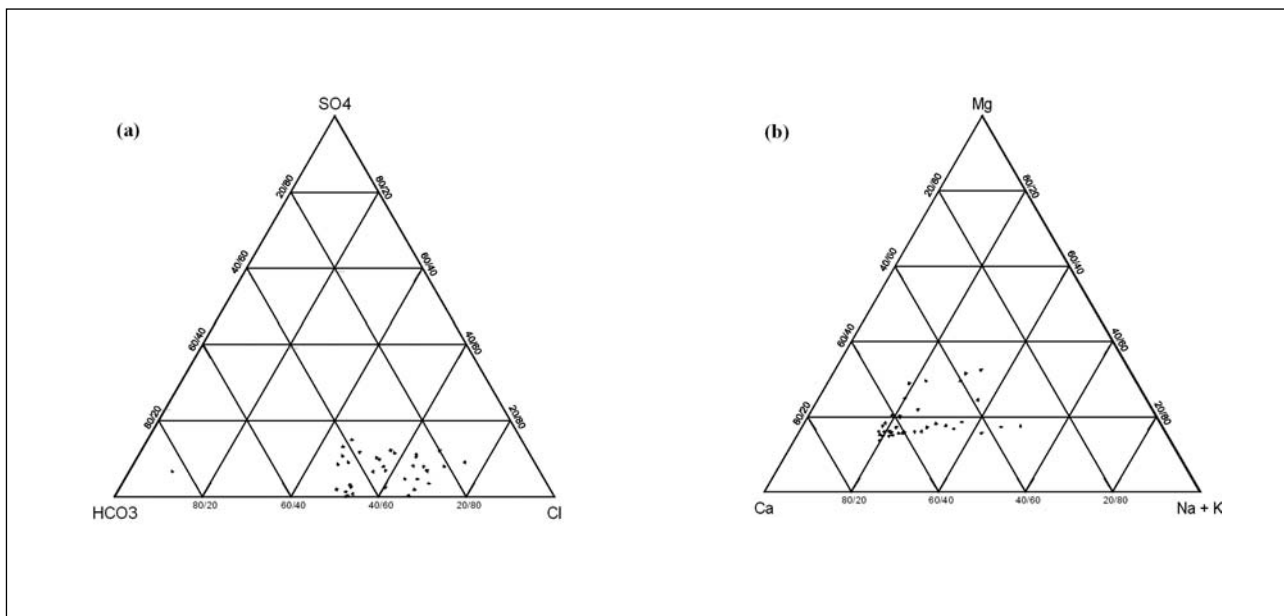
Figure 8. (a) Ternary anion diagram showing the relationship between Cl^-, HCO_3^- and SO_4^{2-} (meq l^{-1}) and (b) ternary cation diagram showing the relationship between $(Na + K)$, Ca^{2+} and Mg^{2+} (meq l^{-1}).

and dolomite, cation exchange occurs between solution and adsorbed cations. With increasing cation exchange driven mostly by gypsum dissolution and recorded by increasing SO_4^{2-} concentrations, Na^+ ions are desorbed from exchange sites, resulting in the increase of aqueous Na^+ concentrations. Therefore, it is suggested that dissolution of halite and gypsum and cation exchange are the primary controls on solute concentrations in YB spring waters.

On a ternary diagram (Figure 8), constructed using RockWare Scientific Software relating HCO_3^-, SO_4^{2-} and Cl^- anions (Figure 8a), most of the data cluster towards the chlorinity apex with secondary trends towards HCO_3^-. A similar cation diagram relating Ca^{2+}, Mg^{2+} and $(Na + K)$ cations shows that most of the data fall on the left corner of the triangle with a trend from the Ca^{2+} apex towards the center of the field (Figure 8b). These arrays indicate the presence of calcite, dolomite, gypsum and volcanic rocks weathering in the present drainage basin.

CONCLUSIONS

The hydrochemistry of the YB spring waters varies in relation to different water-rock interactions (presence of calcite, dolomite, gypsum and volcanic rocks) and evaporation. The chemical analysis of water samples showed three main types of water are present within the study area. These are $Ca-SO_4$, $Mg-SO_4$ and $Na-SO_4$. The dissolution of minerals, such as halite, gypsum, dolomite, calcite in the sediments results in the Ca^{2+}, Na^+, Mg^{2+}, Cl^-, and SO_4^{2-} content in the spring waters. Other reactions such as cation exchange and the weathering of aluminosilicates, also contribute to the content of Na^+ and K^+.

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