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The Dead Sea And Geo-Environmental Trace Influence on The Rainwater Quality In Jordan

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Abstract *- In order to investigate the major factors that affect rainwater quality in Jordan, 28 rainwater samples were collected from seven stations distributed in different eco-zones in the country. Major ions concentration (Ca^{2+}, Na^{+}, Mg^{2+}, SO_4^{2-}, K^{+}, Cl^{-}, NO_3^{-}, HCO_3^{-}, NH_4^{+}) were determined for each sample along with measurement of pH, TDS, and EC. The results show that rainwater chemistry varies from one location to another. The average pH values range from 7.25 to 6.58, and the average TDS range between 10 to 59ppm. The variability of rainwater chemistry was an attribute of regional conditions and anthropogenic effects. However, in the case of Ghor Safi station, the influence of the Dead sea was highly notable on the rainwater quality. The most anthropogenic activity that affects the rainwater quality were the traffic activity emissions and combustion fuel. In the case of Qatraneh station, the cement industry and phosphate mining are among the possible anthropogenic activities. The ionic balances in the rainwater samples for all stations were close to one, which indicates that the analyzed ions were sufficient to depict the rainwater quality in the selected stations*.

Keywords — *Anthropogenic contamination, Dead Sea influence, Jordan, Rainwater chemistry, Rainwater quality*

I. INTRODUCTION

Jordan is a poor country in water resources, where the per capita share of water is less than 100 m / year. Water sources suffer constant pressure, especially with the large increases in the number of inhabitants, whether by natural increase or through asylum from neighboring countries. This pressure on water sources and as a result of the insufficiency of the available resources to meet the requirements, the groundwater sources have often been depleted to the point that most basins suffer from low levels [1]. Also, sometimes, some degradation in the quality of the groundwater was recorded [2]. As for surface water, it is exposed to pollution to a greater degree than groundwater, where degrees of pollution have been recorded in different places during several studies[3]. These difficult water conditions led to think of new resources such as reuse and desalination of brackish water and saltwater.

Rainwater is the main source for all other sources in Jordan, so it is given great attention, as it is noted that there are a large number of studies on the quality of rainwater, the historical pattern and predicted amounts f rainfall, the precipitation distribution, and the process of collecting and storing rainwater [4],[5],[6][7].

As for the precipitation quantities and its distribution, it is generally limited, as less than 1% of the area of Jordan receives precipitations more than 500 mm/year, and about 5.5% of the area of the country receives between 200 mm and 500 mm. More than 71 % of the country's area is a desert and receives less than 100 mm/ year [8]. The annual amount of precipitation ranges between 3 to 17 billion cubic meters, with a long-term annual average of about 8.2 billion cubic meters [9].

This study aims to investigate the quality of rainwater in several regions of Jordan and determine the regional and local factors that influence the quality of rainwater, with special attention to the impact of the Dead Sea and anthropogenic effects on the rainwater quality.

II. STUDY AREA AND METHODOLOGY

Seven sampling stations distributed in different regions of the country have been selected. These stations represent an area where more than 70% of the total population of Jordan lives (Fig. 1). From each station, 4 rainwater samples were taken during the winter season of 2017. Each sample has been collected from a rainfall event greater than 5mm. The sampling was performed manually. The analysis was based on wet chemistry for Total Dissolved Solids (TDS), the potential of Hydrogen (pH), and Electrical Conductivity (EC). The major anions (Cl^{-}, NO_3^{-}, and SO_4^{2-}) were determined by ion chromatography, NH_4^{+} by spectrometry, and the Major cations (Na^{+}, K^{+}, Ca^{2+}, and Mg^{2+}) were measured by flame atomic absorption spectrometry.

III. RESULTS AND DISCUSSION

The rainwater quality in the study area is determined through the average of the four measurements for each factor and each location.Table 1 presents the averagevalues of

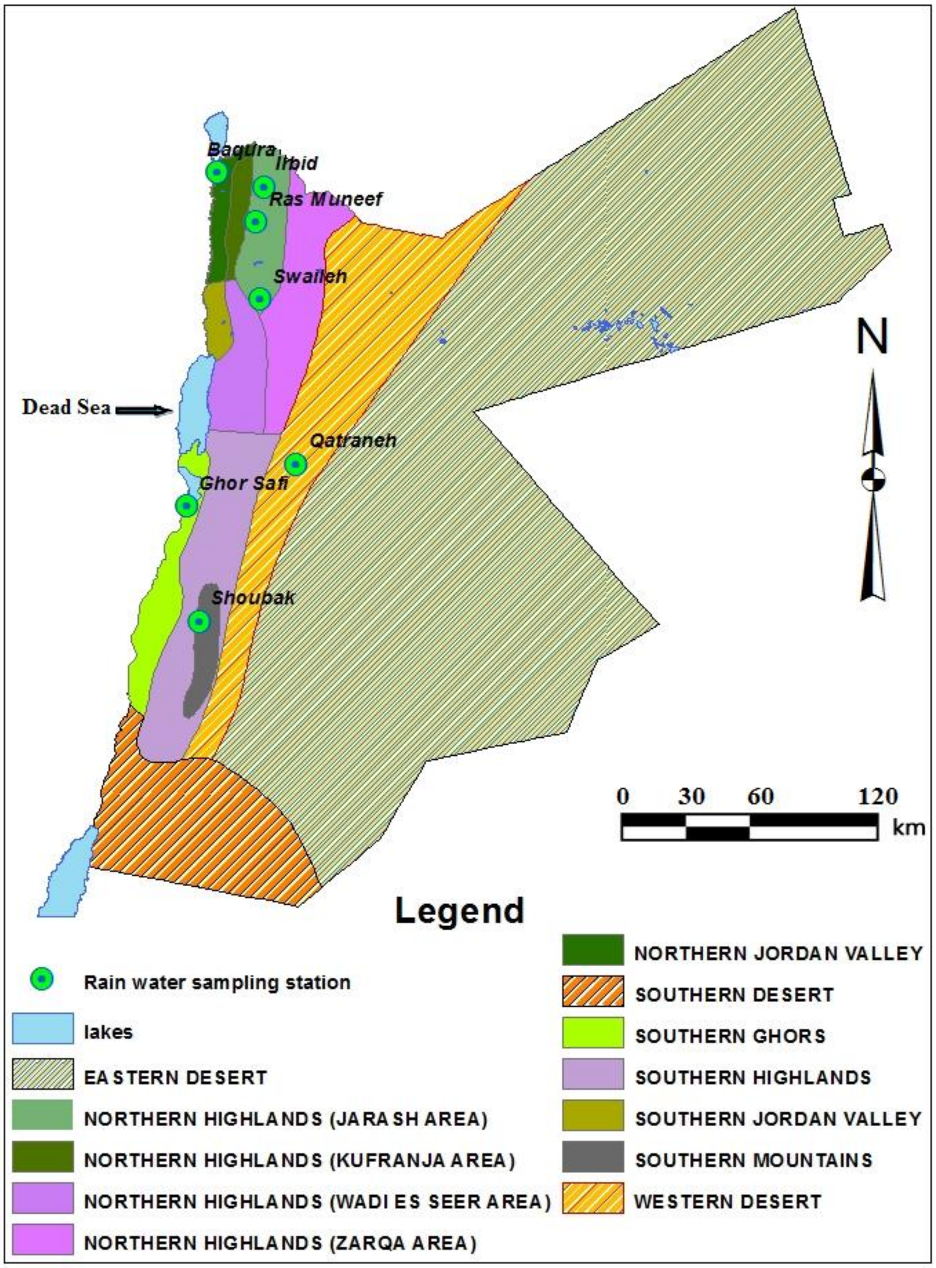

Fig. 1. Location map of the seven sampling sites.

Table1. Average chemical composition of the rainwater samples in the study area

Station	pH	TDS (ppm)	Ec (μs/cm)	HCO_3^- μeq. /l	Cl^- μeq. /l	Ca^{2+} μeq. /l	Mg^{2+} μeq./l	N^{a+} μeq./l	K^+ μeq./l	NO_3^- μeq./l	SO_4^{2-} μeq. /l	NH_4^+ μeq./l
Ras Muneef	7.22	30.31	60.62	94.15	44.46	87.23	51.46	11.71	3.85	3.27	13.19	1.40
Swelieh	6.80	40.19	80.38	128.23	89.85	138.31	83.33	17.85	7.25	6.62	23.77	2.81
Qatraneh	6.58	45.85	91.69	82.62	76.85	106.62	52.00	10.46	4.55	8.62	16.31	1.80
Irbid Av	6.63	39.50	79.00	92.31	46.62	71.38	75.15	7.69	3.48	7.77	17.92	1.44
Baqura	7.16	31.58	63.15	64.03	44.62	67.23	37.02	29.12	4.29	8.70	28.77	3.37
Ghore Safi	6.83	59.27	118.55	82.73	109.09	124.82	95.11	94.55	32.18	2.75	108.18	4.24
Shoubak	7.25	10.50	21.00	21.23	18.41	30.24	14.23	2.07	4.31	2.30	9.03	1.70

pH, EC, and Major ions in the rainwater samples for each station.

The acidity of rainwater is a parameter that is associated with the potential of Hydrogen (pH). The pH of rainwater in a pure atmosphere is around 5.6, produced by the dissolution of CO_2 in the raindrops forming the carbonic acid H_2CO_3 (10). Therefore, pH 5.6 is taken as a quality reference, classifying the rainwater with pH values lower and higher than 5.6 as acid and alkaline rains, respectively (11). The registered pH values presented minimum and maximum values of 6.576 (in Qatraneh) and 7.25 (in Shoubak), respectively.

In general, all the stations were alkaline, showing pH values above the reference level 5.6, with an average value for the seven stations of 6, 92. It was observed that the highest pH values correspond to stations located in higher elevation zones (Ras Muneef pH = 7.22; and Shoubak pH 7.25) where the air is mainly influenced by the regional and local natural loads such as windblown dust. These two stations are also characterized by negligible anthropogenic contaminant loads and no obvious influence of marine currents. Whereas the lowest pH values correspond to stations where there is a greater presence of combustion gases (Swaileh pH = 6.79; Qatraneh pH = 6.576 and Irbid pH = 6.626). The results obtained revealed that low pH is associated with high concentrations of SO_4^{2-}, NO_3^-, Cl^- anions and organic acids, which cannot be completely neutralized with the concentration of cations present in the environment (Na^+, K^+, Ca^{2+}, Mg^{2+}, NH_4^+).

The pH and therefore the acidic or basic character of the rainwater achieved depend on the abundance and type of ions present in the neutralization reactions that occur between them. This analysis is in agreement with those discussed by Kashman 2009 [12], Migliavacca 2005 [13], and Tuncer et al. 2001 [14] for rainwater quality in western Jordan, urban areas of Brazil, and Turkey, respectively. The values obtained from the ion concentration lead to think that among the possible sources of acid anions are industrial emissions, combustion products of vehicular traffic and fossil fuels, or certain polluting activities carried out by a man that produce SOx, NOx, CO_2 gases. This analysis is in agreement with those discussed by Byron et al. 1991 [15]; Kashman, 2009 [12]; and Momani 2003 [16] in rains of Sierra Nevada, West Jordan, and Mediterranean basin, respectively.

The dissolution of the combustion gases in the rainwater results in a mixture of constituents product of their oxidation that in the presence of water gives the acid character. In the case of SO_x (sulfur oxide compounds, predominantly SO_2), it transforms into SO_3^{2-} (sulfite), HSO_3^- (bisulfate), and H_2SO_3 (undissociated sulfurous acid), depending on the pH. At pH values around 5.6 (Reference level), the bisulfate ion is the dominant ion (equation 1). Subsequently, in the atmosphere, various oxidation mechanisms of bisulfate to sulfuric acid occur in the aqueous phase, in which ozone O_3 from clouds and hydrogen peroxide act as oxidants, producing reactions 2 and 3 (eq. 2 & eq. 3) that generate the quantified anion SO_4^{2-} in rainwater (Harrison 2003).

$$SO_2 + H_2O \rightarrow H^+ + HSO_3^- \qquad \text{eq. 1}$$
$$O_3 + HSO_3^- \rightarrow H^+ + SO_4^{2-} + O_2 \qquad \text{eq.2}$$
$$H_2O_2 + HSO_3^- \rightarrow H^+ + SO_4^{2-} + H_2O \qquad \text{eq.3}$$

The concentration of nitrogen oxides NO_x, where x means the formation of NO and NO_2, predominantly the concentration of NO, are formed in the cylinders of internal combustion engines by a direct combination of Nitrogen and Oxygen (eq. 4& q. 5). The high concentrations of NO_x products of the combustion of fuels are absorbed by the water producing nitric and nitrous acid, contributing more acid character to the rain (eq. 6 & eq. 7). In table 1, it's obvious that the station of Suileh, Irbid, and Qatraneh register high NO_3 concentration together with minimum values of pH where the air contamination by vehicle emission is a direct justification of the low pH values and the relative high NO_3 contents. Although Baqurah station registers the highest NO_3 concentration, as it's located in the agriculture zone, the N- fertilizers are mainly the source of NO_3 in rainwater. It should be noted that the electrical discharges that occur in the atmosphere also contribute to the presence of NO_3^- in rainwater. This energy promotes the oxidation reaction between the inert nitrogen in the air and the O_2 and O_3 in the atmosphere, generating the oxides NO and NO_2^-, which in subsequent contact with water produces the anionic acid NO_3^- (eqs. 4, 5, 6, 7).

$$N_2(g) + O_2(g) \rightarrow 2\,NO\,(g) \qquad \text{eq.4}$$
$$3NO + (3/2)O_2 \rightarrow 3NO_2 \qquad \text{eq.5}$$
$$OH + NO_2 \rightarrow HNO_3 \qquad \text{eq. 6}$$
$$NO + NO_2 + H_2O \rightarrow 2HNO_2 \qquad \text{eq. 7}$$

High levels of carbon dioxide CO_2 are one of the most common pollutants in areas with a massive transport effect [17]. The presence of CO_2 influences the pH of rainwater in stations of similar conditions such as Swaileh and Irbid. . CO2 reacts with water generating the intermittent unstable carbonic acid with sodium bicarbonate, but later, together with traces of hydrochloric acid produced by the chloride anion Cl- in an aqueous medium, contribute to decrease the pH, as illustrated in the equations 8 to 11.

$$CO_2 + H_2O \rightarrow H_2CO_3 \qquad \text{eq. 8}$$
$$H_2CO_3 \rightarrow HCO_3- + H^+ \qquad \text{eq. 9}$$
$$HCO_3^- \rightarrow CO_3^{2-} + H^+ \qquad \text{eq. 10}$$

$$Cl^- + H^+(OH)^- \rightarrow HCl + OH^- \qquad \text{eq. 11}$$

Also, the minimum average pH values recorded in the Qatraneh station are partially due to the additional effect of the amounts of SO_4^{2-} and NO_3^- from gypsum dust of cement industries and N-fertilizer of agricultural land close to this station; transported by air masses and incorporated into raindrops. A similar mechanism is repeated in Swaileh station, located in the vicinity of a cement factory.

Among the reported values (table 1), it was found that Ghor Safi station registered the same low pH level as Swaileh (pH = 6.8), a consequence of the high concentration of Cl^-

present in the rainwater. The registered concentrations of Na^+ are associated to a large extent with Cl^-. When the Pearson correlation coefficient (R^2) is close to one, this demonstrates their same origin, which is normally the seawater. In the case of Ghor Safi station, the Pearson correlation coefficient is 0.95, which indicates the marine precedence represented by the Dead Sea and the potash industry lakes derived from the same seawater. Furthermore, the value of Cl/Na for rainwater of Ghor Safi station is 1.15, which identical to seawater [13]. This confirms the effect of the Dead sea on the rainwater quality of the Ghor Safi area. The Cl /Na value obtained in this study is very close to the value obtained by Kashman 2009 (1.09), and he had a similar interpretation for this value [12]. The proximity of Ghor Safi to the Dead Sea and to Potash industry lakes leads to deduce a clear influence of them on the pH, rather than anthropogenic sources. Furthermore, the high values of the basic cations Ca^{2+}, Mg^{2+}, K^+, and Na^+ (124.8, 95.1, 32.2, and 94.5 µeq/l respectively) registered in this station, confirm the effect of the Dead Sea on the quality of rainfall water in Ghor Safi area. For the other stations, R^2 was around 0.76 for Na^+ versus Cl^- (fig. 2); this indicates the partial marine effect on rainwater of all these stations. The relative abundance of Cl^- suggests that the contribution of the anion from marine currents is not the only source, but there are additional sources that are more likely to be anthropogenic.

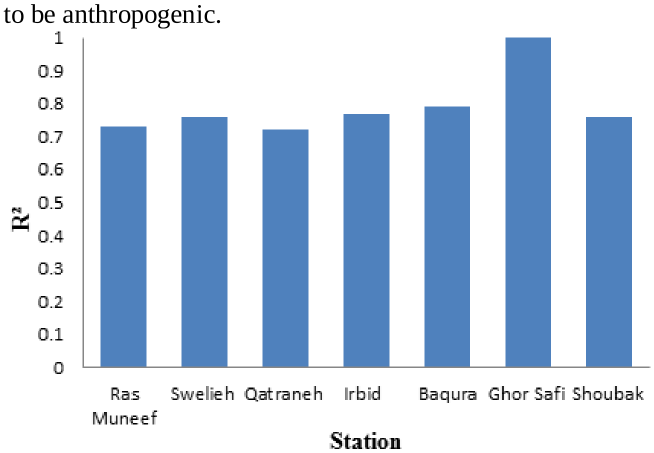

Fig. 2. Pearson correlation coefficients for Cl^- versus Na^+ in rainwater.

The Baqura and Ghor Safi stations, both characteristically agricultural areas located in the Jordan rift valley, presented the highest concentrations of $NH4^+$ and NO_3^- (Table 1). The good Pearson correlation coefficients between these ions (fig. 3 & fig. 4) found in both satiations (Baqura R^2 = 0.68 and R^2 in Ghor Safi = 0.53) suggest that a large part of them come from the same source, which is the fertilizers applied to agricultural soils. The fertilizers contain $(NH_4)_2SO_4$ and $(NH_4)NO_3$, which when applied to the soil, part of these components become NH_3, escaping into the atmosphere and reacts in an aqueous medium to form NH_4^+ (eq.12), which play a role

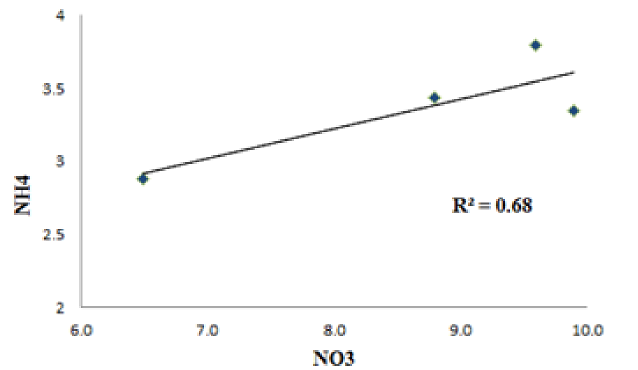

Fig. 3. Linear regression of the NH_4^+ and NO_3^- in Baqura rainwater samples.

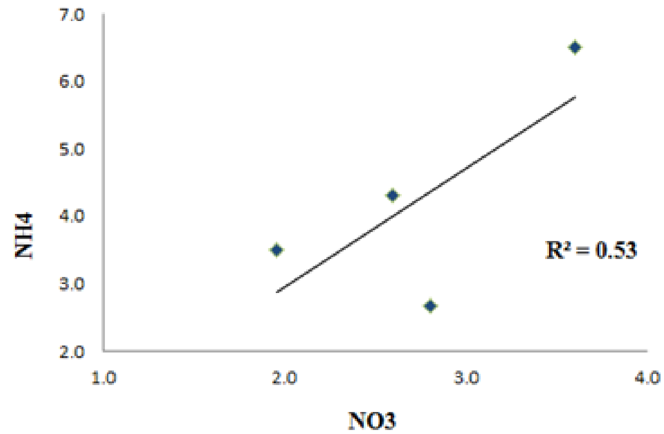

Fig.4. Linear regression of the NH_4^+ and NO_3^- in Ghor Safi rainwater samples.

in neutralizing the acidity of rainwater [18], [19], [20]. Part of these fertilizers can be transported by air that would also introduce SO_4^{2-} and NO_3^- anions into raindrops. This analysis is in agreement with those presented by Kaya and Tuncel 1997 [20] and Al- Momani 2003 [16] for ion traces in rainwater from Turkia and Jordan, respectively. In Ghor Safi station, the maximum K^+ concentration value is remarkably high compared to the average in the other stations (Table 1), thus confirming once again the Dead Sea effect that enriches the adjacent atmosphere with high concentrations of chloride and alkaline ions (Na^+, K^+, Mg^{2+}, ..etc.).

$$NH_3 + H_2O \rightarrow NH_4^+ + OH^- \qquad \text{eq.12}$$

The high Ca^{2+} values found in Swaileh and Qatraneh are strongly affected by the alkaline minerals of the desert dust and probably the emissions from cement factories close to both stations.

The results recorded for the bicarbonate anion for the Sweileh, Qatraneh, and Irbid stations were the highest; (128, 82, and 92 µeq./liter respectively). That attributed to high vehicular flow, which, together with the high population and industrial density, generate notable levels of CO_2. The reaction of CO_2 with water generates unstable carbonic acid (H_2CO_3), which is one of the major sources of bicarbonate anion (HCO_3). This anion originates when the H_2CO_3 acid dissociates into hydrogen ions (H^+) and bicarbonate anion (HCO_3^-). Therefore, the presence of a large number of anthropogenic sources that generate emission gases and with them anions of combustion (NOx, CO_2, SOx, etc.) are directly related to the increase in HCO_3^-.

Total dissolved solids (TDS) is a measure of the ions in solution. In the study area, Qatraneh and Ghor Safi registered the highest TDS contents in their rainwaters (table 1). This leads to conclude that dissolved ions concentrations in rainwater increase if they are found in desert areas such as Qatraneh or areas near the Deadsea such as Ghor Safi. In desert areas, there is a transported dust in the environment that originated from the erosion of nearby or distant soils and even from cement factories and phosphate mines located in the area. At Goor Safi, the abundance of cations is a direct consequence of the Dead Sea effects.

The EC versus TDS graph (fig. 5) shows an excellent correlation index with R^2 close to one, which confirms that the areas with the high presence of dissolved ions have high electrical conductivity and vice versa. These results are in good agreement with those presented by Migliavacca et al. 2005 for rainwater in Brazil [13] and Galeb et al. 2010 for rainwater in Ramallah [21].

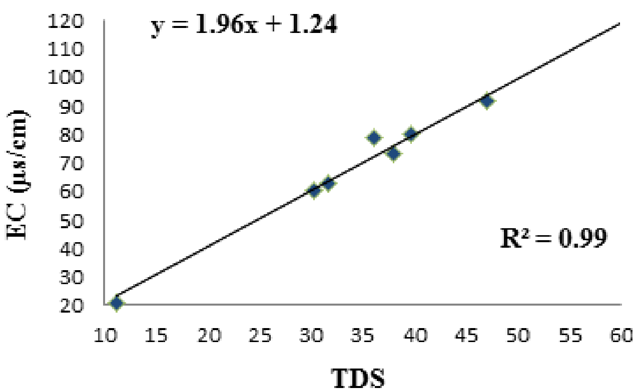

Fig.5. Linear regression of the average EC and TDS values in rainwater samples of the seven stations.

The ionic balances in the rainwater samples from each station were calculated by making the summation of cations and anions concentrations in µeq./liter. According to the neutrality condition, the sum of the anions must be equal to that of the cations [22]. If all the important anions and cations are measured during the chemical analysis, the quotient obtained by dividing the sum of cations by the sum of the anions (Σcations / Σanions) should be equal to one [23]. In the actual study, the registered ratios ranged between 0.95 and 1.159 (fig.6). This indicates that most of the ions present in the samples were measured, corroborated by the correlation index R^2 close to one (fig.7).

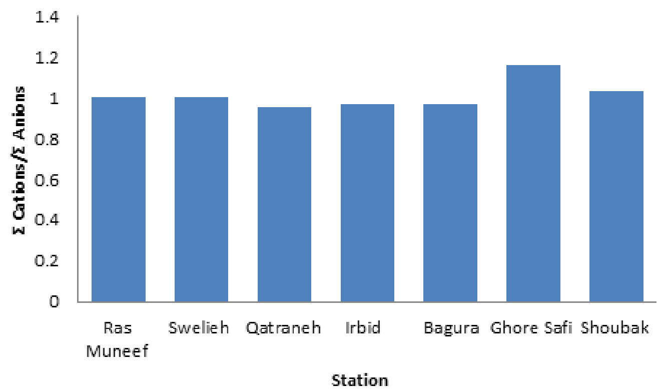

Fig.6. Histogram of Summation cations - anions for the seven stations

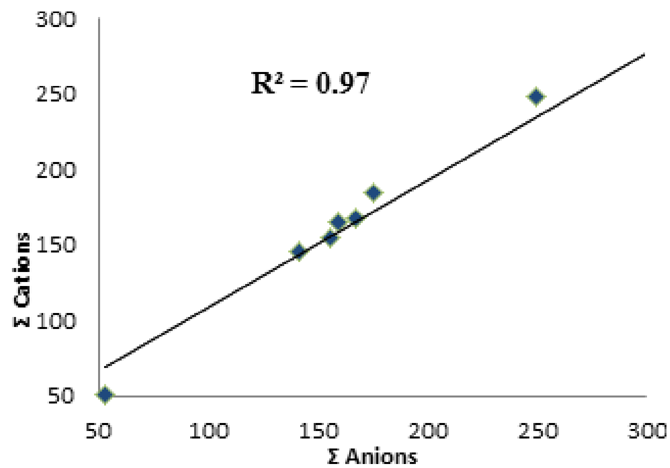

Fig.7. Linear regression of the anions and cations sum.

Shoubak and Ras Muneef have a relatively high pH, 7.22 and 7.25, respectively. Both stations are located in mountainous areas. The elevation of Shoubak station is 1300 m, and that of the Ras Muneef station is 1218m. Both stations are far from sources of contamination, such as massive traffic roads and large industries. The distance between the two stations is more than 280 km. This means that the registered pH in those stations is almost the regional influenced value in the majority of Jordan's geography. This interpretation coincides with the results of Ganem et al. 2010 in Ramallah district [21] and is in agreement with mane 2007 of the pH results in southern Israel [24]. This indicates the presence of local sources of constituents that result in lower pH when it is much less than that of Shoubak or Ras Muneef rainwaters.

IV. CONCLUSIONS

The results of the present study led to the conclusion that the stations with high pH values in rainwater were located in areas far from the Dead Sea or the main sources of pollution such as main roads and large industries. The highest pH values were recorded at Shoubak and Ras Muneef stations; these values were 7.25 and 7.22 for the two stations, respectively. It should be noted that Shoubak and Ras Muneef stations are the highest elevations among the seven stations (1365 and 1150 m asl. respectively).

The impact of the Dead Sea was evident at the closest station, which is Ghor Safi, as well as the impact of the Dead Sea at the Baqura station, was clear, although to a lesser extent. Baqurah is located in the same Jordanian Rift valley, which includes the Dead Sea and Jordan valley.

All stations show a relative abundance of constituents that come from soil dust and marine sources, and this represents the regional background. Local contributions were represented by the provision of anions that reduce pH value through neutralization of the regional effects.

The anthropogenic impact was evident at Irbid, Swaileh, and Qatraneh stations. At these stations, the most obvious effect was combustion fuel. At Qatraneh station, the mining activities and cement industries contribute to reducing the pH values. In Baqura and Ghor Safi, the effect of agricultural activity was evident in rainwater, as NO_3 is closely correlated to NH_4, which most probably indicates fertilizers precedence.

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