

## Stable Isotope Ratios in Meteoric Waters in El Kef Region, Northwestern Tunisia: Implications for Changes of Moisture Sources

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### Abstract

Natural oxygen and hydrogen stable isotopes ( $\delta^{18}\text{O}\text{-H}_2\text{O}$  and  $\delta\text{D}\text{-H}_2\text{O}$ ) in modern precipitation collected in five months event in 2001 in Northwestern Tunisia (El Kef area) are presented in this paper. The total correlation between  $\delta^{18}\text{O}$  and  $\delta\text{D}$  is obtained as the Global Meteoric Water Line and the Local Meteoric Water Line of the Tunis-Carthage. Seasonal variations of the precipitation D-excess provide more details for changes in moisture sources. The lower  $\delta\text{D}$  and  $\delta^{18}\text{O}$  values observed at the El Kef site reflect the combined effects of oceanic and sea vapor sources "Atlantic-Mediterranean origin", fractionation due to local precipitation, and slower equilibration of the larger raindrops nucleated by a maritime aerosol.

**Keywords** Stable isotopes; Precipitation; Vapor sources; Northwestern Tunisia; El kef area

### Introduction

The stable isotopes  $^{18}\text{O}(\text{H}_2\text{O})$  and  $^2\text{H}(\text{H}_2\text{O})$  have many applications in hydrogeological investigations, meteorological, hydrologic, ecological and paleoclimatic studies [1-5]. Due to their high abundance and the simplicity of their analytical determination, spatial and temporal variations in the  $^{18}\text{O}$  and  $^2\text{H}$  contents of precipitation are caused by the isotope fractionation effect accompanying evaporation from the ocean and condensation during the atmospheric transport of water vapor [6,7]. Isotope fractionation is a thermo-dependent reaction, as proved by the existence of a correlation between  $\delta^{18}\text{O}$  in rainwater and environmental temperature [6,8]. Thus, more the temperature is higher; more the heavy isotope is higher. The existence of thermo-dependence in the fractionation of  $^{18}\text{O}$  and  $^2\text{H}$  implies a correlation between the two parameters, which has been defined by different authors:

Craig [9] and Rozanski [10]: Global Meteoric Water Line (GMWL:  $\delta^2\text{H}=8 \delta^{18}\text{O}+10$ );

Zouari [11]: Local Meteoric Water Line of the Tunis-Carthage (LMWL:  $\delta^2\text{H}=8 \delta^{18}\text{O}+12.4$ ).

The Global Meteoric Water Line (GMWL) corresponds to the averaging of numerous local meteoric straight lines (regional data), each of which is influenced by the abovementioned geographic and climatic factors. The intercept of the GMWL is termed the deuterium excess ( $d=\delta^2\text{H}-8 \delta^{18}\text{O}$  after Clark [12]). The value of this parameter is acquired during evaporation, and does not vary significantly during the later history of the cloud mass. It is thus a valuable indicator of the source area of the water vapor Rindsberger [13]; Cruz [14]; Celle-Jeanton [15]:  $d$  values close to 10% indicate waters of Atlantic origin, values close to 22% are characteristic of waters from the Eastern Mediterranean and  $d$  values close to 14%, intermediate between the first 2, are detected in rainwater falling on the Western Mediterranean.

In this study, we report the correlation between  $\delta^{18}\text{O}$ ,  $\delta\text{D}$  and D-excess value in precipitations and their relationship with temperature. More importantly, their responses to changes in moisture sources will be discussed. This may prove beneficial in understanding the response of precipitation  $\delta^{18}\text{O}$  and  $\delta\text{D}$  to climatic/environmental variables in Northwestern Tunisia (El Kef area) and in understanding the palaeoclimatic records associated with precipitation  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in monsoon climate studies.

### General Setting

#### Geography

The study area is located in the North Africa and in the NW part of Tunisia, a site located at the southern margin of the Tethys. The El Kef area is located between  $40^{\circ}30'$  and  $39^{\circ}45'\text{N}$ , and  $6^{\circ}30'$  and  $7^{\circ}30'\text{E}$ , near the Atlantic-Mediterranean confluence (Figure 1). It extends 60 km in the N-S direction and 60-80 km in the E-W direction, covering an area of 420 km<sup>2</sup>. The elevation of the El Kef Plateau decreases from 400-500 m above sea level (m a.s.l.) (Figure 1).

#### Climate

Meteorological data (precipitation and temperature) obtained at the nearest meteorological station (El Kef meteorological station:  $7\text{G}09^{\circ}00'\text{N}$ ,  $40\text{G}20^{\circ}70'\text{E}$ , asl 10 m) around the sampling site includes only the period from 1924 to 2001. During this period, the maximum of precipitation was observed in January (58.6 mm) and a minimum in July (9.4 mm) (Figure 2); the maximum and minimum monthly mean temperatures that were recorded was approximately  $36^{\circ}\text{C}$  in August and  $3^{\circ}\text{C}$  in January, respectively (Figure 3). Relative humidity was maximum in spring ( $\approx 65\%$ ) and minimum in summer ( $\approx 26\%$ ). Over 80% of the annual precipitation occurs during the rainy season from December to May. The precipitation that occurs during dry season from June to November accounts for about 20%. The potential evapotranspiration exceeds 1,300 mm [16].

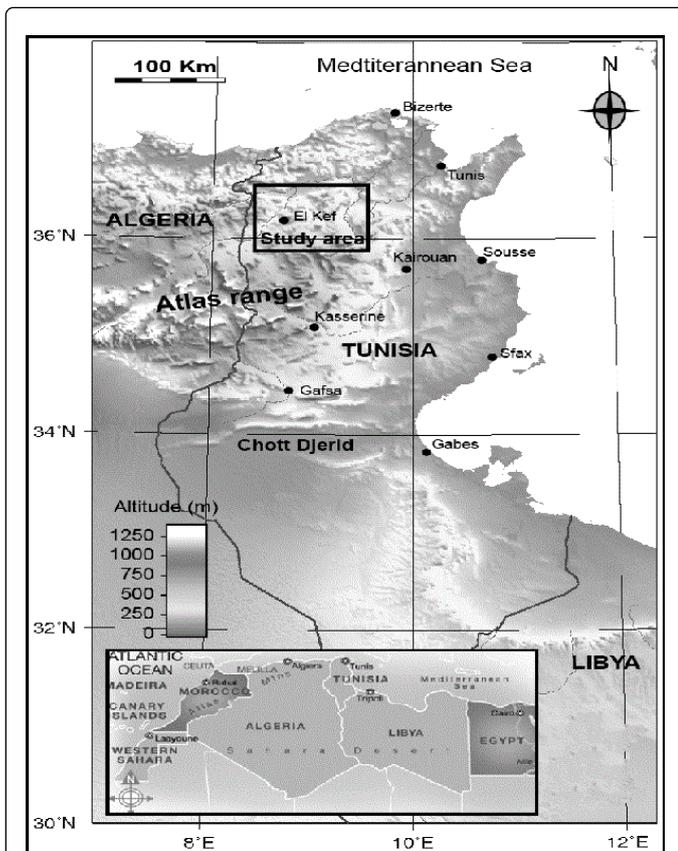


Figure 1: The localization map of El Kef region (NW Tunisia).

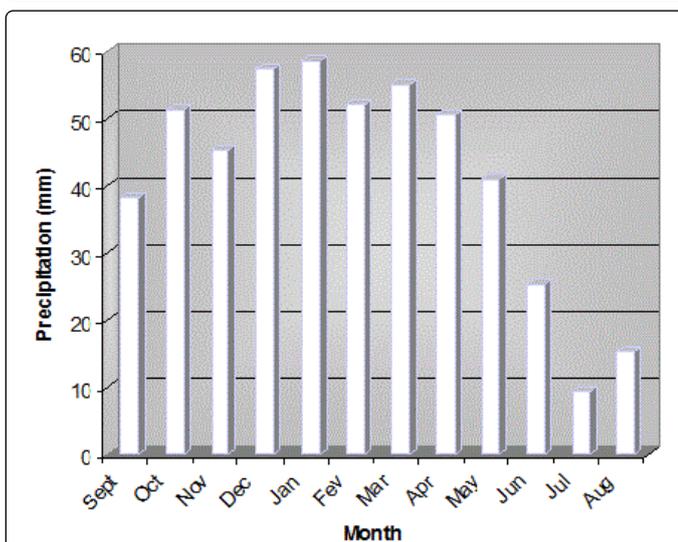


Figure 2: The averages of monthly precipitation from 1924 to 2001 in El Kef area.

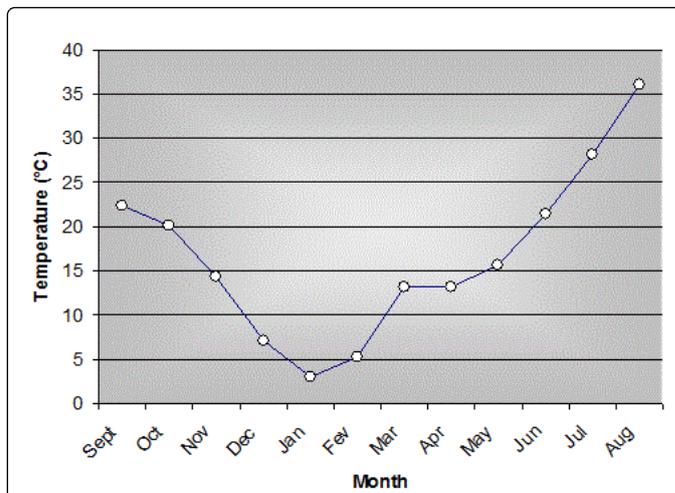


Figure 3: The averages of monthly temperature from 1924 to 2001 in El Kef area.

## Methods and Sampling

Owing to financial constraints, a limited number of modern meteoric water samples were selected for oxygen-18 and deuterium [16]. Hydrogen and oxygen isotope analyses were performed in the Laboratory of the International Agency of Atomic Energy (IAEA) in Vienna, by employing, respectively the standard CO<sub>2</sub> equilibration [17,18] and the zinc reduction techniques [19], followed by analysis on a mass spectrometer. Oxygen and hydrogen isotopes analyses were reported to  $\delta$  notation relative to Vienna-Standard Mean Oceanic Water (V<sub>SMOW</sub>), where  $\delta = [(R_s/R_{SMOW}) - 1] \times 1,000$ ;  $R_s$  represents either the <sup>18</sup>O/<sup>16</sup>O or the <sup>2</sup>H/<sup>1</sup>H ratio of the sample, and  $R_{SMOW}$  is <sup>18</sup>O/<sup>16</sup>O or the <sup>2</sup>H/<sup>1</sup>H ratio of the SMOW. Typical precisions are  $\pm 0.1$  and  $\pm 1.0\%$  for oxygen-18 and deuterium, respectively. The isotopic composition of oxygen ( $\delta^{18}O$ ) was measured after equilibration with reference CO<sub>2</sub> at 25°C for 24 h [17], while reduction on Cr at 800°C was used to determine the isotopic composition of hydrogen ( $\delta D$ ) in water [20]. Both measurements were performed on a Varian MAT 250 mass spectrometer.

Precipitation was captured using a dual bucket system in which a smaller bucket with a small hole at the bottom was placed snugly inside a larger bucket. During a rain event, the water drained through the hole, where it would remain inside the larger bucket until sample retrieval. After a snowfall, the sampler was brought inside where the water was melted and drained into the lower bucket. The precipitation samples were then transferred to 5 ml glass vials with poly-seal cone closures. These samples were collected in 1-L Nalgene bottles and then transferred after melting into 5 ml glass vials.

## Results and Discussion

### Regional isotopic composition of precipitation

The results for meteoric waters are given in Table 1. The isotopic compositions of collected meteoric waters range from -8.45 to -4.12‰ ( $\delta^{18}O$ ) and -55.40 to -22.60‰ ( $\delta^2H$ ) for El Kef area in 2001 [16]. These results are plotted in Figure 6 and in the Table 1. This variation is controlled by local climatic parameters, including the origin of the

vapor mass, re-evaporation during rainfall, the seasonality and monthly of precipitation; and suggestive of different atmospheric sources during these months as compared to the other rainy season months of July through September [12,16,21-23]. Generally, the negative precipitation isotopic values are attributed to the air temperature gradient and the massifs that surrounding the study area (Algerian and Tunisian Atlas: “1,000 ≤ altitude ≤ 1,500m”) (Figure 4). One of the factors that can be identified as influential is the distance of the stations from the Ocean and/or the sea.

Date	Amount of precipitation	Temperature (°C)	Delta <sup>18</sup> O	Delta <sup>2</sup> H	D-excess
January 2001	58	3	-8.45	-55.4	12.2
February 2001	53	5	-7.35	-41.5	17.3
March 2001	55	13	-6.88	-42.4	12.64
April 2001	51	13	-5.48	-36.6	7.24
May 2001	41	16	-4.12	-22.6	10.36

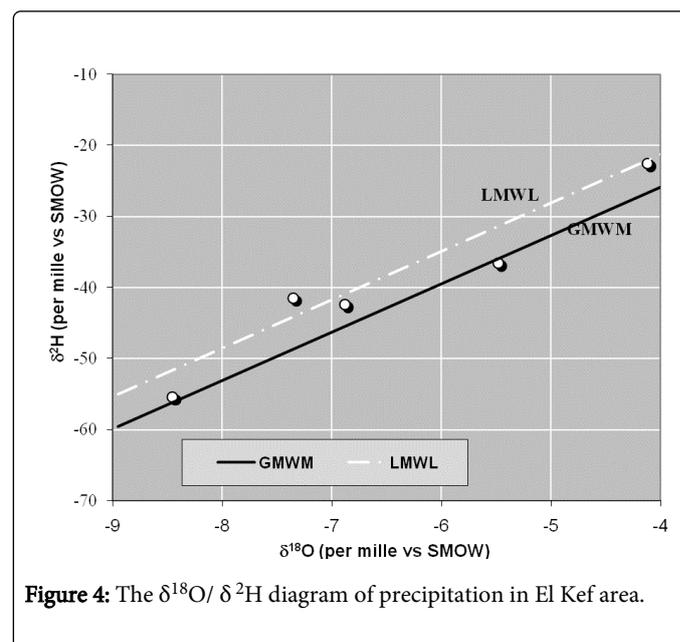
**Table 1:** Isotopic data for the precipitation in El Kef area (2001). (Delta <sup>18</sup>O and <sup>2</sup>H: per mille VSMOW “Vienna-Standard Mean Ocean Water”)- Precipitation (mm)

These negative residuals of the observed  $\delta^{18}\text{O}$  values in precipitation most likely result from strong convective precipitation during the period of Atlantic Ocean monsoon and the Mediterranean Sea activities. Furthermore, positive residuals are observed in the desert area of North Africa by several scientific studies [24-27]. These positive residuals can probably be attributed to the moisture forming precipitation coming from inland vapor cycle. The dry continental air masses usually result in  $^{18}\text{O}$  enrichment in precipitation [28,29]. Simultaneously, strong evaporation in these regions also causes raindrops to become increasingly enriched in  $^{18}\text{O}$  of precipitation. Additionally, it is noticeable that observed stations of these regions are relatively sparse (Tunis-Carthage and Sfax stations belonging to the GNIP network), which imparts influence on the precision of interpolation results to some extent.

By concerning the amount of precipitation and temperature effects, the values of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in precipitation (Figure 5) decrease with increasing precipitation. At a monthly timescale, the precipitation oxygen isotope composition co-varies with the amount of precipitation “amount effect” (showing negative relationship) and temperature (showing positive relationship) “temperature effect” at the study area (Figure 6). Although a longer time series in Northwestern Tunisia (El Kef basin) would be needed in order to confirm exactly the link of amount of rainfall and temperature with the oxygen isotope composition of precipitation, the available data (only 6 months) and mainly the geological and the climatological regional context support that these variables are direct controls of the isotope signal of precipitation. Over this period, each precipitation event higher than 2 mm (excluding potential precipitation altered by partial evaporation of droplets during fall) was collected and analyzed for deuterium and oxygen-18. The variability of oxygen isotopes in precipitation explained by amount of precipitation (discarding months with upper 40 mm of precipitation) (Table 1).

In other words, the evaporation during passage of raindrops through a warm dry atmosphere typical of semi-arid areas causes a

greater enrichment in heavy isotopes in precipitation during small rainfall events than in large rainfall events [6]. This “amount effect” has also been observed in the northern Sahara, where heavy winter rainfalls are generally depleted in heavy isotopes [24,30]. It is also possible that air masses coming from central Europe can contribute to the depletion of isotopic composition (continental effect). The distance between the moisture source and the site of precipitation has been suggested to impact the oxygen isotope composition of precipitation [31,32]. According to the Rayleigh distillation process governing this relationship, the depletion of moisture vapour due to precipitation along the air mass trajectory results in a progressive  $^{18}\text{O}$  depletion of the remaining moisture fraction in the atmosphere and the subsequent precipitation events [6,33-37] was the first to recognize that the  $\delta\text{D}$  and  $\delta^{18}\text{O}$  composition of precipitation was negatively correlated with temperature, latitude, altitude, distance from the coast, and the amount of precipitation. The change in moisture sources is an important contributor to the precipitation isotope variability [13]. Thus, the paleoclimate oxygen isotope records depending on the isotope composition of precipitation are potentially impacted by moisture sources (continental moisture originates from evaporation of soils, surface waters and plants transpiration), the origin of the precipitation “precipitation from Mediterranean in relation to Atlantic sources, the atmospheric dynamic” [22,23,33,38-41].



**Figure 4:** The  $\delta^{18}\text{O}/\delta^2\text{H}$  diagram of precipitation in El Kef area.

### Interpretation of oxygen-18 and deuterium data

The standard diagram of  $\delta^{18}\text{O}/\delta^2\text{H}$  (Figure 4) shows the position of all samples relative to the Global Meteoric Water Line (GMWL:  $\delta^2\text{H}=8\delta^{18}\text{O}+10$ ) [9,10] and the Local Meteoric Water Line of the Tunis-Carthage (LMWL:  $\delta^2\text{H}=8\delta^{18}\text{O}+12.4$ ) [11] closest to our study area belonging to the GNIP network. This diagram shows that all meteoric water samples lay between the GMWL and the LMWL. This arrangement signifies that the precipitation ensuring the recharge of the El Kef area originates from a mixture of Oceanic and Mediterranean vapor masses. This suggests that the precipitation as the predominant source from the Atlantic (with the intercept +40%) and from the Mediterranean (with the intercept up to +60%) during this period of rain (January to May 2001). However, quantifying the

contribution of each of these origins to the total recharge of the basin is difficult, and no simple and reliable methods are currently available. These findings and estimations remain to be developed in the future but with a lot of isotopic data. However, this contribution varies from one period to another and to a geographical area to another; depending on several other parameters (climate, geographical position, storm trajectory, atmospheric moisture, temperature, latitude, altitude, distance from the coast, and the amount of precipitation, also the influence of the isotopic signature of local precipitation “local vapor sources”). The same phenomenon was also observed in the central and southern Tunisia [21,22,42-45]. A mixed isotopic signature of Atlantic and Mediterranean origin was also observed in the karst aquifers of southeastern and south Spain [46,47] and northeastern of Algeria-El Eulma basin [48].

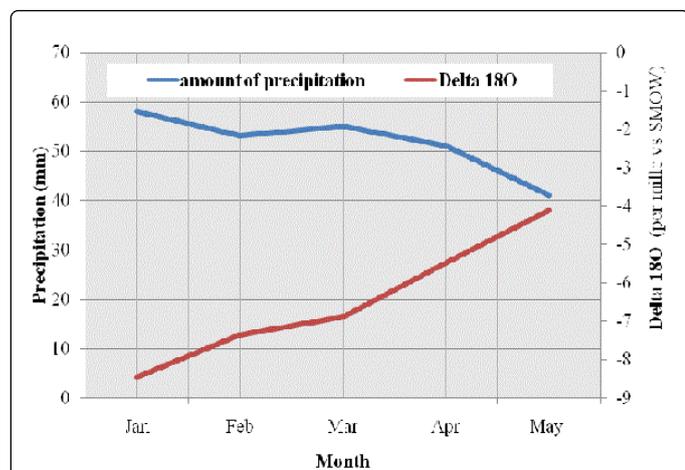


Figure 5: Plot  $\delta^{18}\text{O}$  vs Precipitation vs Months in El Kef area.

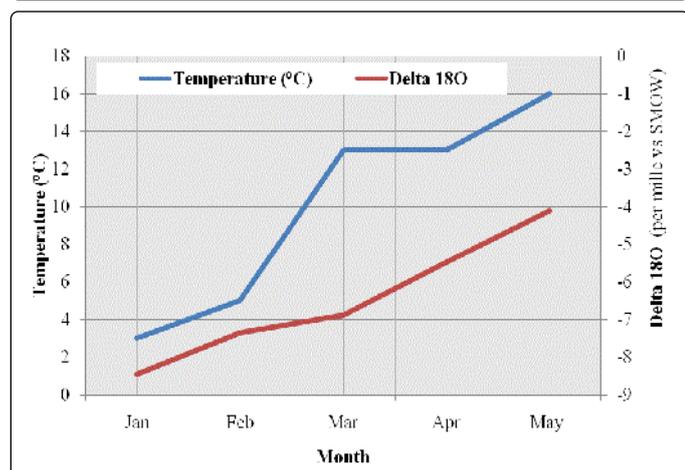


Figure 6: Plot  $\delta^{18}\text{O}$  vs Temperature vs Months of precipitation in El Kef area.

The air passing over El Kef area reflect the combined effects of oceanic-sea vapor sources, fractionation due to local precipitation, and slower equilibration of the larger raindrops nucleated by a maritime aerosol (Mediterranean Sea from the North and from the East). Both larger raindrops as well as the potential for more night-time precipitation [49] may explain the variation isotopic signature of

precipitation at El Kef area [23]. Remember, that the observations at this site are based on only half of one year of precipitation data, and additional monitoring may be necessary to confirm the results presented here.

Additionally, the samples that are characterized by a relatively depleted oxygen-18 and deuterium contents, indicate that they are not significantly affected by evaporated. Therefore, the most  $^{18}\text{O}$ -enriched value corresponds to rain collected during summer (evaporative influence). This value corresponds to summer precipitation with intense evaporation of the raindrops beneath the cloud base with surface air temperatures around  $40^\circ\text{C}$ . The deuterium values are linearly and positively correlated to the  $\delta^{18}\text{O}$  values (Figure 4). The linear correlation was found by many scientists [17,50]. The physical basis for this correlation lies in the fractionation of isotopes during evaporation-condensation processes [51]. This is attributed to the air temperature gradient and the massifs of the country (Tunisian Atlas). One of the factors that can be identified as influential is the distance of the stations from the sea ( $\approx 100$  km).

Tunisia is located in the Western Mediterranean, which represents a climatic transition zone open to the influence of the cool North Atlantic air masses and the warm Mediterranean air masses [13,43]. Moreover, specific geomorphologic characteristics of Tunisia i.e. the absence of high mountains with elevations exceed 1,500 m and the relatively limited geographic extensions allow the integration of Saharan air masses into the atmospheric circulation [34]. However, hydro-meteorological studies [35,52] suggest the existence of two major trajectories for dominant air masses. These are (i) Atlantic air masses that circulate from the west over Northern Africa and (ii) Mediterranean air masses that come from the north (Figure 1). Quantitatively, Mediterranean precipitation represents  $\approx 66\%$  of the total rainfall. The main part of the regional aquifer recharge is supplied by Mediterranean rain events. The question about how this affects the local precipitation and precipitation  $\delta^{18}\text{O}$  needs future detailed work, for this influence may vary in time and in space.

Deuterium-excess (D-excess) (Table 1), defined as ( $\text{D-excess} = \delta^2\text{H} - 8 \delta^{18}\text{O}$ ), is generally associated with the moisture sources of the precipitation and Sea Surface Temperature (SST; positive correlation) at the moisture source [6,53,54]. It is generally negatively correlated with the relative humidity of the air masses formed above the ocean [53-55], and appears to be used to indicate climatic changes in the moisture source regions [56,57]. Using this relationship, D-excess has been used in many studies to determine the temporal changes in moisture supply for a given location [58]. The average D-excess in El Kef basin during 2001 (from January to May) is 11.94‰. The d values, varying from 7.24‰ (April) to 17.3‰ (February). The high D-excess for the winter samples indicating evaporation in low humidity conditions and low D-excess in summer (coupled with the higher  $\delta^{18}\text{O}$ ) indicating the possible re-evaporation below the cloud.

These results indicate a larger proportion of local moisture for the precipitation in 2001 in northwestern Tunisia. To improve the quality of this moderate interpretation, probably we should adopt a model analysis on trajectory of air-mass with combination with several isotopic analyses. Moreover, like several scientific authors in the word, I hope that application of these isotopic and modeling methods (GIS) would become of common use in order to advance in our knowledge of the oxygen, deuterium isotopes and D-excess composition at different areas in the word and to improve and facilitate the interpretations of paleoclimate depending on it and with the inter-discipline inter-collaboration.

## Conclusions

Isotopic composition of precipitation in semi-arid northwestern Tunisia (El Kef area) is highly variable and barely seasonally controlled. This variability is at least in part explained by different pathways of the rain-bringing air masses. We were able to show that air masses coming from southern and western directions in general have significantly lower “d” values than rains from northern and eastern directions due to rainout effects of air-masses. Evaporation effects dominate the isotopic composition of many rainfall events with less than 2 mm, as indicated by their low “d” values.

The relationship between  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  for local precipitation in El Kef region shows that precipitation originating from cold and wet to hot and dry and the differing contribution of vapor derived from the closed marine basin of the Mediterranean Sea and the Atlantic Ocean vapor source that contains a maritime aerosol. However, in Tunisia geographical diversity influences the complicated mixing of continental and maritime air masses and the sources of air masses even change during a rain event. A mixed isotopic signature of Atlantic and Mediterranean origin was also observed in the aquifers of south Tunisia, Algeria, Morocco and Spain [21,22,47,59-61]. In reality, this seasonal variability of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values apparent in precipitation in the El Kef area is likely to be reflected in the isotopic composition of the surface waters and the ground waters [23].

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