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Cave dripwater isotopic signals related to the altitudinal gradient of Mount-Lebanon: implication for speleothem studies

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Abstract: An important step in paleoclimate reconstructions based on vadose cave carbonate deposits or speleothems is to evaluate the sensitivity of the cave environment and speleothems to regional climate. Accordingly, we studied four caves, located at different altitudes along the western flank of Mount-Lebanon (Eastern Mediterranean). The objectives of this study are to identify the present-day variability in temperature, pCO_2, and water isotopic composition and to assess the possible influence of the altitudinal gradient on cave drip waters and cave streams. We present here an overview of the spatial variability of rainwater based on local and regional data, and we compare these data with our results, i.e., temperature, air pCO_2, and the isotopic composition of cave water and modern cave calcite collected in 2011 and 2014. The results show that the rainwater isotopic signal is generally preserved in the cave dripwater isotopic composition with some exceptions in large caves with high ceilings where evaporation effects may influence its isotopic composition. The altitude effect observed in rainwater isotopic composition seems to be transferred to the cave dripwater. Different $\delta^{18}O$/100 m gradients between dripwater and rainwater (0.13‰ and 0.21‰, respectively) are noted. This is mainly attributed to the $\delta^{18}O$/100 m value of the dripwater which is site-specific and dependent on i) local processes within the epikarst/soil, ii) the relation to the precipitation altitude gradient and iii) the extension of the defined infiltration basin.

Keywords: drip water, isotopic signal, Lebanon, caves, altitude gradient

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INTRODUCTION

Speleothems, which are secondary cave carbonate deposits that precipitate from cave drip water are increasingly used to reconstruct changes in regional climate and vegetation. Their isotopic composition, $\delta^{18}O$, and $\delta^{13}C$ is influenced mainly by respectively the isotope signature of rainwater linked to temperature (Clarck & Fritz, 1997; Lachniet, 2009) and by the carbon isotopic composition of dissolved carbon influenced by the soil bioactivity ($\delta^{13}C$), linked to vegetation and thus to temperature and water availability (Hellstrom et al., 1998; Genty et al., 2006). Rainwater and dissolved carbon circulate through the unsaturated zone, i.e. the upper part of the epikarst, which is affected by dissolution and is characterized by a mainly vertical transfer of percolation water to the cave (Hendy, 1971; Bar-Matthews et al., 1996; Ford & Williams, 2007; Fairchild & Baker, 2012).

Several cave monitoring programs have been conducted worldwide, providing information on the role of local cave environment and hydrology that possibly influence stalagmite-based palaeoclimate proxy records (Bar-Matthews et al., 1996; Spotl et al., 2005; Baldini et al., 2006; Verheyden et al., 2008a; Mattey et al., 2010; Miorandi et al., 2010; Tremaine

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et al., 2011; Johnston et al., 2013; Genty et al., 2014; Deininger et al., 2014; Van Rampelbergh et al., 2014; Suric et al., 2016; Beddows et al., 2016). These studies aim at understanding better the registration of the nowadays climatic signal in speleothems in order to more precisely constrain the past climatic records.

In the Levant region, although speleothems from Palestine/Israel are well studied and proxy interpretations well-supported by monitoring programs, the rest of the region is still understudied in terms of speleothem-based paleoclimate studies (Nader et al., 2007, Verheyden et al., 2008b, Cheng et al., 2015, Nehme et al., 2015; 2018). Nearly no cave monitoring information is available and only limited rainwater isotopic data is available (Aouad-Rizk et al., 2005; Gat el al., 2005, Abou Zakhem & Hafez, 2010). Aouad-Rizk et al. (2005) and Koeniger & Margane (2014) each defined a local meteoric water line (LMWL) based on data from central Mount-Lebanon. Both studies display some differences which will be further discussed in the paper. The Levant (East-Mediterranean) is characterized by abrupt temperature and rainfall gradients, due to its current location on the arid/semi-arid boundary and to its steep topography between coastal and inland areas.

In a first attempt to understand the local environmental conditions, four Lebanese caves were investigated for their temperature, *p*CO2 concentration, and dripwater isotopic composition. Here, modern changes in the rainwater isotopic composition across the steep altitudinal trend of Mount-Lebanon, are compiled based on a literature review, and compared with the observed changes in modern cave drip waters. The main objectives of this paper are to discuss if rainwater signal is generally preserved in the cave dripwaters and to assess the possible influence of the altitudinal gradient on cave drip waters and cave streams. This study will help also verify the ability of cave waters in Lebanon to transfer spatial changes in isotopic composition of rainwaters and by extension of spatial and temporal changes in regional climate.

THE STUDY AREA: REGIONAL CLIMATIC CONTEXT AND SITE DESCRIPTION

The Levant region in general is mainly influenced by the mid-latitude westerlies (Fig. 1A), which originate from the Atlantic Ocean, forming a series of subsynoptic low-pressure systems (Gat et al., 2003; Ziv et al., 2010) across the Mediterranean Sea. In winter, cold air plunging south over the relatively warm Mediterranean enhance cyclogenesis, creating the Cyprus Low (Alpert et al., 2005). This low-pressure system drives moist air onshore, generating intense orographic rainfall across the mountains of the northern Levant. The duration, intensity, and track of these storm systems strongly influence the rainfall amount in this region. In summer, the westerly belt is shifted to the north, following the northern shift of the North-African subtropical high pressures, and the region experiences hot and dry conditions with more southward winds. In Lebanon (Fig. 1B and 1C), the annual rainfall varies between 700 and 1000 mm along the coastline and more than 1400 mm in higher mountains with 4 months snow coverage (Shabaan et al., 2015). As a consequence of the above circulation system, the climate is seasonal with wet winters (November to February) and dry, hot summers (May to October). A general N-S gradient in rainfall amount and mirrored by the isotopic signal ($\delta^{18}O$ and $\delta^{2}H$) is clearly evident from northern Syria (Abou Zakhem & Hafez, 2010), to southern Israel/Palestine (Gat et al., 2005). A West-East gradient, i.e. from the Levantine coastline to inner regions (Fig. 1A), is also visible as a consequence of the continental and/or altitudinal effects related to the Rayleigh distillation processes (Dansgaard, 1964; Rozanski et al., 1993).

Four caves are selected at different altitudes along a transect from the coast to the Makmel Mountain, which is the highest peak in the Mount-Lebanon range (Fig. 2). These are: Kanaan Cave (96 m above sea level - asl), Jeita Cave (98 m asl), Mabaage Cave (770 m asl), and Qadisha Cave (1720 m asl).

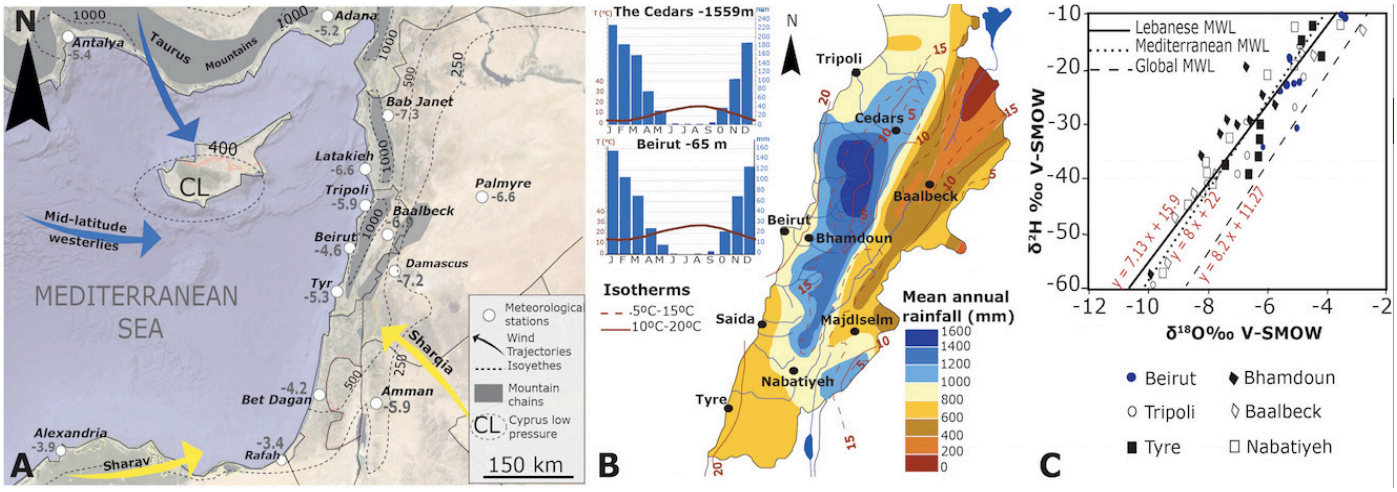


Fig. 1. Climate and geographic setting of the study area. A) Eastern Mediterranean map showing the position of the mid latitude winds (http://iridl.ldeo.columbia.edu/Maproom), NS and EW precipitation gradients and $\delta^{18}O$ mean values NS and EW precipitation gradients, of rainwater stations over coastal and inner cities (Kailani et al., 2003; El-Asrag, 2004; Aouad-Rizk et al., 2005; Dirican et al., 2005; Gat el al, 2005; Saad et al., 2005; Abou Zakhem & Hafez, 2010; GNIP database); B) Precipitation gradients of Lebanon and histograms of Beirut and the Cedars with mean annual rainfall and temperature (Abi-Saleh & Safi, 1988; http://fr.climate-data.org); C) Rainwater isotope graph with several published meteoric waterlines: the Lebanese MWL in Saad et al. (2005), the Global MWL in Rozanski et al. (1993), and the Mediterranean MWL in Gat (1980).

Except for Qadisha Cave which is developed mainly in Quaternary deposits and Cretaceous limestones (Dubertret, 1975), Mabaage, Jeita and Kanaan caves develop in the middle Jurassic Kesrouane Formation, a faulted micritic limestone and dolomite sequence (Walley, 1998). The studied caves (Table 1) are located in the western flank of Mount-Lebanon (Fig. 1) along a N-S altitudinal transect. All four caves were previously studied for their speleothem content (Verheyden et al., 2008a; Cheng et al., 2015; Nehme et al., 2015, 2018).

Kanaan Cave (162 m long) is located 15 km northeast of Beirut. This fossil cave was discovered after quarrying activity in the late 1990s (Nehme et al., 2009). The Jeita multi-level system cave, located at 4.5 km distance from the coast, hosts a series of dry and active galleries (Karkabi, 1990), a permanent stream with a discharge of 1 to 25 m^3/s (Doummar, 2012) and is the most visited show cave in Lebanon. A 75 m deep canyon connects fossil galleries with the lower galleries in the downstream extremity of the 10 km karstic network, making the cave a well-ventilated system (Fig. 2). Mabaage Cave 400 m long, located at 40 km northeastern of Beirut and in the inner part to the Fidar valley (Jabbour-Gedeon & Zaatar, 2013) was recently transformed into a touristic cave during summer but closes in winter due to flooding of the cave stream. Finally, Qadisha Cave, located in the northern part of Mount-Lebanon, hosts a permanent spring with a discharge rate up to 1 m^3/s (Edgell, 1997). Qadisha cave was partially transformed into a touristic cave in 1934.

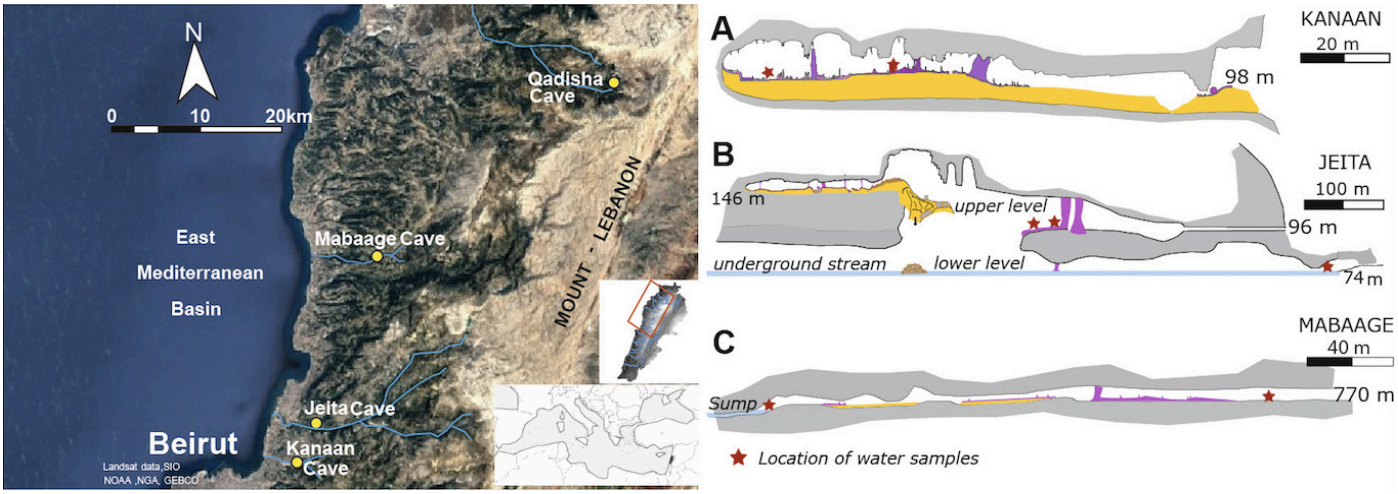


Fig. 2. The study area and cross-sections of A) Kanaan Cave (Nehme et al., 2013); B) Jeita Cave (Karkabi, 1990; Nehme, 2013); C) Mabaage Cave (Zaatar et al., 2013). For the cross-sec`tion of Qadisha Cave, see Tawk et al. (2008). Red stars indicate the location of water samples taken inside each cave and cave elevations are in m above sea level (asl). Refer to Table 1 for additional cave site data.

The vegetation cover above the caves mainly develops in shallow Mediterranean soil. Between 100 and 800 m asl, the vegetation consists of densely evergreen shrubs (juniper, oaks, and partially pine trees) growing on calcareous slopes above Kanaan, Jeita and Mabaage caves. The vegetation cover above Qadisha Cave (1720 m asl) is composed of sparse herbs, shrubs, and conifers (Table 1).

Table 1. Locations, morphology of the studied caves, and their soil characteristics.

Cave	Coordinates	Cave type	Entrance (m)	Infiltration basin elevation (m)	Aspect	Length (m)	Host Rock	Vegetation type
Kanaan	33°54'25"N; 35°36'25"E	Horizontal, relict	98	547	SE-NW	162	J4-J5	dense garrigue, pine forest
Jeita upper	33°56'35"N; 35°38'48"E	Horizontal, relict	96	1067	N-S	1300	J4-J5	dense garrigue, pine forest
Jeita Lower	33°56'35"N; 35°38'48"E	Horizontal, active	60	1669	E-W	8750	J4-J6	dense garrigue, pine forest
Mabaage	34°06'25"N; 35°46'01"E	Descending cave	770	1379	E-W	400	J6	sparse garrigue, oaks, pine
Qadisha	34°14'38"N; 36°02'11"E	horizontal, spring	1720	2244	NE-SW	1076	Q; C4	Sparse hurbs, shrubs, conifer

SAMPLES AND METHODS

A total of 35 cave drip water and 12 underground stream water samples were collected in the Jeita and Qadisha caves for $\delta^{18}O$ and $\delta^{2}H$ analyses, respectively. The samples were obtained during two sampling campaigns: a first one held in September 2011 in Jeita and Qadisha caves and a second one between September and November 2014 in Jeita, Qadisha, Mabaage, and Kanaan caves.

Temperature and pCO_2 of cave air were measured using a hand thermometer with a precision of 0.5°C and a Dräger pump system (σ ± 50 ppmv), respectively. Continuous temperature monitoring using a Niphargus (Burlet et al., 2015) temperature logger (precision of 0.1°C and resolution of 0.05°C)

was pursued from December 2015 to March 2017 in Qadisha and Jeita caves with one measurement every 20 minutes.

Isotopic analyses of cave waters collected in 2014 were carried out using a PICARRO L2130-i Cavity Ring-Down Spectrometer (CRDS) at the Vrije Universiteit Brussel. Measured values were corrected using three house standards calibrated against the international VSMOW2, GISP, and SLAP2 standards following the method described in De Bondt et al., (2018). Analytical uncertainties (2σ) equal 0.06‰ for $\delta^{18}O$ values and 0.3‰ for $\delta^{2}H$ values. Water samples collected were analyzed in 2011 at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE-CEA), Paris. Hydrogen isotopes were measured on an ISO-PRIME mass spectrometer and a PICARRO CRDS with a 1 sigma error of ±0.7‰. Oxygen isotopes were analyzed using a Finnigan MAT 252 by equilibration with CO_2. The 2 sigma error of the $\delta^{18}O$ is ±0.05‰. All values obtained from both laboratories are calibrated against and reported in permill (‰) relative to Vienna Standard Mean Ocean Water (V-SMOW2).

To calculate the altitudinal gradient of the cave dripwaters with respect to the altitude of the entrance and the infiltration basin of the studied caves, the infiltration basin elevation (Table 1) was derived after plotting the georeferenced caves maps on a Digital Elevation Model (DEM) using a Geographical Information System (ArcGIS). The infiltration watershed area of the cave is defined by considering the altitudes between the cave entrance and the limit of the surface watershed. The underground waterflow main directions identified in Hakim (1985) and Hakim et al. (1988) for the Lebanese karst basins were considered to derive the most significant infiltration surface above the caves. The mean altitude is then calculated for the delimited infiltration basin for each cave using the DEM. Note that the Jeita Cave develops on two-levels (an upper fossil and a lower active gallery) thus has two different infiltration elevations.

RESULTS

Cave air temperature and pCO_2

Cave air and underground stream temperatures, measured at different sites inside each cave (see Supplementary Data), a fairly constant (Fig. 3A) with variations of less than 1°C over the sampling period. The measured air temperatures in Kanaan (19°C ± 0.5), Jeita upper (20°C ± 0.5), Mabaage (13°C ± 0.5), and Qadisha caves (9°C ± 0.5) all display autumn values roughly in agreement with the outside mean temperature (Fig. 3B) data (Karam, 2002), despite the small offset compared to the surface temperature trendline and some small internal changes (up to 0.3°C) as shown by the continuous monitoring data in Jeita and Qadisha (Fig. 3C).

As for the pCO_2 concentrations in each cave, the measured values reached 3,600 and 8,000 ppmv in Jeita and Mabaage caves respectively, whereas low values (600 ppmv) close to the atmospheric concentrations are detected in Qadisha Cave (see Supplementary Data).

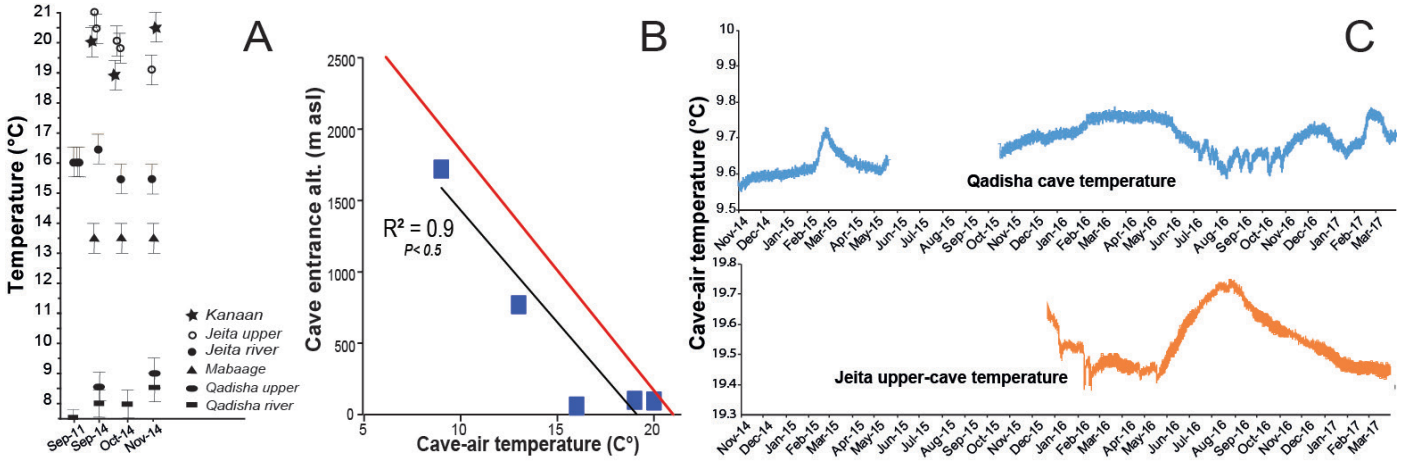


Fig. 3. A) Cave-air and underground stream temperature measurements collected each month in both 2011 and 2014 171 campaigns; B) cave-air temperature trendline (black line) vs the cave entrance altitude. The air-temperature trendline from compiled data of Lebanese meteorological stations (Karam, 2002) is represented in red line; C) continuous cave-air temperature monitoring from November 2014 to March 2017 for Qadisha and from January 2016 to March 2017 for Jeita Cave.

Cave dripwater $\delta^{18}O$ and $\delta^{2}H$

Cave dripwaters and stream waters $\delta^{18}O$ and $\delta^{2}H$ are summarized in Table 2 and detailed in the Supplementary Data. Jeita Cave dripwaters exhibit an average of -5.7 ± 1.1‰ for $\delta^{18}O$ and –26.6 ± 6.9‰ for $\delta^{2}H$ (Table 2), with an amplitude of 2.9 and 20.5‰, respectively. As for Kanaan Cave, measurements show an average of -5.40 ± 0.04‰ for $\delta^{18}O$ and –24.0 ± 0.2‰ for $\delta^{2}H$ (Table 2), with an amplitude of 0.2 and 0.5‰ respectively. Mabaage Cave located at higher altitude (770 m) shows an average of -7.2 ± 0.6‰ for $\delta^{18}O$ and –36.6 ± 6.2‰ for $\delta^{2}H$, whereas the amplitude varies between 1.8 and 14.0‰, respectively.

Qadisha Cave, located at the highest altitude in our study area, shows an average of -8.48 ± 0.05‰ for $\delta^{18}O$ and –46.1 ± 0.31‰ for $\delta^{2}H$ with an amplitude of 0.2 and 1.1‰, respectively. The variability of the dripwater oxygen isotopic signal in Kanaan (avg. -5.4‰) and Qadisha (avg. -8.5‰) does not exceed ±0.1‰ (Table 2). Drip water values in both Jeita and Qadisha caves show lower isotopic values than Jeita (avg. -7.2‰) and Qadisha (avg. -9.0‰) stream waters. However, the difference is much higher between dripwater and stream water isotopic values in Jeita Cave (~1.5‰) than those of Qadisha Cave (~0.4‰).

Table 2. Summary of the isotopic results of the drip and stream waters collected from the studied caves. Note that Jeita upper is the fossil cave and Jeita lower in the active cave with a permanent stream. (n) is the number of samples and oxygen isotopic composition ranges. The mean (avg.), maximum and minimum values of the $\delta^{18}O$ drip and stream water and mean (avg.) of the $\delta^{2}H$ of the autumn values, are reported in ‰ VSMOW.

Fossil Caves	***n***	**Dripwater $\delta^{18}O$ (‰ VSMOW)**				**Dripwater $\delta^{2}H$ (‰ VSMOW)**	
		min	**max**	**avg.**	**2s**	**avg.**	**2s**
Kanaan	6	-5.48	-5.37	-5.43	±0.04	–24	± 0.2
Jeita Upper	11	-6.92	-4.05	-5.71	±1.11	–26.6	± 6.9
Mabaage	8	-8.28	-6.64	-7.18	±0.66	-36.6	± 6.2
Qadisha	10	-8.55	-8.38	-8.48	±0.05	-46.1	± 0.3
Cave Streams	***n***	**Stream water $\delta^{18}O$ (‰ VSMOW)**				**Stream water $\delta^{2}H$ (‰ VSMOW)**	
		min	**max**	**avg.**	**2s**	**avg.**	**2s**
Jeita lower	6	-7.35	-7.17	-7.28	±0.06	-35.73	± 0.48
Qadisha	6	-8.96	-8.92	-8.95	±0.02	-49.24	± 0.26

DISCUSSION

In order to determine if the current spatial gradients in rainwater isotopic composition are recorded in the cave dripwater, we discuss *i)* the available meteoric water lines of Lebanon and their altitudinal trends, *ii)* the cave waters $\delta^{18}O/\delta^{2}H$ signals compared to the available $\delta^{18}O/\delta^{2}H$ rainwater data, and *iii)* the altitudinal trend in rainwater $\delta^{18}O/\delta^{2}H$ and the potential altitudinal trends in cave water $\delta^{18}O$ to test for their agreement.

Rainwater data of Lebanon: different meteoric water lines and altitudinal trends

Several studies on the rainwater isotopic signal in Lebanon (Aouad-Rizk et al., 2005; Saad et al., 2005; Saad and Kazpard, 2007; Koeniger & Margane, 2014; Koeniger et al., 2017) exist in the literature (Table 3). The trendline slopes of the MWL (Lebanon, Mount-Lebanon, etc.) are different than that of the Mediterranean MWL, due mainly to a secondary evaporation effect during rainfall events (Saad et al., 2005; Saad & Kazpard, 2007). The evaporation occurs mostly during hot (dry) seasons and is particularly impacting light rains. Consequently, this process will determine the lowering of the slope and the "d-excess" value of the rain sample (Clark & Fritz, 1997).

In general, the constructed Lebanese Meteoric water lines based on $\delta^{18}O$ and $\delta^{2}H$ data of rainwater are roughly in agreement with a general depletion trend with elevation. However, the local MWL (Koeniger & Margane, 2014; Koeniger et al., 2017) for the Kelb basin, the Mount-Lebanon (Aouad-Risk et al., 2005) and the general Lebanese MWL (Saad et al., 2005; Saad & Kazpard, 2007) are calculated based on different locations of the meteorological stations (Fig. 4A).

Table 3. Summary of the Global MWL and the MWLs previously calculated for the Mediterranean, Lebanon and Kelb basin.

Meteoric water line	**Equation**	**Author**
Global meteoric water line	$\delta^{2}H = 8 * \delta^{18}O + 10$	Craig, 1961
Mediterranean water line	$\delta^{2}H = 8 * \delta^{18}O + 22$	Gat, 1980; Gat et al., 2003
Lebanese water line	$\delta^{2}H = 7.13 * \delta^{18}O + 15.98$	Saad et al., 2005 ; Saad & Kazpard, 2007
Mount-Lebanon water line	$\delta^{2}H = 6.3 * \delta^{18}O + 8.2$	Aouad-Rizk et al., 2005
Kelb basin water line (Local)	$\delta^{2}H = 6.04 * \delta^{18}O + 8.45$	Koeniger & Margane, 2014; Koeniger et al., 2017

The MWL after Aouad-Risk et al. (2005) referred here as the Mount-Lebanon MWL, is constructed using data from meteorological stations which display the same E-W trend than the stations used for the MWL of the Kelb basin (Koeniger & Margane, 2014). Indeed, the MWL after Aouad-Risk et al. (2005) and the local MWL after Koeniger & Margane (2014) show the same gradient (Table 3).

The altitudinal trendline (Fig. 4B) used in this study is constructed after the latest rainwater data (Koeniger & Margane, 2014; Koeniger et al., 2017) from stations located in the Kelb basin (central Mount-Lebanon) since the collected data covers an elevation range up to 1600 m (Chabrouh station) close to the basin altitude of the studied caves and includes snowfall isotopic signals. This altitudinal trendline show a linear $\delta^{18}O$-altitude relation of -0.13‰/100 m in West Mount-Lebanon (Fig. 4B), i.e., a decrease in rainwater $\delta^{18}O$ of 0.13‰ per 100 meters altitudinal increase.

Cave waters $\delta^{18}O/\delta^{2}H$ signals compared to the available $\delta^{18}O/\delta^{2}H$ rainwater data

The isotopic results of cave waters (drip and stream) of the four studied caves fall well on the Mount Lebanon MWL (Aouad-Risk et al., 2005), except for some of the Jeita Cave dripwaters (Fig. 4C). In general, Kanaan, Jeita, Mabaage and Qadisha $\delta^{18}O_{water}$ (drip and stream) values seems to fall more closely to the Mount Lebanon and Lebanese MWL than the regional Mediterranean MWL (Gat, 1980; Gat et al., 2003).

The $\delta^{18}O_{drip}$ values of Kanaan Cave are at the lower part of both Lebanese and Mount-Lebanon MWLs whereas Mabaage $\delta^{18}O_{drip}$ values are located at the center. $\delta^{18}O_{drip}$ values of Qadisha cave correspond to the highest part on both MWLs. Both Qadisha and Jeita $\delta^{18}O_{drip}$ results of 2014 fall generally close to the Lebanese MWL trend. Jeita $\delta^{18}O_{drip}$ results of 2011 show a distinct displacement to the right of the Mount Lebanon MWL, that clearly indicates evaporation

processes (Saad & Kazpard, 2007). The positive $\delta^{18}O_{drip}$ values of the 2011 campaign in Jeita Cave (Fig. 4C) suggest several processes and factors that might explain this particularity. Mickler et al. (2004) and Day and Handerson (2011) showed that the evaporation effect often occurs in caves with higher cave-air temperatures, which is the case for Jeita Cave (20°C), and less so for Qadisha Cave (9°C). Another possibility is related to other cave environment parameters which include enhanced ventilation (Muhlinghaus et al., 2009; Deininger et al., 2012) that would lead to enhanced out-of-equilibrium processes.

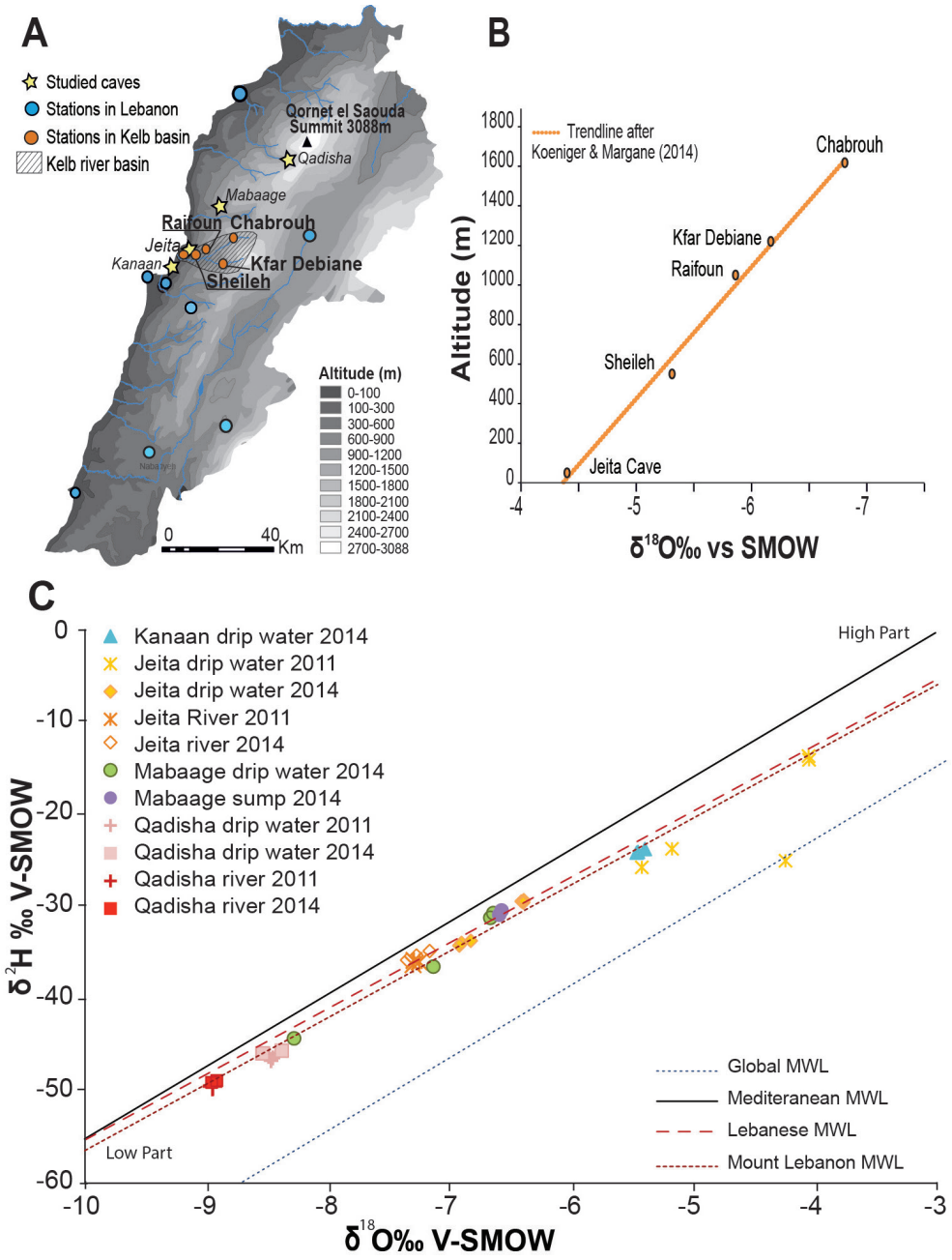


Fig. 4. Summary of major cave drip and stream water $\delta^{18}O$ and $\delta^{2}H$ values plotted on the available meteoric water lines (MWL) with the location of their meteorological stations. A) location of the studied caves (yellow stars) and the meteorological stations (blue circles) in Saad et al. 2005 and Saad & Kazpard, 2007 and (orange circles) in Koeniger & Margane, 2014; B) the altitudinal trendline used in this study and derived from the Local MWL of 222 the Kelb basin (Koeniger & Margane, 2014); C) $\delta^{18}O$ and $\delta^{2}H$ values of 47 dripwater and stream samples plotted on the available MWLs.

The $\delta^{18}O$ values for stream waters in the Qadisha and Jeita caves plot along the Mount Lebanon and Lebanese MWLs. The Qadisha stream water displays $\delta^{18}O$ values close to the drip water isotopic signal of the same cave suggesting a similar water infiltration source for the vadose and the karst aquifer (phreatic) zones. However, the isotopic signal of Jeita stream exhibits higher values than the drip water isotopic signal in Jeita upper cave advocating for different infiltration reservoir for the unsaturated and saturated zones. Indeed, the Jeita underground stream exhibits a $\delta^{18}O$ signal which is very close to the average $\delta^{18}O$ signal of the highest karstic springs feeding the Kelb basin: Nabaa el-Labane spring at 1647 m (avg. -7.26‰) and Nabaa al-Assal spring at 1528 m (avg. -7.32‰) (Aouad-Rizk et al., 2005; Koeniger et al., 2017). The isotopic signals are also in agreement with the well-known infiltration basin (or recharge area) for the Jeita underground stream situated at a mean altitude of 1669 m asl (Table 1).

The spread of the isotopic values cave waters along the Mount Lebanon MWL is related to the variability in $\delta^{18}O$ and $\delta^{2}H$ in rainwater and therefore in cave drip water. This is mainly due to the spatial variability, inter-seasonal, or interannual variations in isotopic composition of rain. It suggests that all sampled dripwaters in the caves, especially in Jeita and less in Qadisha may be related to rainwater from different seasons or even years depending on the residence time of the water in the vadose, here epikarst zone.

The deuterium excess (d-excess) value is calculated from $\delta^{18}O$ values and $\delta^{2}H$ using this equation:

$$\text{d-excess} = \delta^{2}H - 8 * \delta^{18}O$$

The d-excess, an indicator for the source and trajectories of atmospheric moisture (Rozanski, 1993; Sharp, 2007), is associated with evaporation at the moisture source. The comparison of d-excess_{drip} vs d-excess_{rain} is necessary to understand if the measured cave waters are controlled by a similar vapor source than the rainwater. The Eastern Mediterranean source waters have d-excess values ranging from 14‰ to 19‰ (Kattan, 1997), whilst they reach 15‰ (Frot et al., 2007) for Western Mediterranean sourced waters, and are close to 10‰ for Atlantic-sourced moisture. The Lebanese MWL exhibit a d-excess of 15.98‰ which is within the range of the Eastern Mediterranean waters' values (Kattan, 1997). The d-excess for the dripwater in cave indicate a value of 16.25, which is close to the one defined by the Lebanese MWL (Aouad-Rizk et al., 2005; Saad & Kazpard, 2007; Koeniger & Margane, 2014).

Altitudinal trends in cave water $\delta^{18}O$

The Mediterranean air masses arriving from the west are orographically uplifted as they reach the Mount-Lebanon range. As the air rises and cools, the rainwater with a heavier isotope falls first, resulting in rainwater exhibiting more negative isotopic values with altitude (Bowen & Wilkinson, 2002). Globally, the average change in $\delta^{18}O_{rain}$ is -0.2‰ per 100 m elevation gain (Rozanski et al., 1993). Locally, we determined this trend as -0.13‰/100 m (Fig. 4B). In order to understand the altitude effect on the isotopic composition of cave water (drip and stream), the $\delta^{18}O$ cave water values are plotted first against altitude of the cave entrances (Fig. 5A).

Only the $\delta^{18}O_{drip}$ and $\delta^{18}O_{stream}$ values that fall closely on the local MWL were retained for the altitudinal trend analysis (Fig. 5A). Figure 5A clearly shows a poor altitudinal trend when considering only the altitude of the cave entrance. However, there is a clear altitudinal trend with a regression coefficient $R^2 = 0.86$ ($P < 0.001$) when considering the mean altitude of the infiltration basin (or recharge area) of these caves (Fig. 5B and 5C). Indeed, the basin from where water infiltrates is at higher elevation relative to the cave entrance due to the thick limestone overburden and the topography above the caves entrance. The $\delta^{18}O_{water}$ values of Jeita stream are a clear example of that particularity, showing that the underground water originates from an infiltration basin at a higher altitude (avg. alt. 1669 m) than the cave entrance (60 m) (Doummar, 2012; Koeniger et al., 2017).

With the infiltration basin altitude (Table 1) taken into consideration here, all points fall within the 95% confidence interval (Fig. 5B and 5C) except one point, which represent cave waters taken from Mabaage Cave at the end of August 2014. All drip and stream waters $\delta^{18}O$ values reach 0.2‰ per 100 m (Fig. 5B) with an overall offset of 0.07‰ compared to the rainwater trend of 0.13‰ per 100 m calculated form the trendline (Fig. 4B) after Koeniger & Margane (2014).

Comparable studies in the Mediterranean region showed different small offsets between altitudinal gradients for precipitation and dripwater $\delta^{18}O$ values. In the steep northern Italian Alps, eight caves aligned along two transects, show slightly different gradients of 0.15 and 0.08‰ in dripwater (Johnston et al., 2013). In the eastern Adriatic coast and Dinaric mountains (Croatia), the offset reaches up to 0.2‰ (Suric et al., 2016) similar to the offset measured in Mount-Lebanon cave waters.

Clearly, the $\Delta\delta^{18}O$/100 m value of the dripwater (Fig. 5C) is site-specific, mainly due to: *i)* local processes that influence the $\delta^{18}O_{drip}$ on its way to the cave, such as those within the litter, soil, and epikarst (Beddows et al., 2016), *ii)* the relation to the precipitation altitude gradient (rainfall quantity, patterns, and frequencies above the cave infiltration

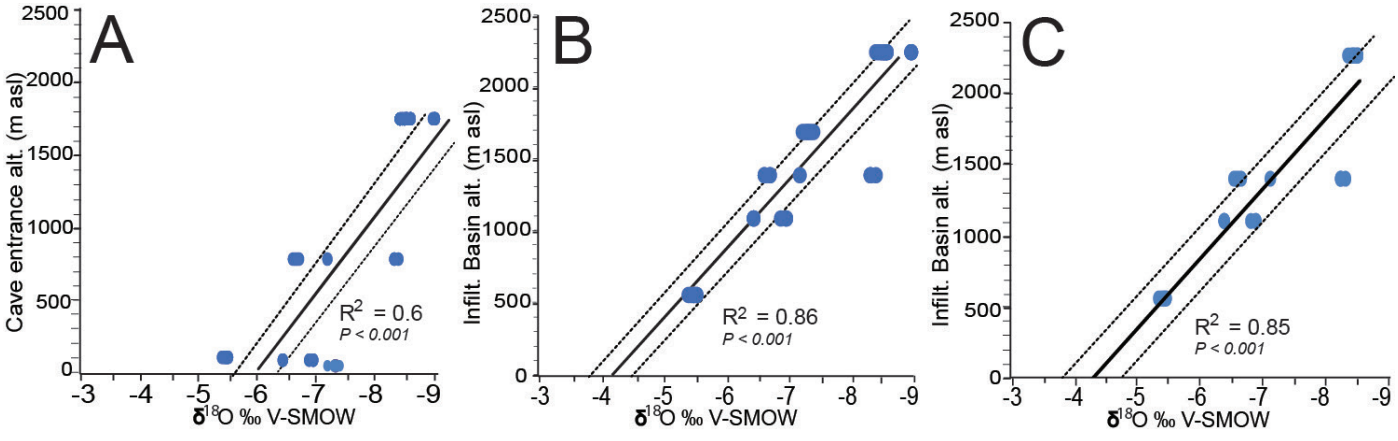


Fig. 5. Altitudinal trends in cave water $\delta^{18}O$ (drips and streams). A) Adjusted linear regression between the $\delta^{18}O_{cave\ water}$ and the altitude of the cave entrance; B) $\delta^{18}O_{cave\ water}$ and the altitude of the infiltration basin; C) Plot showing the $\delta^{18}O_{dripwater}$ trendline only vs the infiltration basin altitude. The calculated interval of confidence (dashed line) for each linear regression is 95% and the significance p-value for all three graphs is $P < 0.001$.

basin), or *iii)* the hilly topography above the cave and the limits of the calculated infiltration basin defined herein.

Figure 6A compares the offset between altitudinal gradients for precipitation and dripwater $\delta^{18}O$ values. Within the limit of the 95% confidence interval of the precipitation $\delta^{18}O$ trendline and considering the 2σ sigma error of the dripwater, the drip $\delta^{18}O$ values fall generally close to the precipitation $\delta^{18}O$ trendline except for Qadisha Cave. There is, however, a minor negative offset (0.2‰ at low altitude to 1.2‰ at high altitude) between the dripwater and the precipitation $\delta^{18}O$ trendline.

This offset, similar to the one observed for the Adige, Valsugana valleys, northern Italy (Johnston et al., 2013), the Adriatic coast (Suric et al., 2016), and Vancouver, Canada (Beddows et al., 2016) represents a bias due to the infiltration effects of rainwater. In winter, the infiltrated water from rainfall/snowmelt with lower $\delta^{18}O$ values reaches the cave, while in summer seasons, ^{18}O (^{2}H)-enriched water will partially evaporate in the unsaturated zone, especially when shallow overburden exists above the cave (Wackerbarth et al., 2010, 2012). The cave waters are therefore normally biased towards lower/lighter $\delta^{18}O/\delta^{2}H$ values compared to the rainwater isotopic signal (Wackerbarth et al., 2012).

Generally, cave dripwaters in Lebanon are mostly the result of percolation happening during the wet season (from autumn to spring snowmelt), with a longer infiltration period at higher altitudes due to snowmelt. For Qadisha Cave, which is located at a higher altitude, the offset between the precipitation $\delta^{18}O$ trendline and the dripwaters isotopic signals is the most negative when compared to the other studied caves. This is explained by the infiltration of winter water enhanced by a negative isotopic value of winter snow, especially at higher altitudes (Aouad-Rizk et al., 2005) and contributing into the vadose water budget.

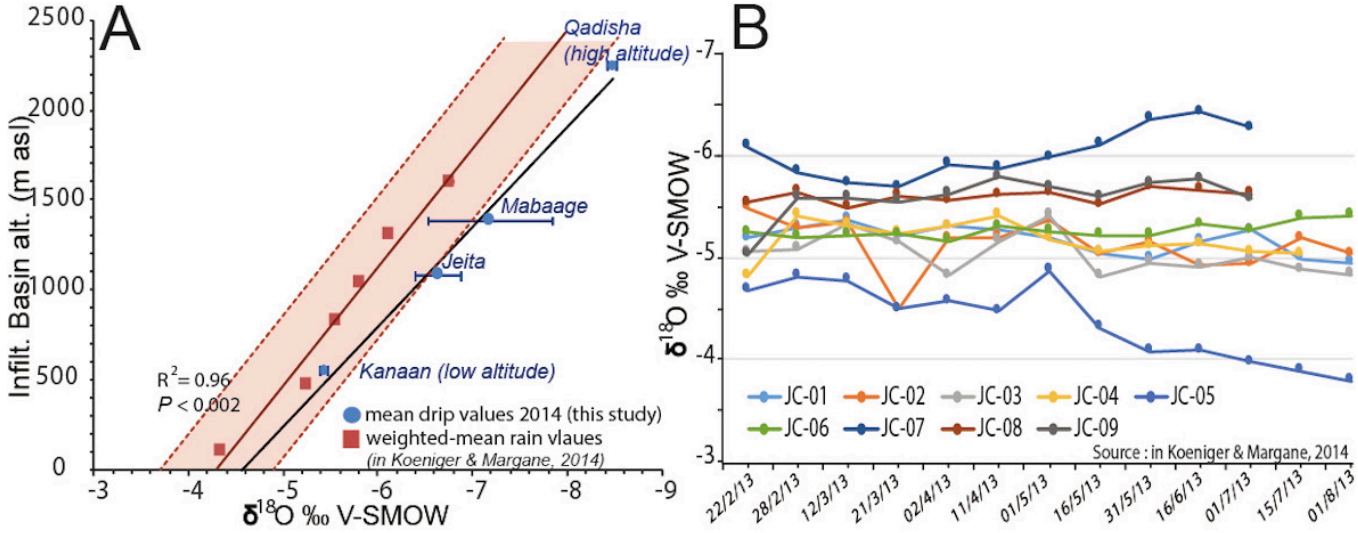


Fig. 6. Cave dripwaters $\delta^{18}O$ values in Lebanese caves compared to $\delta^{18}O_{rain}$: A) trendline showing the mean $\delta^{18}O_{dripwater}$ (blue dots) of each cave (this study) compared to the altitudinal trendline used in this study (red rectangles) and derived from the Local MWL ($\delta^{18}O$ weighted-mean rainwater values) after Koeniger & Margane (2014). The calculated interval of confidence (dashed line) for the $\delta^{18}O$ weighted-mean rainwater regression is 95% and the significance p-value is P < 0.002; B) $\delta^{18}O_{dripwater}$ measured at nine different sampling sites (JC-01to 09) in Jeita Cave on a yearly basis (data in Koeniger & Margane, 2014).

Regarding the altitudinal effect on the ^{18}O- and ^{2}H-depleted dripwater, our study shows that $\delta^{18}O_{drip}$ values decrease up to 3‰ between Kanaan and Qadisha caves (Fig. 6A). This amplitude attributed to the altitudinal effect could theoretically be increased by variations in the $\delta^{18}O_{drip}$ values related to site-specific characteristics or to a seasonal bias between winter and summer dripwaters values transferred by the rainwater seasonal variations. Indeed, rainwater $\delta^{18}O$ values in Lebanon show clearly a seasonal bias with a variation up to 5‰ (Saad & Kazpard, 2007; Koeniger & Margane, 2014) between early-winter and winter-spring seasons. In fact, seasonal variations in meteoric precipitation may range up to >15‰ for $\delta^{18}O$ (Genty et al., 2014). However, the majority of cave sites studied around the world (Genty et al., 2014; Beddows et al., 2016) demonstrated that drip waters typically show little or no isotopic seasonality compared to the variations in meteoric precipitation. For instance, the $\delta^{18}O_{drip}$ variability of caves in Vancouver Island, Canada is reduced in amplitude by 60–90% compared to the Victoria rainfall records of the same year. In Villars, Chauvet, and Orgnac caves, southern France, the $\delta^{18}O_{drip}$ values stayed stable for 15 years with little seasonal variations compared to drip rate measurement. In Lebanon, our $\delta^{18}O_{drip}$ data, even though stable during the autumn season prevent us from assessing a seasonal variability for all four cave sites. However, a previous campaign on dripwater isotopic measurement completed at nine drip sites in Jeita Cave (Koeniger & Margane, 2014) show little variability at each drip site over a complete rainy and early-summer season (Fig. 6B), but rather a spatial variability between each drip site. Indeed, the maximum seasonal variability of 1‰ is only recorded in JC-05 site (Fig. 6B). The $\delta^{18}O_{drip}$ yearly average is -5.24‰ in Jeita cave showing a low seasonal variability with a standard deviation of ±0.48. Therefore, the seasonal variations in cave dripwaters as seen in Jeita $\delta^{18}O_{drip}$ measurement, account less in the altitudinal effect on the lowering of the dripwater isotopic values.

Implications for future speleothems-based paleoclimate studies

The isotopic signal of the dripwater in Lebanese caves located on the western flank of Mount-Lebanon falls generally on the local MWL (Koeniger & Margane, 2014) as well as the Mount-Lebanon MWL (Aouad-Rizk et al., 2005). This implies: *i)* an identical source of water being derived from rain forming over the Mediterranean basin as indicated by similar d-excess values of the water, *ii)* a reduced evapotranspiration effect, observable on only some samples with a clear offset to the right of the MWL, probably due to increased cave ventilation, and *iii)* a longer infiltration period occurring in the unsaturated zone at higher altitudes.

Whilst some exceptions might occur as seen in some drip water in Jeita Cave during the 2011 campaign, which were more exposed to ventilation at some locations, most of the cave dripwater exhibits a similar $\delta^{18}O$ and $\delta^{2}H$ signal as the local rainwater. However, a slight offset towards lower 'winter values' may occur due to a preferential water recharge during winter months, including the recharge by melting snow.

Regarding the altitudinal trend observed in the rainwater over the Mount-Lebanon range (Fig. 4B), the isotopic signal in dripwater exhibits an altitudinal trend, but with a slightly different gradient (-0.21‰ per 100 m) than the rainwater (-0.13‰ per 100 m). This is however, more significant when the dripwater isotopic signal is compared to the altitude of the infiltration basin of each cave (Fig. 5C).

CONCLUSION

The preliminary dripwater isotopic measurements and temperature conducted on four caves located in the western flank of Mount-Lebanon, revealed the following important conclusions for future speleothem-based interpretation of paleoclimate changes at both local and regional scales:

- Despite for some water samples influenced by evaporative processes, the drip water exhibits isotopic values in agreement with the local rainwater. Therefore, stalagmites for paleoclimatic reconstructions (or fluid inclusion analysis) should be preferentially chosen outside a possible ventilation-influenced area of the cave.
- The altitudinal trend confirmed previously in the rainwater isotopic composition on the western flank of Mount-Lebanon is demonstrated also in cave drip water indicating the transfer to the cave through the vadose zone of the spatial isotopic signals of the rainwater. The isotopic composition of the dripwaters, however, exhibits a slightly higher negative $\delta^{18}O$/100 m gradient for cave drip water due to slower infiltration of winter waters. The isotopic dripwater signal represents therefore mostly a lower limit of the isotopic signal of the corresponding rain/snow melt.
- The results of this study can further help in the interpretation of past altitudinal trends based on speleothems. Additional future cave water and calcite monitoring with automated logging equipment (pCO_2, temperature, humidity, etc.) will continue to refine the interpretations that have been based on the initial monitoring findings presented here.
- To build further on this study, the altitudinal trend signal should be confirmed by modern calcite from the same caves, in which the trend should have a similar gradient.

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