L.M. Boop, J.G. Wynn, G. Thompson, J.J. Fornós, and B.P. Onac – Interactions between surface conditions, the Mediterranean Sea, and cave climate within two littoral caves in Mallorca: implications for the formation of phreatic overgrowths on speleothems. *Journal of Cave and Karst Studies*, v. 79, no. 1, p. 59–72. DOI: 10.4311/2016ES0114

INTERACTIONS BETWEEN SURFACE CONDITIONS, THE MEDITERRANEAN SEA, AND CAVE CLIMATE WITHIN TWO LITTORAL CAVES IN MALLORCA: IMPLICATIONS FOR THE FORMATION OF PHREATIC OVERGROWTHS ON SPELEOTHEMS

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Abstract: Phreatic overgrowths on speleothems from Mallorca's littoral caves are valuable markers of former sea-level stands. These carbonate encrustations form as CO_2 degasses from brackish cave water that is hydrologically connected to the Mediterranean Sea. This study uses time series analysis to document relationships between surface conditions of temperature, barometric pressure, precipitation, tidal level of the Mediterranean Sea, and the coastal caves' microenvironment of temperature, partial pressure of CO_2 (pCO_2), and water level to contextualize overgrowth formation in Cova des Pas de Vallgornera (Vallgornera) and Coves del Drac (Drac). Water level in both caves was an attenuated semidiurnal function of Mediterranean Sea level with a lag of about four hours. The impact of individual rainfall events on cave water level was negligible during the study period. pCO_2 of cave air at both sites reached an annual maximum in September and decreased rapidly when surface air temperature fell below the cave air temperature (mean ~19 °C). As this threshold was reached, cooler and denser tropospheric air descended into the caves, initiating cave ventilation and displacing high-pCO_2 cave air that had accumulated. Observed pCO_2 was lower in Drac than in Vallgornera, and had small daily fluctuations because of bigger passages, fewer constrictions, and a large collapse entrance. The frequency and magnitude of pCO_2 fluctuations were higher in Vallgornera than in Drac, with both caves showing twice-daily water level maxima causing displacement of high-pCO_2 air from the cave alternating with water level minima causing tropospheric air to enter the cave via a "piston effect." A secondary control on pCO_2 variation can be attributed to variation in tropospheric barometric pressure. Thus, the geochemical conditions favorable for overgrowth formation are, in part, the result of this tidally-controlled cycle of cave-water level. The cycle causes cave-troposphere air exchange that drives CO_2 degassing, and therefore, the formation of phreatic overgrowths on speleothems.

INTRODUCTION

Mallorca's diverse Quaternary geomorphological features have made the island a world-renowned location for Quaternary sea level research (Ginés et al., 2012a). Coastal features including wave cut notches, marine terraces, fossil assemblages, and beach deposits document former sea level stands (Butzer, 1962; Butzer & Cuerda, 1962, Goy & Zazo, 1986; Hearty et al., 1986; Hearty, 1987; González-Hernández et al., 2001). The littoral caves of Mallorca host carbonate encrustations known as phreatic overgrowths on speleothems (POS) that have been used to refine the western Mediterranean Sea eustatic curve (Vesica et al., 2000; Tuccimei et al., 2006; Dorale et al., 2010; Ginés et al., 2012b). POS are robust sea level proxies because they provide precise geographic location, elevation, age, and tidal range (Ginés & Ginés, 1972; Pomar et al., 1979; Ginés & Ginés, 2007; van Hengstum et al., 2015). POS form when CO_2 degasses at the air-water interface (Pomar et al., 1976, 1979; Csoma et al., 2006; Boop et al., 2014); they are widespread in Mallorca, with over 30 identified POS paleolevels from 46 m above to 23 m below current sea level (Ginés, 2000). Some of the POS observed above the water table have been correlated with fossil marine terraces and beach deposits (Cuerda, 1975; Pomar and Cuerda, 1979). POS precipitated by former sea level stands are generally left intact within the stable cave environment, whereas beach deposits may be reworked and terraces or wave cut notches may be removed or overprinted by subsequent transgressions. In addition, encrustations that correspond to former sea level lowstands are preserved and accessible,

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whereas surface geomorphological evidence of lowstands is often inaccessible or destroyed by surficial processes.

Studies on the mechanisms of POS formation have focused on their distribution and mineralogy (Pomar et al., 1979; Csoma et al., 2006; Ginés et al., 2012a). However, the interactions of the subterranean environment with external forcings, including surface air temperature, precipitation, barometric pressure, tide, and sea level have not been quantified in high temporal resolution. This study uses time series analysis on data collected *in-situ* to investigate the effect that external forcings have on the subsurface air and water temperatures, partial pressure of CO_2 (pCO_2), and cave water level to address the following research questions: what are the effects of rainfall and tidal fluctuations on cave water level and to what extent is cave air CO_2 controlled by atmospheric forcings like barometric pressure, temperature, or tidal pumping?

METHODS

SITE CHARACTERISTICS

The island of Mallorca is located in the western Mediterranean and is the largest of the Balearic Islands. The geological structure of Mallorca consists of a set of northeast-southwest trending horsts and grabens formed during the middle-upper Miocene. Horsts correspond to the mountain ranges structured as a thrust and fold belt during the Alpine Orogeny, the Tramuntana range to the west and the gentler Llevant ranges in the east, and the grabens correspond to the basins, Es Pla and Migjorn among others, where horizontal upper Miocene carbonate platforms crop out (Fornós et al., 2002; Sàbat et al., 2011). This extensional event responsible for Mallorca's present day topography occurred mainly in the upper Miocene; since this event, Mallorca has remained relatively stable in terms of vertical movements (Just et al., 2011).

The climate of Mallorca is typical of the Mediterranean, with hot, dry summers and mild winters. Mean annual temperature is 16.6 °C (Guijarro, 1995), and yearly rainfall totals are highly variable, ranging from 300 mm in the central and south-eastern part to 1400 mm along the Tramuntana range (Ginés et al., 2012b). The coastal areas of Mallorca are considered low-energy environments characterized by semidiurnal microtides (< 25 cm). Tidal range, however, varies seasonally due to wind stress and changes in barometric pressure, but remains between 0 and 23 cm (García et al., 2000).

Two caves were selected for this study, both hosting modern phreatic overgrowths on speleothems at the present water table: Cova des Pas de Vallgornera (Vallgornera), located on Mallorca's southern coast near Cala Pi in the Llucmajor municipality, and Coves del Drac, on the eastern coast in the outskirts of Porto Cristo (Fig. 1). The two littoral caves are each within a horizontal distance of about 400 m of the coast and developed in upper Miocene calcarenites and limestones at less than 25 m below the surface (Ginés & Ginés, 2007; Ginés et al., 2014). Vallgornera has 74 km of mapped passage that is accessed by a single nearly-sealed entrance (Merino et al., 2014). With less than 4 km of passages (Gràcia et al., 2007), Drac is the most visited cave in Europe (Robledo & Durán, 2010). Since about 3000 tourists a day spend about an hour covering 1.2 km of the cave, their respiration likely increases the cave atmosphere pCO_2. To maintain the natural conditions of temperature, relative humidity, and CO_2 concentration, the cave is artificially ventilated, further altering the natural conditions. The study area is in the non-touristic part of the cave. Therefore, any impacts of present day tourism, including altered temperature, relative humidity, and pCO_2, are minimal at the monitoring site. However, samples collected by Boop (2014) and JJF suggest that higher pCO_2 is present in non-tourist areas of the cave, likely due to the decomposition of bat guano, passage morphology, and less efficient natural ventilation in this section of the cave.

DATA COLLECTION

This study was designed to monitor the physical and chemical conditions at the air-water interface where overgrowths precipitate. Sensors were installed in both caves for almost 16 months between December 2011 and March 2013. Sample intervals varied by parameter based on the needs of resolution, sensor memory, and the field schedule. Sensor malfunctions resulted in incomplete records.

A YSI 6920 sonde was fitted into a foam float and tethered to monitor conditions at 15 cm of depth within the water column at a fixed location. Cave-water temperature (T_w) (accuracy ±0.15 °C; precision 0.01 °C) was measured at 3 hr intervals. Cave-water level (WL_c) was calculated at 15 min intervals using In-Situ Baro Merge software with water pressure from an In-Situ Aqua TROLL (accuracy ±1.05 hPa; precision 0.5 hPa), and barometric pressure (p) (accuracy ±1.5 hPa; precision 0.075 hPa) from an In-Situ Baro TROLL sensor, which also collected cave air temperature (T_c) (accuracy ±0.1 °C; precision 0.01 °C). The accuracy of the barometric-pressure-based water level is ±0.021 m. The mean was subtracted from WL_c to compare relative water level fluctuations. The Aqua TROLL sensors in both caves failed 11 months after their deployment.

A CO2meter.com K33-ELG logger (accuracy ±30 ppm; precision 20 ppm) recorded hourly CO_2. The CO_2 readings were converted to partial pressure (pCO_2) using the relationship $pCO_2 = CO_{2(\mathrm{raw})} \times 1013/p$, where $CO_{2\,(\mathrm{raw})}$ is the input data and p is in-cave barometric pressure (Spötl et al., 2005).

Additional data were obtained from governmental organizations to contextualize the cave specific data. Surface temperature (T_s) (°C; 3 hr intervals) and precipitation (P) (mm; daily totals) from the Agencia Estatal de Meteorología (AEMet) weather stations Llucmajor II (Llucmajor) and Manacor-Poliesportiu (Manacor) were the closest available to Vallgornera and Drac, respectively. To determine maxima, minima, and lags during 2012, the daily averages of cave air

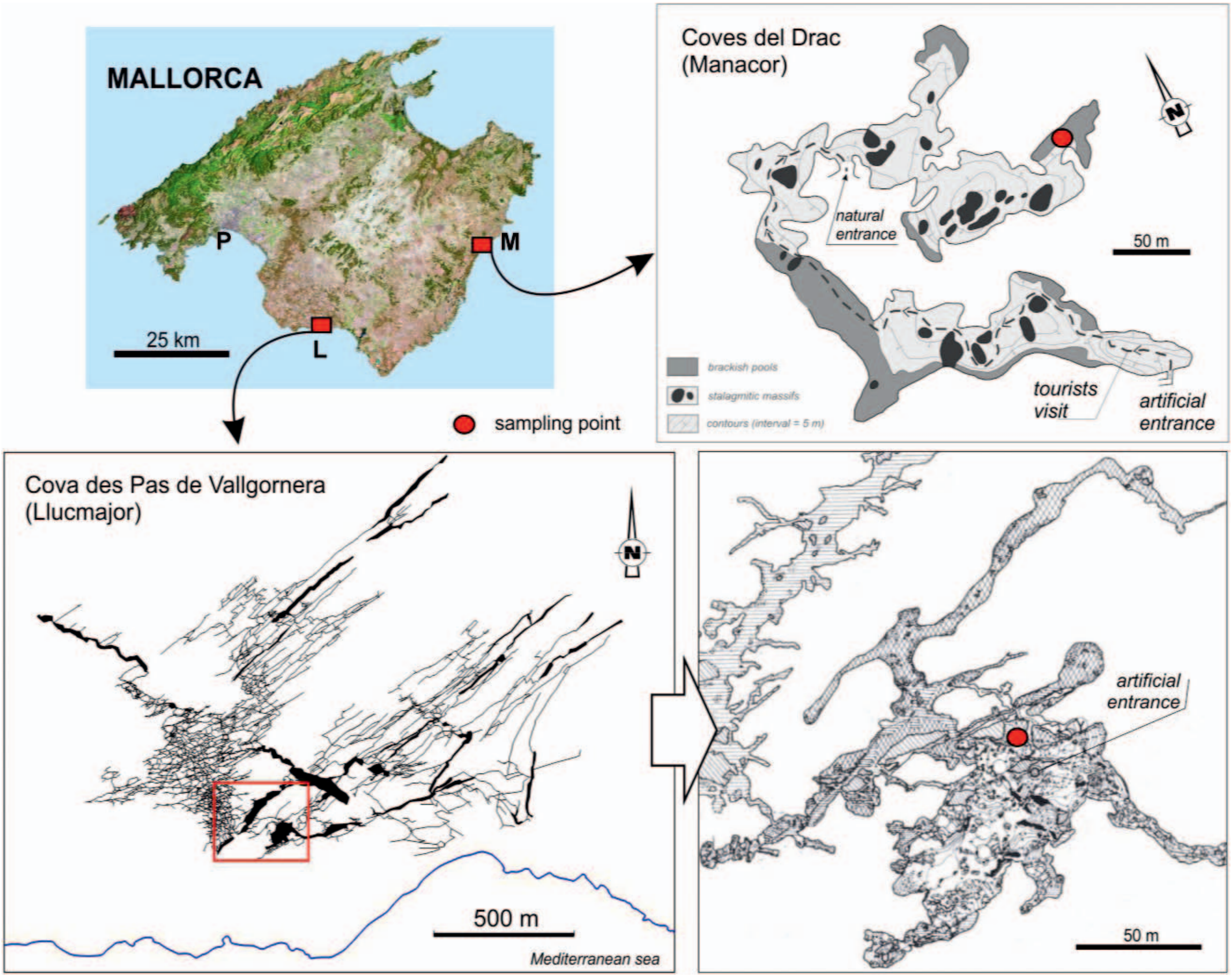

Figure 1. Map of Mallorca and caves studied. Palma Sea Level Monitoring Facility is indicated by P; Llucmajor II AEMet and the Manacor weather stations are indicated with L and M, respectively. The map of Drac is modified after Ginés and Ginés (2007) and those of Vallgornera are modified from Merino et al. (1993, 2014).

Table 1. Maxima, minima, and averages for temperature (T) and pCO_2 during 2012. The average surface temperature (T_s), cave air temperature (T_c), cave water temperature (T_w), and pCO_2 were computed for each day. This daily data was smoothed using a 7 day moving-average filter. pCO_2 averages were excluded due to missing data.

	Vallgornera				Drac			
Parameter	T_w, °C	T_c, °C	T_s, °C	pCO_2, ppm	T_w, °C	T_c, °C	T_s, °C	pCO_2, ppm
2012 Maximum	19.69	19.44	27.82	2,328	18.51	18.43	26.86	1,072
	9/12/12	10/19/12	8/11/12	9/23/12	11/19/13	12/24/12	8/11/12	8/28/12
2012 Minimum	19.23	18.99	6.05	1,007	18.45	18.31	6.34	399
	4/17/12	4/15/12	2/8/12	12/18/12	5/22/12	5/7/12	2/7/12	1/6/12
Study Average	19.4	19.2	15.6	···	18.5	18.4	15.5	···

and cave water temperatures, surface temperatures, and pCO_2 were computed, and then smoothed with a 7 day moving average filter. Finally, absolute tide gauge level (WL_P) from the Palma de Mallorca UNESCO-IOC Sea Level Station Monitoring Facility (2016) was downloaded in 15-min intervals that corresponded with the measurements of the cave-water level.

TIME SERIES ANALYSIS

Time series analysis was used to evaluate temporal trends in data collected at regular intervals. Analyses presented below were computed using commands in the Signal Processing Toolbox of Matlab 2013a. Frequency-domain figures are terminated below the Nyquist frequency, equal to half of the sampling frequency.

The relationship between tide gauge and cave water levels at each cave was investigated using coherency analysis. To examine controls on cave pCO_2, coherency analysis was also used to investigate barometric pressure, cave and surface temperatures, and water level in the caves. Coherency is a function of the power-spectral densities of each time series, and their cross-power spectral density. Coherency measures how well correlated two time series are as a function of frequency, with values between 0 (incoherent) and 1 (perfectly coherent). It also measures the phase angle, ψ, between these time series as a function of frequency. Thus if there is a strong coherency at a particular frequency, both time series must have significant energy at that frequency, and from ψ the time lag in hours can be determined. Magnitude-squared coherency estimates were computed using the MSCOHERE Matlab command, using a Hamming window of length 256 samples with an overlap of 128 samples.

Spectrograms are computed to investigate the temporal behavior of periodic signals. To identify high-frequency pCO_2 fluctuations, a stationary time series was created: the annual signal was removed by subtracting a 20-point (20-hr) moving average. Spectrograms were computed using the SPECTROGRAM Matlab command, a window length of 24 samples, an overlap of 12 samples, and a 2048-sample fast Fourier transform.

RESULTS

METEOROLOGICAL DATA

Surface temperature and precipitation show similar patterns at both sites (Fig. 2). During the study period, mean T_s was 15.6 °C at Llucmajor and 15.5 °C at Manacor. Relative to T_s, which displays annual and daily fluctuations, cave air temperatures were nearly stable within 0.5 °C (Figs. 2A–2C). Total precipitation P differed between sites, with 485.4 mm recorded at Llucmajor (Fig. 2A) and 392.2 mm at Manacor (Fig. 2B).

CAVE AIR AND WATER TEMPERATURE

Over the study period, mean water temperature in Vallgornera was 19.4 °C, whereas mean cave air temperature was 19.2 °C (Table 1), and T_w was always greater than T_c (Fig. 2C). During 2012, maximum T_c lagged that of T_w by 37 days (Table 1). In contrast, minimum T_c lagged that of T_w by two days.

During the study, mean T_c and T_w in Drac were 18.4 and 18.5 °C, respectively, with ranges of only 0.2 and 0.15 °C, respectively (Table 1). These values are similar to spot measurements recorded in the touristic part of the cave by BPO, where temperatures would rise well above 21 °C when large groups (about 500 people) visited the cave and when forced-air ventilation was not in operation.

As in Vallgornera, T_w in Drac was always greater than T_c (Fig. 2C). The range of T_w was lower in Drac, making it difficult to determine maxima and minima and determine lag times. During 2012, the maximum T_c in Drac lagged T_s by 135 days; the same lag in Vallgornera was only 69 days (Fig. 2; Table 1). Maximum T_w in Vallgornera followed maximum T_s after 32 days. Minimum T_c and T_w in Vallgornera lag T_s by 67 and 69 days, respectively. Minimum T_c and T_w in Drac were recorded 90 and 105 days after minimum T_s, respectively.

CAVE pCO_2

In both caves, peak pCO_2 occurred in August/September (Table 1, Fig. 2D). pCO_2 in Vallgornera was higher than in Drac and showed greater amplitude of variation on annual and daily scales. The time between the highest and lowest recorded pCO_2 values was 157 days in Vallgornera, and only 121 days in Drac.

There is significant surface temperature $T_s - pCO_2$ coherency in Vallgornera (Table 2) at a 12 hr period. The lag time of the January – April 2012 data was 4 hr, while that of the May 2012 – March 2013 was 4.8 hr. An 8 hr period is also present in the Vallgornera data; the lag is 3.9 hr in the January – April 2012 data and 3.2 hr in the May 2012 – March 2013 series. No notable T_s - pCO_2 relationship is observed in Drac (Table 2).

The cave water levels in Vallgornera and Drac are shown in Fig. 3A. $WL_c - pCO_2$ coherency is present in Vallgornera at the 11.6hr period (Fig. 4A, Table 2). Lead times are 4.4 hr for the January – April 2012 time series, and 2.7 hr for the July – November 2012 data (Fig. 4B). $WL_c - pCO_2$ coherency is also present at a 12.8 hr period with a 5 hr lead in the January – April 2012 data from Drac (Fig. 4C and D), but no cycles were identified in the July – November 2012 dataset.

Barometric pressure $p - pCO_2$ coherency is present at a 12.2hr period in Vallgornera, with lead times of 5.3 hr in the January – April 2012 data and 4.5 hr in the July – November 2012 dataset (Table 2). Drac $p - pCO_2$ coherency analysis identified an identical 12.2 hr period in the July – December 2012 series and a 12.8 hr period in the January – March 2013 data. The corresponding lead times are 9 min and 5.5 hr, respectively. A similar $p - pCO_2$ coherency peak is not present in the January – April 2012 data from Drac.

Table 2. Notable peaks from magnitude squared coherency analyses for surface temperature–$p\mathrm{CO}_2$ (T_s–$p\mathrm{CO}_2$), barometric pressure–$p\mathrm{CO}_2$ (p–$p\mathrm{CO}_2$), and absolute tide gauge level – cave-water level ($\mathrm{WL}_P - \mathrm{WL}_c$).

	Vallgonera							Drac						
Parameter	Date Range	Coherency	*f*, Hz	Period, hr	*Ψ*, rad	Lead, hr	Lag, hr	Date Range	Coherency	*f*, Hz	Period, hr	*Ψ*, rad	Lead, hr	Lag, hr
$\mathrm{T}_s - p\mathrm{CO}_2$														
	01–2012 – 04–2012	0.91	2.32×10^{-5}	12.0	–2.10		4.0	12–2011 – 04–2012						
		0.66	3.47×10^{-5}	8.0	–3.04		3.9							
	05–2012 – 03–2013	0.32	2.32×10^{-5}	12.0	–2.53		4.8	07–2012 – 12–2012	0.34	2.24×10^{-5}	12.4	0.63	1.3	
		0.4	3.47×10^{-5}	8.0	–2.48		3.2		0.38	2.39×10^{-5}	11.6	1.21	2.2	
								01–2013 – 03–2013	0.74	1.88×10^{-5}	14.8	–3.12		7.3
									0.35	2.46×10^{-5}	11.3	1.86	3.4	1.9
									0.35	2.60×10^{-5}	10.7	–1.11		
$\mathrm{p} - p\mathrm{CO}_2$														
	01–2012 – 04–2012	0.97	2.28×10^{-5}	12.2	2.74	5.3		01–2012 – 04–2012						
		0.57	2.82×10^{-5}	9.8	2.47	3.9								
		0.75	3.15×10^{-5}	8.8	2.50	3.5								
		0.87	3.47×10^{-5}	8.0	2.51	3.2								
		0.73	4.12×10^{-5}	6.7	2.18	2.3								
		0.81	4.56×10^{-5}	6.1	1.74	1.7								
		0.65	6.94×10^{-5}	4.0	1.22	0.8								
	05–2012 – 03–2013	0.62	2.28×10^{-5}	12.2	2.33	4.5		07–2012 – 12–2012	0.38	2.28×10^{-5}	12.2	0.008	0.1	
		0.32	3.47×10^{-5}	8.0	2.26	2.9			0.24		8.0	–1.71		2.2
									0.23		5.6	–2.85		2.5
								01–2013 – 03–2013	0.26	1.74×10^{-5}	16.0	1.81	4.6	
									0.28	2.17×10^{-5}	12.8	2.69	5.5	2.5
									0.24	2.50×10^{-5}	11.1	0.72	1.3	0.2
									0.23	2.82×10^{-5}	9.8	1.14	1.8	0.1
									0.37	3.91×10^{-5}	7.1	–2.23	0.8	
									0.36	5.64×10^{-5}	4.9	1.08		
									0.38	9.98×10^{-5}	2.8	–0.47		
									0.45	1.17 × 10–5	2.4	–0.15		
$\mathrm{WL}_P - \mathrm{WL}_c$														
	12–2011 – 11–2012	0.94	1.09×10^{-5}	25.6	–1.19		4.9	12–2011 – 11–2012	0.92	1.09×10^{-5}	25.6	–0.96		3.9
		0.96	2.17×10^{-5}	12.8	–1.98		4.0		0.96		12.8	–1.75		3.6

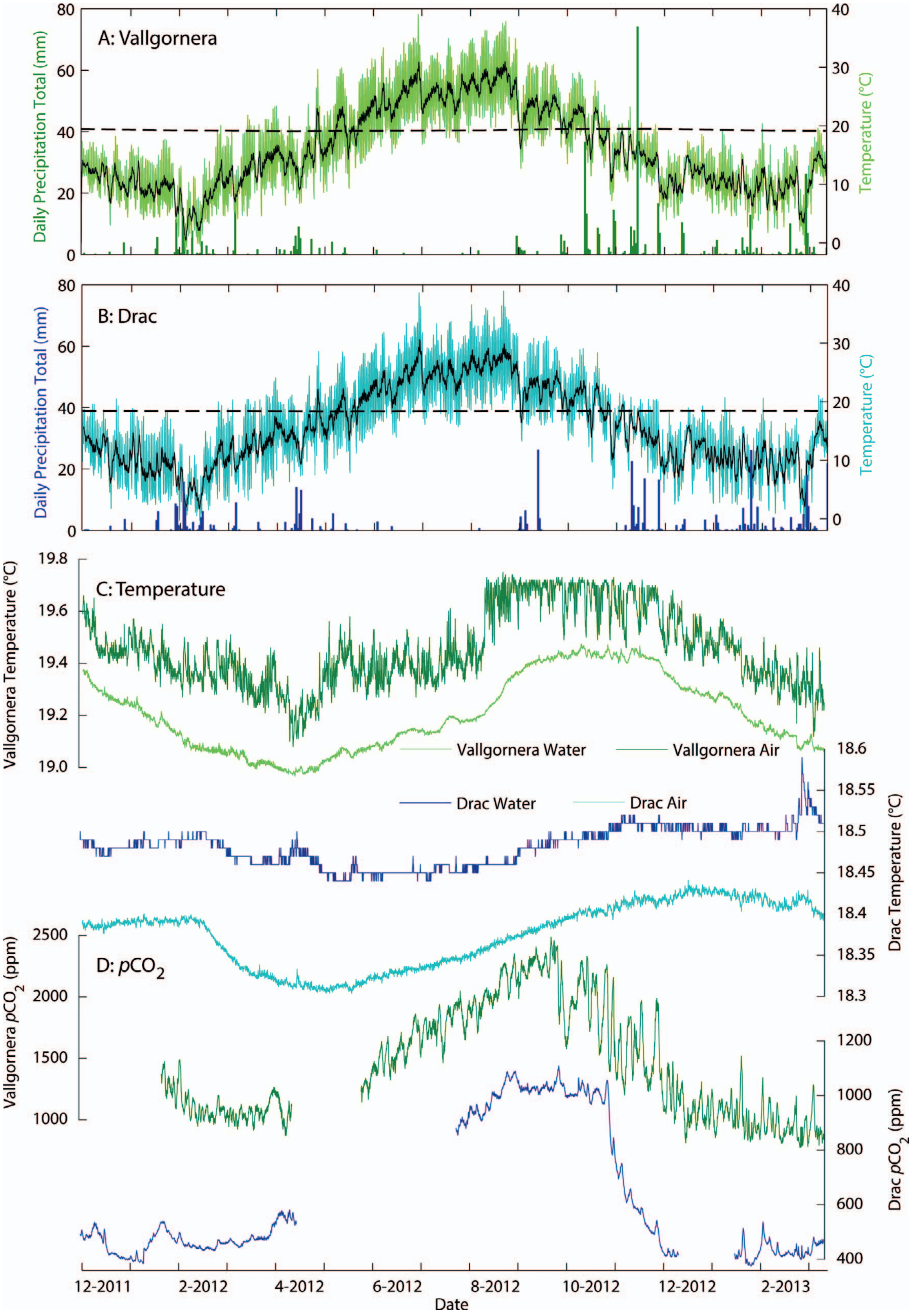

Figure 2. A) Surface temperature (T_s) and daily precipitation (P) from AEMet Llucmajor II and B) Manacor stations. Right axis: T_s (black line shows a 10-point moving average). Black dashed lines show cave air temperature (T_c) for Vallgornera (A) and Drac (B) from part C. C) Detailed cave air temperature (T_c) and water temperature (T_w) and D) cave pCO_2 (corrected for cave barometric pressure) from Vallgornera and Drac.

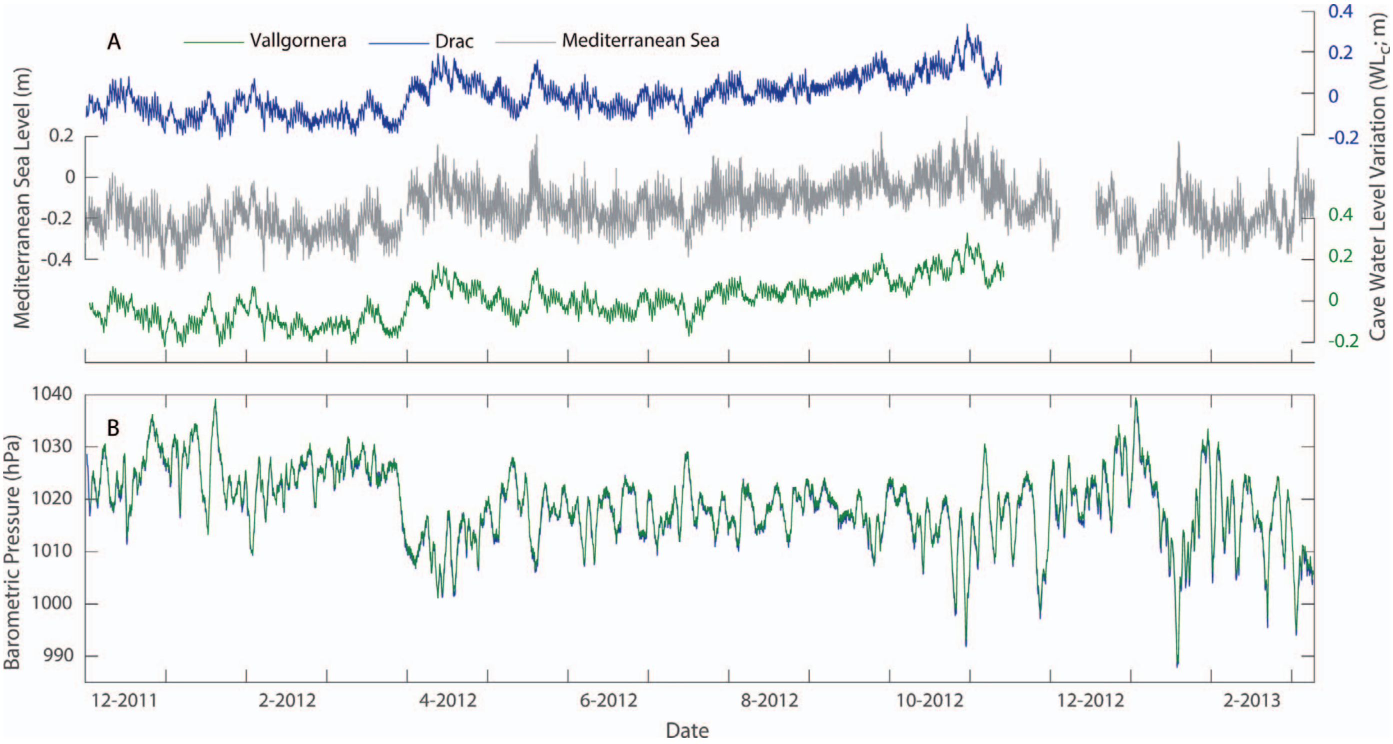

Figure 3. A) Cave water levels (WL_c) and Mediterranean Sea Level from tide gauge (WL_p). B) Barometric pressure (p).

pCO_2 records are reproduced next to the spectrograms of filtered data for both Vallgornera and Drac in Fig. 5. High-frequency pCO_2 fluctuations in Vallgornera (Fig. 5A) create spectral power at the 12-hr period in Vallgornera (Fig. 5B). In Drac, which has lower-amplitude pCO_2 fluctuations (Fig. 5C), lower spectral power exists at the 12 hr cycle at some points throughout the year (Fig. 5D).

CAVE WATER LEVEL

The cave water level WL_c is similar in both caves (Fig. 3A), and both records show a similarity to the tide gauge water level WL_P. The strongest $WL_P - WL_c$ coherency peak (>0.96) in both caves corresponds to a 12.8-hr period, and the second strongest peak (>0.92) occurs at a 25.6 hr period (Fig. 6, Table 2). In Vallgornera, the 12.8-hr WL_c cycle lags WL_P by 4.0 hr, whereas the 25.8-hr cycle lags by 4.9 hr. The corresponding lag in Drac of the 12.8-hr cycle was 3.6 hr, while that of the 25.6-hr cycle was 3.9 hr.

The results from the coherency analyses indicate a period close to, but not matching the lunar day (24.8 hr). However, strong similarity between the cave (WL_c) and tide gauge (WL_P) water levels is clearly seen when the data are considered in the time domain (Fig. 7). Data from July 20 – 23, 2012 was chosen because no rainfall was recorded for $>$ 30 days before this interval. The discrepancy between results in the frequency and time domains is a consequence of the 3-hr sample interval of the coherency analyses; the 24.8-hr lunar day signal rounds up to the closest possible frequency output (25.6 hr). The same discrepancy applies to the mixed semi-diurnal 12.4-hr cycle that is output as strong coherency at 12.8 hr.

Over the entire study period, there was strong positive linear correlation between WL_P and WL_c in both caves. In Vallgornera, the r-value from Pearson's correlation coefficient for WL_P and WL_c was 0.88 (slope $y = 0.82x + 1.41$, $n =$ 32,372), while that of Drac was 0.87 (slope $y = 0.76x + 1.71$, n $=$ 32,419). WL_P ranges 0.76 m, whereas the ranges in WL_c at Vallgornera and Drac are almost identical, 0.55 and 0.56 m, respectively (Fig. 3A). Thus, over the study period, the attenuation of WL_P is approximately 0.2 m at both sites.

DISCUSSION

CAVE AIR AND WATER TEMPERATURES

While it is generally accepted that cave temperature is a good approximation of the annual average surface temperature in the cave area (Wigley & Brown, 1976), over the study period, mean cave T_c and cave water T_w temperatures in both caves are approximately 3 °C greater than mean surface temperature during the same time interval. This observation could be explained by either differences in cave ventilation regimes or the heat exchange between seawater and cave atmosphere due to each cave's hydrologic connection to the Mediterranean Sea. The higher T_c and T_w in Vallgornera compared to Drac may be due to the cave's proximity to a region known to host a deep thermal reservoir (López-García & Mateos-Ruíz, 2006). Merino et al. (2011) report temperatures of 27.1 °C and 23.6 °C in Cova de sa Guitarreta and Pou de Can Carro, located less than 10 km northwest of Vallgornera. Based on the presence of various hypogene features and unusual mineral assemblages, thermal basal recharge may have played a major role in the speleogenesis of Vallgornera (Ginés et al., 2009, 2014; Fornós et al., 2011; Onac et al., 2014).

pCO$_2$

Higher pCO$_2$ in Vallgornera compared to Drac may be attributed to limited ventilation in the former cave, which has a single closed entrance. A large, natural collapse entrance in Drac, as well as less-restricted cave passages, likely allows for stronger ventilation, maintaining lower pCO$_2$ in cave air (Ek & Gewelt, 1985; Hu et al., 2008).

In both caves, pCO$_2$ increases in the summer months, with the highest concentration observed at the end of the growing season. Similar findings are reported in Banner et al. (2007), Liñán et al. (2008), Faimon et al. (2012), Cowan et al. (2013), Mandić et al. (2013), and Lang et al. (2016). The pCO$_2$ plateau noticed in Drac between August and early November is due to this cave's particular air circulation. Annual ventilation is driven by the disparity between surface and cave temperatures during winter, when cool, relatively dense air descends into cave passages, forcing the CO$_2$-rich air out of the cave. Throughout the summer months, the outside temperatures exceed those inside the cave causing the ventilation to slow down or cease, allowing CO$_2$ to build up. Dumitru et al. (2015) confirmed this bi-directional ventilation regime in Drac by deploying radon sensors throughout the non-touristic branch of the cave, including one detector at the same site investigated in this study (Fig. 1). In Vallgornera and Drac, this exchange is sufficiently slow that rapid, high-amplitude decreases of T_c do not coincide with the seasonal decrease of pCO$_2$ (Fig. 2). Instead, the thermal signal is buffered between the entrances and the study sites. It is likely that the network of fractures in the overlying bedrock at Vallgornera plays a major role in the seasonal overturn, whereas in Drac, this ventilation occurs through the cave's large natural entrance.

The calculated surface temperature $T_s - p\mathrm{CO}_2$ coherency values and corresponding lag times suggest the possibility of overnight thermal- and density-driven ventilation in Vallgornera, but there is no peak in spectral coherency at a 24 hr period (Table 2). Additionally, during the summer months when T_s consistently exceeds cave temperature T_c, thermal cave gradients could not drive ventilation and high-frequency pCO$_2$ variations.

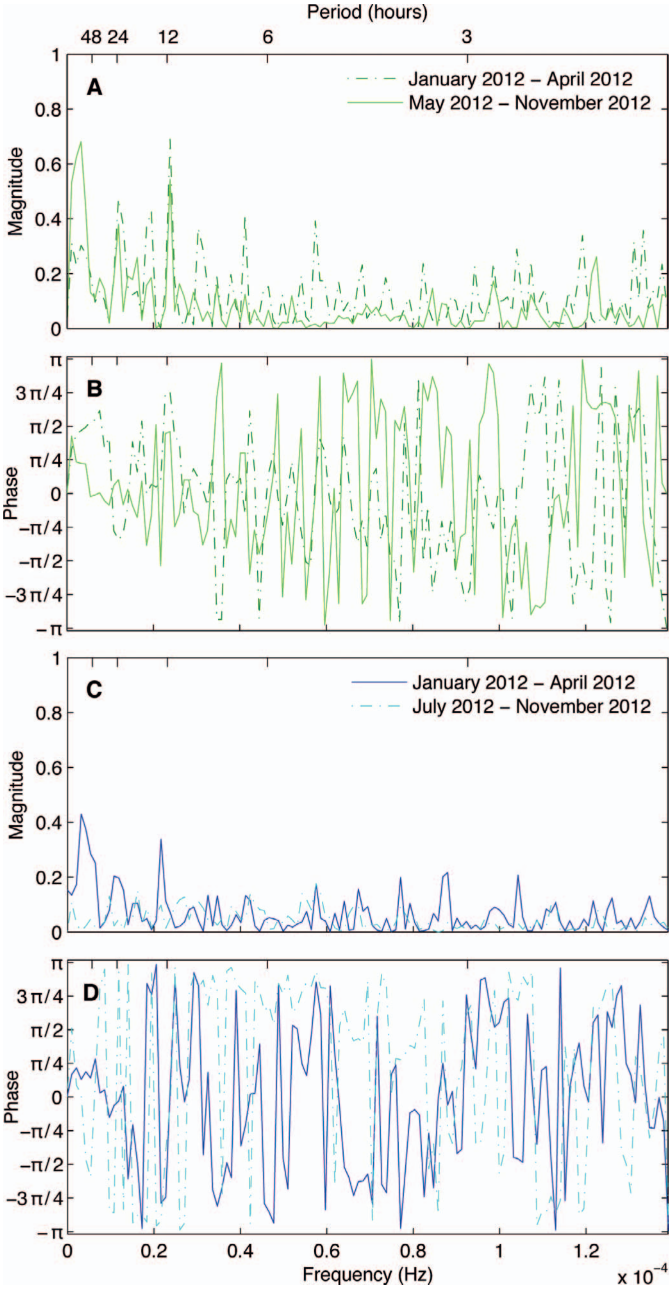

Figure 4. Cave water level $\mathrm{WL_C} - p\mathrm{CO}_2$ coherency for Vallgornera and Drac (A and C) and the corresponding phase angles (B and D). Top axis labels: time domain; bottom axis: frequency domain.

pCO$_2$ can be controlled by cave-barometric pressure p in some caves. For example, Baldini et al. (2006) report nightly ventilation due to p minima. Cowan et al. (2013) present diurnal fluctuations that correspond to p as global atmospheric tide in caves in Texas, USA. Similarly, Benavente et al. (2011) suggest that p may drive cave-air circulation during summer, but state that their data are insufficient to support the hypothesis. $p - p\mathrm{CO}_2$ coherency was found throughout the year in both Vallgornera and Drac in approximately 12-hr cycles (Table 2). The different lead times do not support the potential relationship identified by $p - p\mathrm{CO}_2$ coherency values, because p should exert a constant control irrespective of surface temperature and other variables. Further, the lack of replication of $p - p\mathrm{CO}_2$ coherency values in Drac between the January – April 2012 data and the January – March 2013

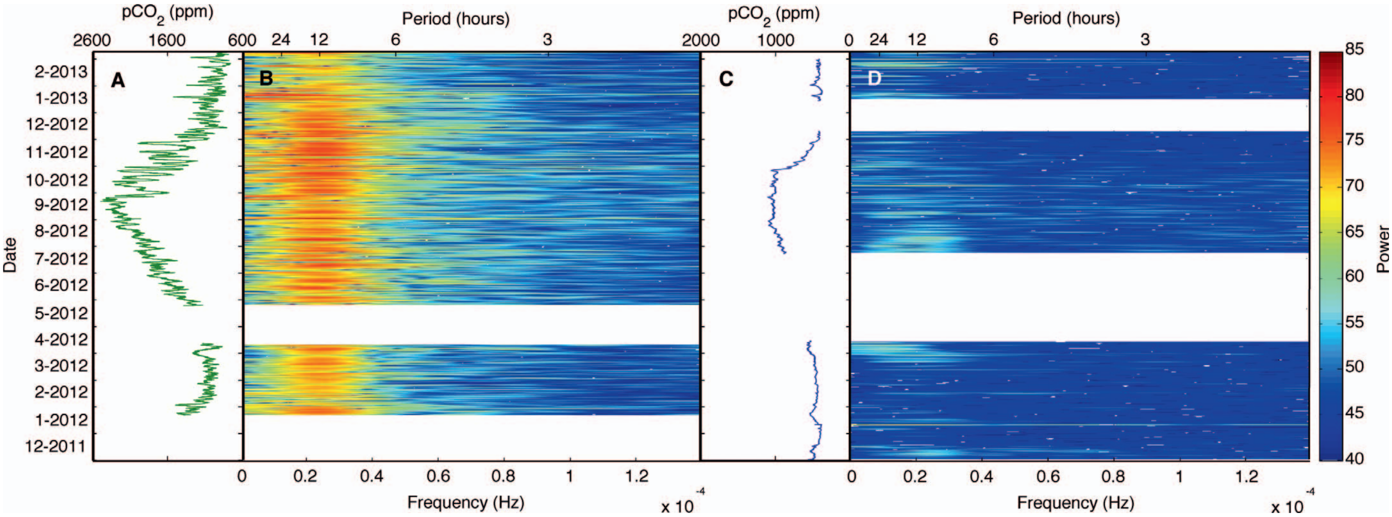

Figure 5. Vallgornera and Drac $p\mathrm{CO}_2$ time series (A and C) and their spectrograms (B and D).

series indicates that further work is necessary to evaluate the control of p on $p\mathrm{CO}_2$ fluctuations, at least in Drac. $p - p\mathrm{CO}_2$ coherency values and consistent period and lead times in Vallgornera support the hypothesis that $p\mathrm{CO}_2$ responds to p in this near closed-system cave, but it is not the primary control.

Cave water level $\mathrm{WL}_c - p\mathrm{CO}_2$ coherency values support the hypothesis that WL_c affects $p\mathrm{CO}_2$ throughout the year in Vallgornera and to a lesser extent during the summer months in Drac (Fig. 4A and C, Table 2). This is reinforced by the spectral power observed at approximately 12 hr in Vallgornera (Fig. 5B). These observations suggest that the transition from low- to high-water level forces higher-$p\mathrm{CO}_2$ air from the cave (Fig. 8A). Regardless of surface temperature, as the water recedes, tropospheric air sinks into the cave, with a volume equivalent to the volume reduction of seawater in the cave (a function of water level WL_c) (Fig. 8B). Significant cave temperature variation was not observed at either study site; any external thermal signal is attenuated.

In Vallgornera, a forward and reverse piston effect seems to dominate semidiurnal cave air exchange through the overlying bedrock, which is less than 20 m thick (Fig. 8). Twice-daily $p\mathrm{CO}_2$ variations on the order of 300 ppm were recorded throughout the year. It is possible that in Vallgornera's extensive horizontal network of passages with limited vertical extent, the sensor recorded the movement of a boundary layer of CO_2 maintained by degassing from the water (Badino, 2009), though no data are currently available to support or refute the presence of such a boundary layer at this site.

Exchange through the entrance likely dominates in Drac (Fig. 8). In addition to maintaining lower $p\mathrm{CO}_2$, Drac's large passages and few constrictions allow for cave water level driven exchange through its large entrance, which explains the lower amplitude of semidiurnal fluctuations that are only recorded during the summer months, when large-scale temperature-driven ventilation is inactive. Compared to Vallgornera, the Drac study room has greater volume, thus the amplitude of recorded $p\mathrm{CO}_2$ fluctuations may be much smaller due to the homogenous cave atmosphere maintained by temperature- and density-driven convection currents. More in-depth spatial (horizontal and vertical) $p\mathrm{CO}_2$ sampling is necessary to better constrain the dynamics of cave troposphere $p\mathrm{CO}_2$ exchange in both caves.

CO_2 degassing from the cave water is an essential process for the formation of phreatic overgrowths on speleothems (Pomar et al., 1976, 1979; Csoma et al., 2006) and is driven by the gradient between $p\mathrm{CO}_2$ in the cave water and air (Boop et al., 2014). The lower overall $p\mathrm{CO}_2$ in Drac allows for periods of degassing that cause water to become supersaturated with respect to calcium carbonate, as evidenced by ephemeral rafts of calcite observed throughout the year. Conversely, relatively high $p\mathrm{CO}_2$ in Vallgornera is not presently conducive to extended periods of rapid CO_2 exsolution. Thus, the conditions for supersaturation with respect to carbonate minerals and subsequent precipitation of POS are not currently satisfied (Boop et al., 2014). Based on the U/Th dates from a Vallgornera POS sample, these unfavorable geochemical conditions may have started 600 years ago (Tuccimei et al., 2009, 2010).

CAVE WATER LEVEL

The attenuation of the tide gauge water level WL_P inside the littoral caves and the strong relationship between variation in it and the cave level WL_c, illustrated by very high coherency and robust linear correlations, further confirm that POS are excellent recorders of mean sea-level position.

Though WL_c fluctuates with a roughly 4-hr lag behind WL_P, there is limited evidence of direct Mediterranean Sea water intrusion during this tidally-driven process (Boop et al., 2014). Cave water temperatures T_w fluctuated less than 1 °C

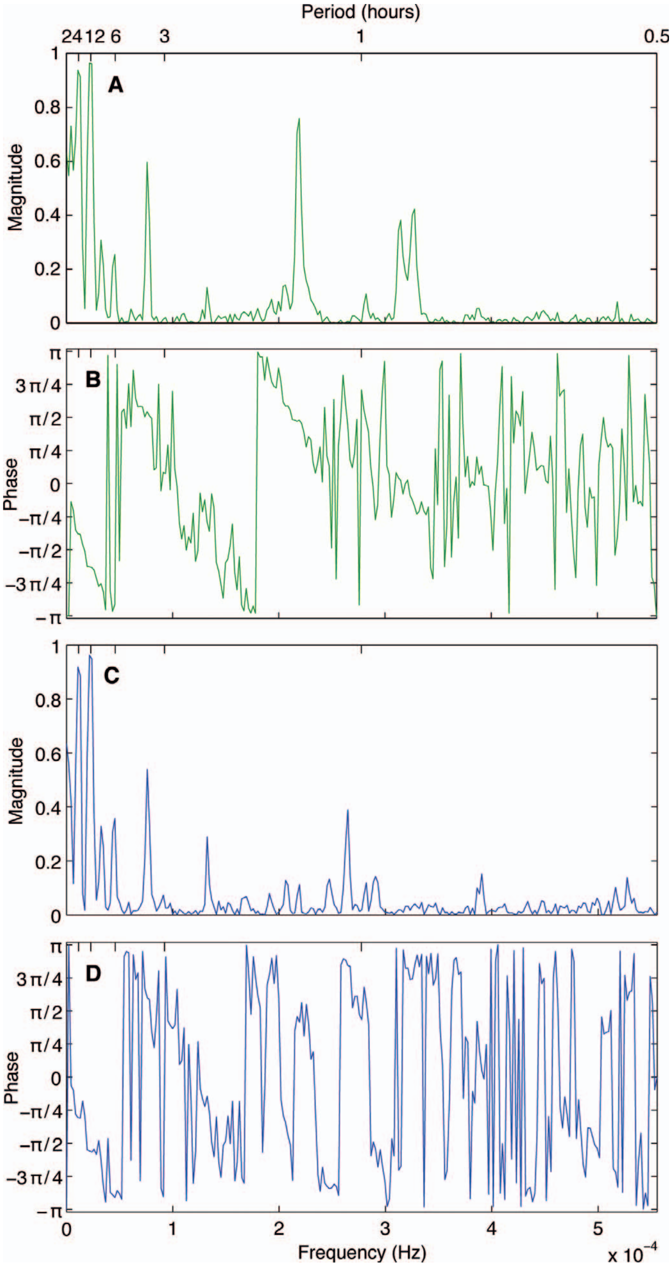

Figure 6. Vallgornera tide gauge level WL_P – cave water level WL_C coherency (A) and corresponding phase angles (B); same analyses for Drac (C and D). Top axis: time domain; bottom axis: frequency domain.

over the study period, with a higher range observed at Vallgornera than in Drac. Direct mixing with the Mediterranean Sea would introduce a strong annual period in T_w because of changes in seawater temperature. Lack of mixing is further supported by the low turbidity of the water, lack of observed currents, and the maintenance of brackish water at the surface of both study sites (Boop et al., 2014). The latter condition is likely maintained throughout the year by meteoric recharge.

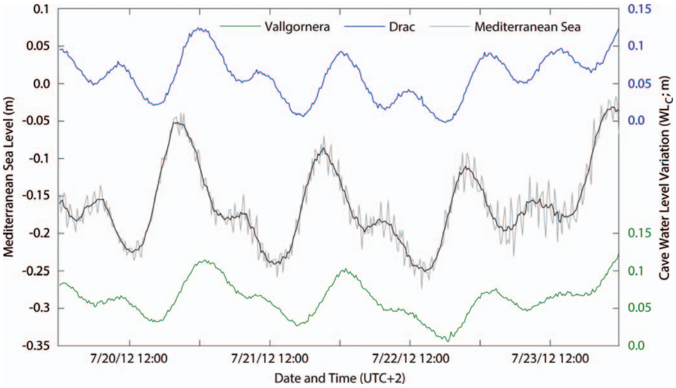

Figure 7. Expanded-scale cave water level (WL_C) variation from July 20–23, 2012, and Mediterranean Sea level (WL_P) (black line: 5-point moving average).

Time series analysis did not reveal a relationship between cave water levels and precipitation. In non-coastal areas, time series analysis can identify the effect of rainfall on karst aquifers (Padilla & Pulido-Bosch, 1995; Larocque et al., 1998; Panagopoulos & Lambrakis, 2006; Bailly-Comte et al., 2008). However, analyses based on spring discharge or piezometric head are not applicable to understand controls on water level in Mallorca's littoral caves because water level is dominantly controlled by the Mediterranean Sea level. Similarly, Kim et al. (2005) were unable to identify any effect of precipitation on coastal water levels in Korea and attribute fluctuations solely to the tidal effect using time series analysis.

CONCLUSIONS

The phreatic overgrowths on speleothems found in the littoral caves of Mallorca are valuable paleo-sea level index points. These encrustations form at the cave water table, which is, and was in the past, coincident with sea level because of the negligible hydraulic gradient between the caves and the Mediterranean Sea. The present study validates the assumption that the cave water levels fluctuate in response to the Mediterranean Sea tide by identifying an approximately 4-hr lag of WL_c behind the tide gauge level in the sea WL_P. The semidiurnal signal is preserved, though slightly attenuated, in WL_c at each study site. The primary control of WL_P masks any direct effect of P on WL_c.

The present study, Boop (2014), and Boop et al. (2014) monitored conditions relevant to the precipitation of phreatic overgrowths on speleothems. Some caves may be more likely to host the specific geochemical conditions necessary for their precipitation. These conditions are met when cave water is supersaturated with respect to carbonate minerals at times when CO_2 degasses from water into the cave atmosphere. A high concentration gradient between pCO_2 of the water and air promotes this process. Thus, faster degassing, and therefore

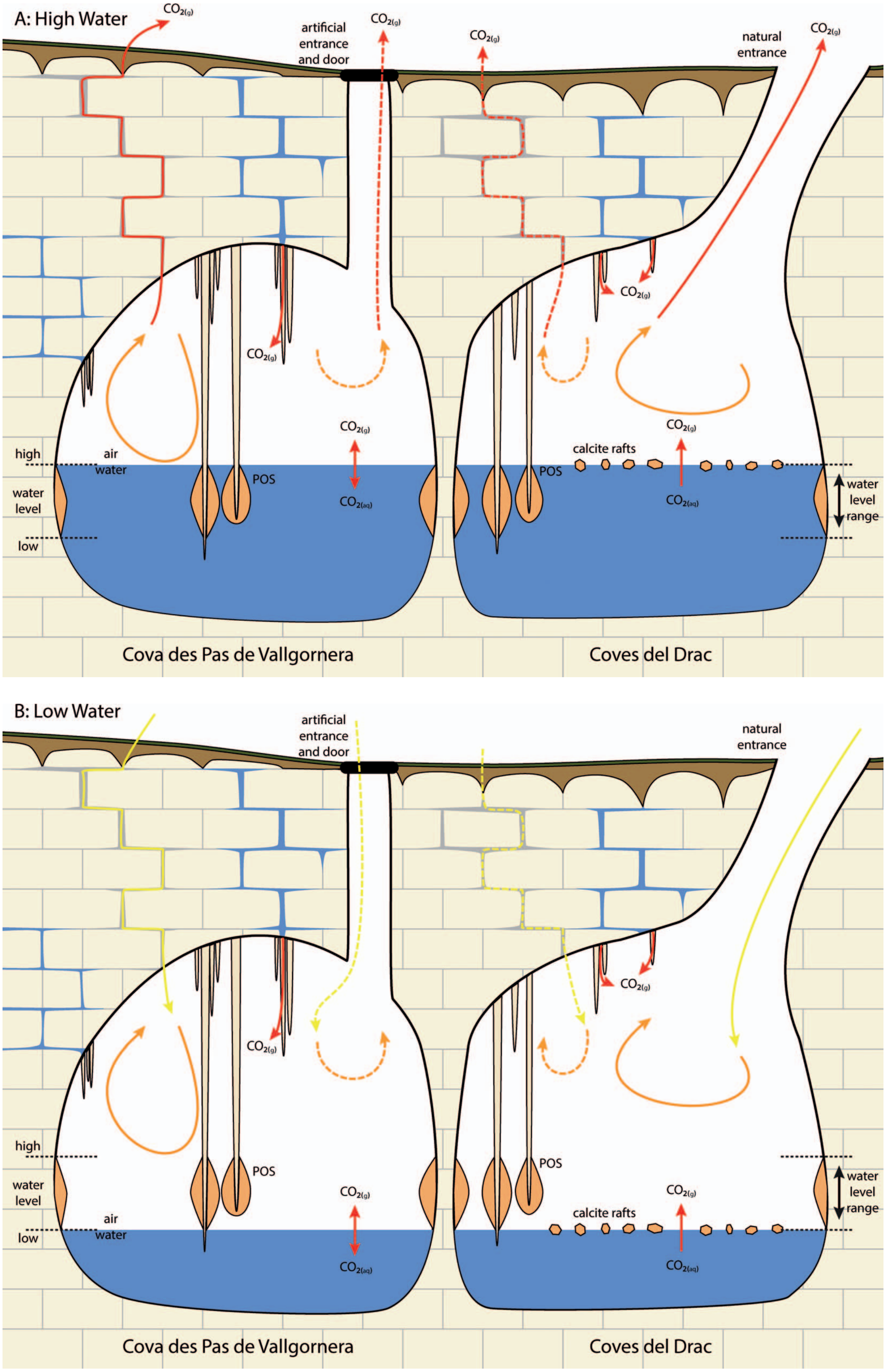

Figure 8. A simplified model of semidiurnal pCO$_2$ dynamics in Vallgornera (left) and Drac (right) associated with the rising (A) or receding (B) of the cave water level, which is overprinted on seasonal (winter) ventilation. Phreatic overgrowths on speleothems appear to be actively growing today in Drac but not in Vallgornera. See text for additional explanations.

solution supersaturation and possible precipitation of POS, is expected to occur with low cave air $p\mathrm{CO}_2$. The annual cycle in $p\mathrm{CO}_2$ is driven by cave ventilation in the winter months, when colder, denser tropospheric air sinks through the bedrock or entrances into subsurface voids, displacing cave air and reducing $p\mathrm{CO}_2$. The thermal signature of this tropospheric air is quickly attenuated within the caves; cave air and cave water temperatures vary by less than 1 °C throughout the year and lag behind surface temperature by several months. $p\mathrm{CO}_2$ fluctuations have larger amplitudes in Vallgornera, likely due to the overall higher $p\mathrm{CO}_2$ and the constricted nature of the study site. It is likely that at both sites, rising cave water level due to barometric pressure changes and tidal pumping (Fig. 8A) forces high $p\mathrm{CO}_2$ air out from the cave, driving semidiurnal tropospheric exchange throughout the year with a roughly 12-hr period. In Vallgornera, air escapes dominantly through the bedrock fissures (solid red line in Fig. 8A), whereas Drac's less-constricted passages promote displacement through its large natural entrance. When WL_c recedes (Fig. 8B), tropospheric air sinks into the cave through the bedrock in Vallgornera (solid yellow line) and primarily through the entrance in Drac. Vallgornera's nearly sealed entrance, maze-like passages, and constrictions promote the buildup of higher concentrations of $p\mathrm{CO}_2$ throughout the year, restricting CO_2 degassing from the water, thus suppressing calcium carbonate supersaturation and POS precipitation. In Drac, $p\mathrm{CO}_2$ is lower throughout the year, which maintains conditions for CO_2 to degas from the cave water to air; this promotes calcium carbonate supersaturation, and thus, precipitation of ephemeral rafts and POS.

Acknowledgments

This material is based upon work supported by the National Science Foundation under grant AGS-1103108 to BPO and MINECO projects CGL2013-48441-P and CGL2016-79246-P (AEI/FEDER, UE) to JJF. LMB received student research grants from the National Speleological Society and the Geological Society of America. AEMet is thanked for providing the temperature and precipitation records for Llucmajor II and Manacor-Poliesportiu that were used in this study. Scott McBride (USGS Tampa) is thanked for lending two YSI 6920 sensors for this study, as well as his invaluable guidance. We are grateful to Tony Merino, Marta Rodríguez-Homar, Joaquín Ginés, and Angel Ginés for their dedication in the field. We are indebted to the administration of Coves del Drac (Porto Cristo) for granting permission to perform research in the cave. We additionally appreciate the Federació Balear d'Espeleologia and the Department de Cultura i Patrimoni del Consell de Mallorca, as well as the Direcció General de d'Espais Naturals i Biodiversitat del Govern de les Illes Balears for helping with legal permissions for our work in Vallgornera. This paper benefited from discussions at the NSF- and PAGES-sponsored workshop on *Sea-level changes into MIS 5: from observation to prediction* (award AGS-1032243 to BPO). Finally, this manuscript was greatly improved through the thoughtful suggestions of three anonymous reviewers.

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