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Climate monitoring in the Caumont cave and quarry system (northern France) reveal near oxygen isotopic equilibrium conditions for carbonate deposition

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Abstract: The study of modern cave deposits forming under near isotopic equilibrium conditions can potentially help disentangle the processes influencing the oxygen isotope system and suitability of stalagmites as archives of past hydrological or thermal changes. We used cave monitoring to evaluate the impact of kinetic isotope fractionation and assess the conditions under which modern cave carbonates form in the Caumont cave and quarry system, located in Normandy, northwest France. Over 20 months, we collected climatological data, dripwater, and modern carbonate samples at 2–4-week intervals at three different stations inside the Caumont cave and quarry system. We find highly stable ($10.4 \pm 0.3 - 11.3 \pm 0.1^\circ\text{C}$) temperature in the deeper sections of the Caumont cave and quarry system. The temporal dynamics of $\delta^{18}\text{O}_{\text{drip}}$ indicates that the drip water composition in Caumont reflects the original (though subdued) signal of precipitation, rather than the impact the seasonal to interannual cave air temperature has on isotopic fractionation. The monitoring reveals that $\delta^{13}\text{C}$ of modern carbonate is influenced by prior carbonate precipitation that occurs during the summer season when evapotranspiration can minimize effective infiltration. Comparison of $\delta^{18}\text{O}$ from dripwater and modern calcite, precipitated on glass plates and collected every two to four weeks, reveals that modern calcite forms near oxygen isotope equilibrium. A Hendy test on modern carbonate deposited on a stalagmite-shaped glass flask over 20 months confirms this finding because neither does $\delta^{13}\text{C}$ increase with distance from the apex, nor are $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ positively correlated. We conclude that the $\delta^{13}\text{C}$ signal in speleothems reflect summer (and longer-term) prior carbonate precipitation in response to effective infiltration dynamics, and that the $\delta^{18}\text{O}$ signal likely reflects annual to multi-annual changes in the composition of precipitation above the cave.

Keywords: Water isotopes, modern cave calcite, PCP, climate monitoring, chalk cave, Normandy

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INTRODUCTION

In recent years, palaeoclimate studies based on speleothems have benefited greatly from the information gleaned from cave monitoring. Many studies emphasize the importance of monitoring cave environments to support interpretations on speleothem-based palaeoclimate reconstructions and focus on cave climate dynamics and the local processes and conditions that make each cave a distinctive system (Mattey et al., 2010; Tremaine et al., 2011; Breitenbach et al., 2015; Czuppon et al.,

2017; Riechelmann et al., 2011, 2019; Surić et al., 2018; Verheyden et al., 2008; Van Rampelbergh et al., 2014). Although monitoring is a powerful strategy to understand the processes that influence the environmental proxies that are recorded in stalagmites, it must be considered with caution. The implied uniformitarianism can be disrupted by random factors like earthquakes or sudden opening of new entrances, or human activities.

The formation of speleothems is influenced by numerous factors, both external and internal to the karst system. Processes linked to local surface

environment, bedrock lithology, soil type, local vegetation and seasonal setting to karst hydrology, morphology and cave climate all affect the way in which environmental signals are transferred from the surface into the speleothem archive (Mattey et al., 2010; Breitenbach et al., 2015). Conducting extended monitoring of selected internal parameters (e.g., air temperature, drip rates) helps to understand the sensitivity that a site has (i.e., the speed and magnitude of its response) to external environmental processes, like rainfall and temperature changes at different timescales. It helps to assess whether and to what extent, environmental parameters are reflected in drip rates and dripwater chemistry. Thus, environmental monitoring in caves guides the interpretation of speleothem proxy records regarding past climate and environmental variability (Spötl et al., 2005; Fairchild, 2007; Riechelmann et al., 2011).

Although numerous monitoring studies have been reported from Central Europe (Dreybrodt & Scholz, 2011; Labuhn et al., 2013; Genty et al., 2014; Baker et al., 2018), some regions remain understudied, such as Normandy, north-western France. Here, we find the cave and quarry system of Caumont that hosts actively growing stalagmites, one of which is currently the subject of detailed investigation. The results of that study will be published in a separate publication. Here, we present the results from an 20-month monitoring campaign and discuss the implications for future speleothem proxy records from Caumont. We analyse how surface precipitation and temperature dynamics are reflected in the dripwater stable isotope composition and evaluate how these key parameters are archived in cave carbonate deposits. We aim to characterize the present condition of the cave and how the internal settings (e.g., temperature, drip rate, etc.) influence the palaeoenvironmental signals recorded in stalagmites.

SITE SETTINGS

Normandy's chalk plateau in northwestern France hosts many caves along the valley of the Seine River, which cuts deep meanders into the chalk (Nehme et al., 2020). Caumont study site (N 49°22'41"; E 0°54'47"; 15 m above sea level) is the largest cave and quarry system in Normandy (Fig. 1A), located on the left bank of the Seine, ca. 25 km southwest of Rouen.

Excavated for building stone since medieval times until the early 20th century, the Caumont cave and quarry system has numerous entrances that are located ca. 200 m from the Seine River (Fronteau et al., 2010). The underground quarry system comprises a ~12 km long network of ~20 m wide and ~50 m long galleries (Fig. 1B) (Ballesteros et al., 2022). The quarry galleries intersect natural cave passages, which total ~4 km (Rodet, 1985). During WWII, the Caumont quarry was occupied in 1943 by German soldiers who began to build an underground factory for liquid oxygen. In 1944, the factory was abandoned leaving behind a ~250 m long bunker made of concrete (Fig. 1B) (Sibout, 2011).

The thickness of the epikarst is ca. 120 m (Ballesteros et al., 2021) and the overlying vegetation is composed of hardwoods and conifers forest (C3 type). For the last two centuries, successive survey maps of the area show no evidence of a drastic change of the vegetation cover over the study area (Geoportail.gouv.fr).

The cave is developed in dedolomitized chalk with flint layers from the upper Cretaceous chalk group, specifically the Soteville Formation (Fm) (Coniacian, 86.3-89.8 Ma) (Fig. 1B). The chalk group is covered by Quaternary deposits, mostly sand and clay residue weathered from the overlying Paleogene sediments. The chalk bedrock presents subhorizontal bedding (Ballesteros et al., 2021) and is moderately deformed, with low amplitude anticlines and local faulting (Nehme et al., 2020). The soils above the cave are predominantly classified as poorly developed rendzina rich in calcium and magnesium. Towards the West, some brown leached soils are found which correspond to slightly clayey aeolian silts (SIGES, 1974). The cave has an underground stream, "Rivière de Robots", which flows in a NE direction and drains into the Seine River (Fig. 1B).

The nearest meteorological station is Rouen-Boos, located 20 km east of Rouen. Normandy's climate is classed as temperate maritime (Cantat, 2004) where the wettest months are October to January and the driest March to July. Monthly rainfall varies from 25 to 163 mm and the average annual air temperature is 11.3°C, with a range from 7°C to 15.5°C (Cantat, 2004). Recent research suggests that the potential evapotranspiration (PET) in the region since 1959 is higher in the spring and summer months (Diomard & Chéron, 2020).

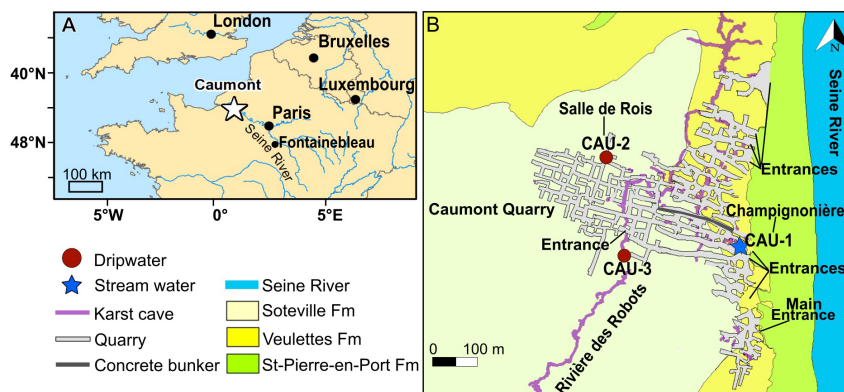


Fig. 1 A) Location of the Caumont cave and quarry system in northwest France relative to the Seine River. B) Geological map of Caumont displaying the quarry, natural cave, and concrete bunker contours, along with the location of the three monitoring stations and the different cave entrances. Map modified from Ballesteros et al. (2020).

METHODS

Collection of regional climatic data

Regional climate data from the nearest meteorological station Rouen-Boos, located ca. 20 km from the cave, is used for direct comparison with monitoring results from the cave. We selected monthly temperature and precipitation (P) for the monitoring period (November 2019 to July 2021). Effective infiltration was obtained by calculating the potential evapotranspiration (PET) with the online Thornthwaite method (<https://ponce.sdsu.edu/onlinethornthwaite.php>; Palmer & Havens, 1958) and then, subtracting precipitation (P) with evapotranspiration (P-PET). Precipitation isotope data were acquired from the nearest available global network of isotopes in precipitation (GNIP) station Fontainebleau, located at ca. 169 km south-east of Caumont, which provides a local meteoric water line (LMWL).

Collection of monitoring data in the cave and quarry system of Caumont

The monitoring campaign was carried out in both underground quarry and cave. Two galleries in the quarry were studied, 'La Champignonière' and 'Salle des Rois', which host the monitoring stations CAU-1 and CAU-2, respectively (Fig. 1B). The third station (CAU-3) is located in the 'Rivière des Robots', a natural karst conduit with variable dimensions along the Robots stream (Ballesteros et al., 2022). The stream opened a passage of ~1.60 m wide, and as it intercepts larger chambers it widens up to ca. 3-5 m. The 'Rivière des Robots' flows in a north-eastern direction towards the Seine River (Fig. 1B).

We collected calcite grown on glass plates and water (drip and stream), and measured cave parameters every 2-4 weeks between November 2019 and July 2021, with a few gaps in 2020, due to Covid lockdown. Both cave air and water temperatures were measured using the same instruments in CAU-1. For sites CAU-2 and CAU-3, Niphargus temperature loggers (Burllet et al., 2015) were left from January 2020 to July 2021 and recorded air temperature every 20 minutes. CO₂ concentrations were measured twice (December 2022 and February 2023) at the three monitoring sites using a handheld Vaisala CARBOCAP with a

GM70 CO₂ probe. Several CO₂ measurements were performed along the Rivière de Robots up to the monitoring site CAU-3.

Stable isotope measurements in water and calcite

During the whole monitoring period, 74 stream and dripwater samples were collected in 50 ml and 12 ml plastic vials respectively. At station CAU-1 only stream water from the Rivière des Robots was collected (Fig. 2A) and at stations CAU-2 and CAU-3 dripwater samples were collected. The water sampling for the three stations was done every 2-4 weeks. At station CAU-2, the dripwater from two drip spots runs through fractures and falls ca. 4.7 m from the ceiling to plastic collection bags with funnels. At station CAU-3, the dripwater was collected directly from stalactites (Fig. 2C) at two adjacent spots. During the monitoring period from November 2019 to July 2021, drip rates (drip counts per minute) were measured during each field visit at station CAU-2.

All water samples were measured at the Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research (AWI), Potsdam (Germany). Of each sample, 12 µl of water were processed using a PICARRO L2130i Isotopic Water Liquid Analyzer. The results are given in δ¹⁸O and δ²H permil (‰) relative to the Vienna Standard Mean Ocean Water (VSMOW) standard (Meyer et al., 2000; Juhls et al., 2020). We collected dripwater samples and modern calcite on glass plates at station CAU-2 (Fig. 2B). The glass plate was exchanged during each field visit (every 2-4 weeks). A glass flask was left for 20 months upside-down 50 cm from the monitored site of CAU-2 and collected in July 2021 for calcite sampling and analysis. The calcite harvested from the glass plates was collected using a cleaned scalpel and analysed for stable oxygen and carbon isotopes. Calcite deposited on the glass flask was sampled with a scalpel from different ~3 x 3 mm areas from side to side, mimicking sampling along a stalagmite growth axis for a HENDY test (Hendy, 1971). The glass flask left on site was cut vertically and scanning electron microscope (SEM) images were taken of the exposed crystal fabrics at the Department of Geography and Environmental Sciences, Northumbria University, Newcastle upon Tyne (England).

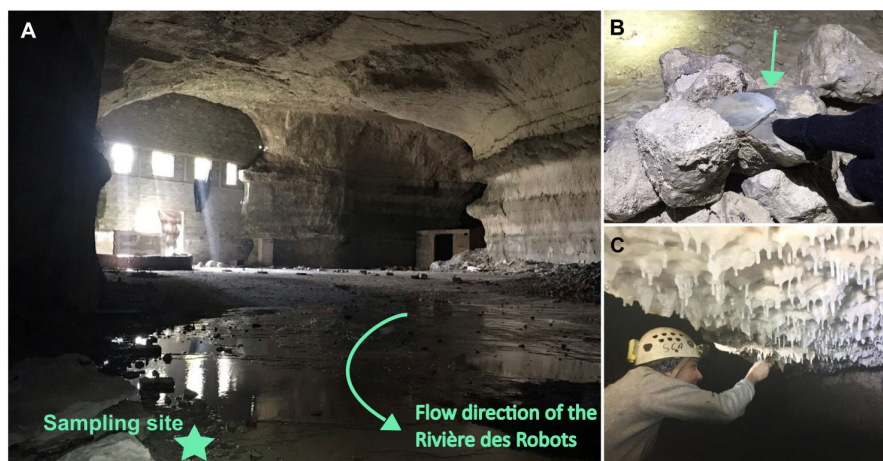


Fig. 2. A) Station CAU-1 in La Champignonière with the stream flowing in NE direction. B) Station CAU-2 in Salle de Rois, showing the glass plate where calcite was harvested approximately bi-weekly through the study. C) Station CAU-3 at the Robots stream, where dripwater was collected directly from soda straws.

Calcite powders were measured for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ using an isotope ratio mass spectrometer (IRMS) at the Department of Geography and Environmental Sciences, Northumbria University. 95–125 μg of calcite powder were weighed using a Mettler Toledo MT5 FACT microbalance and placed into 12 ml borosilicate glass vials. Sample vials were flushed with Helium using a Sercon autosampler. The samples were acid digested at 72°C using orthophosphoric acid (H_3PO_4) and analysed using a Gasbench II coupled with a ConFloIV and a ThermoFisher Scientific Delta V Advantage IRMS following the methodology of Spötl and Vennemann (2004) and Breitenbach and Bernasconi (2011). The raw isotope values were corrected using the in-house carbonate standard, Plessen and the international standards NBS18, NBS19, and IAEA603. In-house standard Pol-2 was measured in each run as control for long-term accuracy. Isotope results are reported in delta notation against the international Vienna Pee Dee Belemnite (VPDB) standard. The long-term external precision of both, $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$, is 0.1‰ (1 standard deviation) or better.

RESULTS

Regional climatic parameters

Over the monitoring period the air temperature recorded in Rouen-Boos meteorological station shows an average of ca. 11°C, with a maximum of 20.4°C in August 2020 and minimum of 3.8°C in January 2021 (Fig. 3).

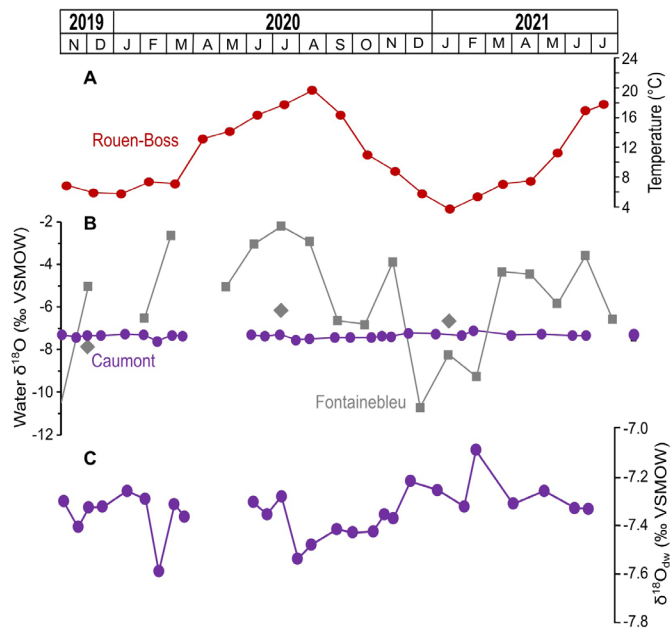


Fig. 3. A) shows mean monthly temperature recorded at the meteorological station of Rouen-Boos. B) shows $\delta^{18}\text{O}_{\text{prec}}$ at the GNIP station of Fontainebleau, with the weighted annual mean of 2019, 2020, and 2021 (diamonds), and $\delta^{18}\text{O}_{\text{dw}}$ values observed at station CAU-2 in Caumont. On the right CAU-2 average, purple circle, which covers the small error of $\pm 0.1\text{‰}$. C) shows the expanded version of $\delta^{18}\text{O}_{\text{dw}}$ record from station CAU-2. CAU-2 $\delta^{18}\text{O}_{\text{dw}}$ data falls between the three values of annual weighted mean of precipitation.

The rainfall distribution between November 2019 and June 2021 reveals a clear seasonal pattern, with overall higher rainfall during the winter seasons and drier conditions during summer. Precipitation

was higher in November and December 2019 and rains persisted until March 2020. The following rainy season was slightly less pronounced at the end of 2020. The driest month in both 2020 and 2021 was April with an average of 0.8 mm and 0.5 mm, respectively. The average $\delta^{18}\text{O}$ of precipitation ($\delta^{18}\text{O}_{\text{prec}}$) observed at Fontainebleau between -10.7 and -2.2‰ (Fig. 3) and the precipitation $\delta^2\text{H}$ over the sample period ranges between -74.5 and -38.7‰ . The lowest $\delta^{18}\text{O}_{\text{prec}}$ values are recorded in November 2019 (-10.5‰) and December 2020 (-10.7‰) and the highest values in March (-2.6‰) and July 2020 (-2.2‰). The $\delta^{18}\text{O}$ of the dripwater ($\delta^{18}\text{O}_{\text{dw}}$) in the Caumont quarry (station CAU-2) is very stable with a mean value of $-7.34 \pm 0.1\text{‰}$ VSMOW, this $\delta^{18}\text{O}_{\text{dw}}$ value is ca 1.6‰ lower compared to the average value in annual precipitation. The effective recharge results show the highest infiltration in November 2019 (143 mm) and December 2020 (130 mm). Negative infiltration was recorded from April until September 2020 plus February and April 2021, with the lowest point being July 2020.

Cave microclimate and monitoring data

Cave air temperatures are $10.4 \pm 0.3^\circ\text{C}$ at station CAU-2 and $11.3 \pm 0.1^\circ\text{C}$ deeper in the cave and quarry system at CAU-3 (Fig. 4). A wider temperature range was observed at site CAU-1 near the entrance to the Champignonière. Here a minimum temperature of 4°C and a maximum of 12.5°C was observed. The stream water temperature at CAU-1 is similar to the air temperature, with a range of 6.5 to 11°C over the monitoring period.

CO_2 concentrations at stations CAU-1 range from 490–560 ppm and at CAU-2 from 480–570 ppm, only slightly higher than atmospheric values. In the Rivière des Robots, CO_2 values were measured along the natural conduit towards the monitoring station CAU-3. The values increased drastically from 510–550 ppm at the entrance of the conduit to 2580–4940 ppm at CAU-3 inside the gallery (Supplementary Table S1).

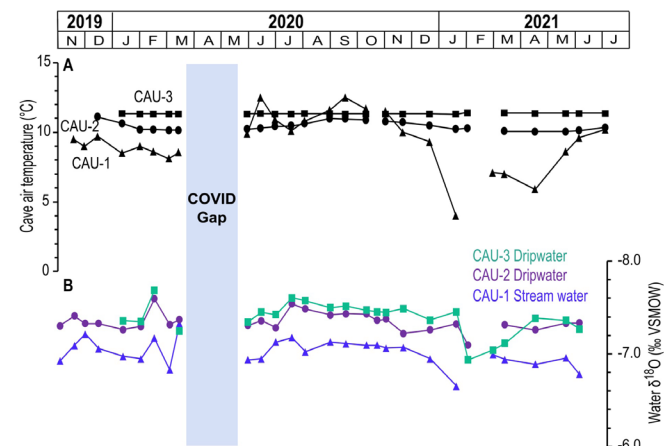


Fig. 4. Comparison of monitored cave air temperature (A) and water $\delta^{18}\text{O}$ (B). At CAU-1, samples were taken from stream water and at sites CAU-2 and CAU-3 from dripwater.

Isotopic composition of stream and dripwater

Twenty-eight stream water (sw) samples collected at CAU-1 yielded $\delta^{18}\text{O}_{\text{sw}}$ values from -7.3 to -6.7‰ . These are slightly higher than the dripwater samples

collected at CAU-2 and CAU-3 (Fig. 5). The dripwaters collected at stations CAU-2 and CAU-3 show a range of -7.7 and -6.9‰ for $\delta^{18}\text{O}_{\text{dw}}$ and -36.2 and -49.6‰ for $\delta^2\text{H}_{\text{dw}}$ (Fig. 5). The results for $\delta^{18}\text{O}_{\text{dw}}$, $\delta^{18}\text{O}_{\text{sw}}$, and $\delta^2\text{H}$ show that all samples fall close to the global meteoric water line (GMWL) and Fontainebleau local meteoric water line (LMWL) (Fig. 5).

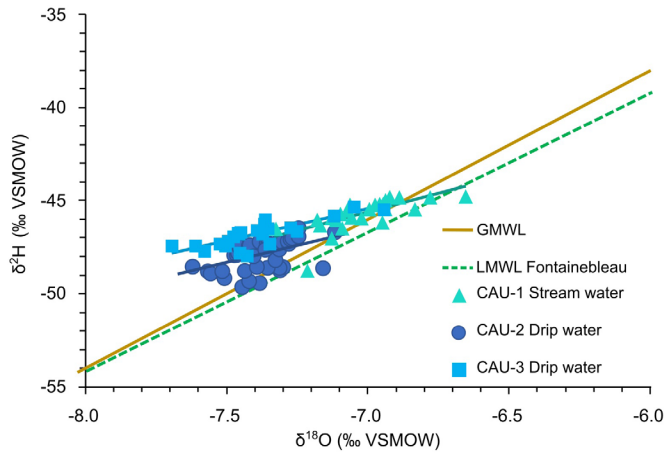


Fig. 5. The $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of the 83 samples collected at stations CAU-1 (triangles), CAU-2 (circles), and CAU-3 (squares) for both stream and dripwater in relation to the GMWL and LMWL at Fontainebleau.

Isotopic composition of modern calcite

The 22 modern calcite samples collected from glass plates placed at station CAU-2 yielded a carbon isotope ($\delta^{13}\text{C}_{\text{calc}}$) signal between -11.1 and -5.7‰ . The lowest $\delta^{13}\text{C}_{\text{calc}}$ value is observed in December 2020 and the highest in September 2020. For the carbonate oxygen isotope ($\delta^{18}\text{O}_{\text{calc}}$) the average value is $-5.2 \pm 0.3\text{‰}$ over the monitoring period (every 2-4 weeks) (Fig. 6). The $\delta^{18}\text{O}_{\text{calc}}$ varies only $\sim 0.6\text{‰}$ from -4.3‰ in October 2020 to -5.6‰ in March 2021 (Supplementary Table S2). Both, $\delta^{13}\text{C}_{\text{calc}}$ and $\delta^{18}\text{O}_{\text{calc}}$ seem to covary, albeit with minimal variance in $\delta^{18}\text{O}_{\text{calc}}$ (Fig. 6C, D). The samples obtained from the glass flask, collected at the end of the monitoring period, also show stable $\delta^{18}\text{O}_{\text{calc}}$ values with a range of -5.04 and -5.5‰ . The $\delta^{13}\text{C}_{\text{calc}}$ values range from -11.3 to -10.9‰ .

DISCUSSION

Cave atmospheric dynamics and internal parameters

Cave air temperature and CO_2 concentrations

Temperature and CO_2 variations in the cave affect calcite precipitation rates and $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ signatures (Deininger et al., 2012; Fohlmeister et al., 2020). Since cave air temperature and CO_2 concentration depend on ventilation dynamics the latter indirectly affects calcite precipitation (Verheyden et al., 2008; Riechelmann et al., 2011). Temperature gradients between surface and cave can enhance or subdue cave ventilation, which in turn can affect CO_2 level and relative humidity in the cave air. Cave ventilation could lower the relative humidity sufficiently to induce evaporation

from the water film on stalagmites and enhance CO_2 degassing from incoming dripwater (Fairchild et al., 2007). Both processes can alter the isotopic signature in speleothem carbonate (Verheyden et al., 2008; Matthey et al., 2010; Fairchild & Baker, 2012). The temperature gradient between cave and surface can vary at diurnal to seasonal timescales (e.g., Breitenbach et al., 2015; Giesche et al., 2023). The ventilation of a cave also depends on the geometry of the cave, the number of entrances, the dimensions of passages, and orientation of entrances towards main wind direction(s) (e.g., Riechelmann et al., 2019). Assessing the temperature variability and amplitude at different sites is essential to characterize the environmental controls on calcite precipitation.

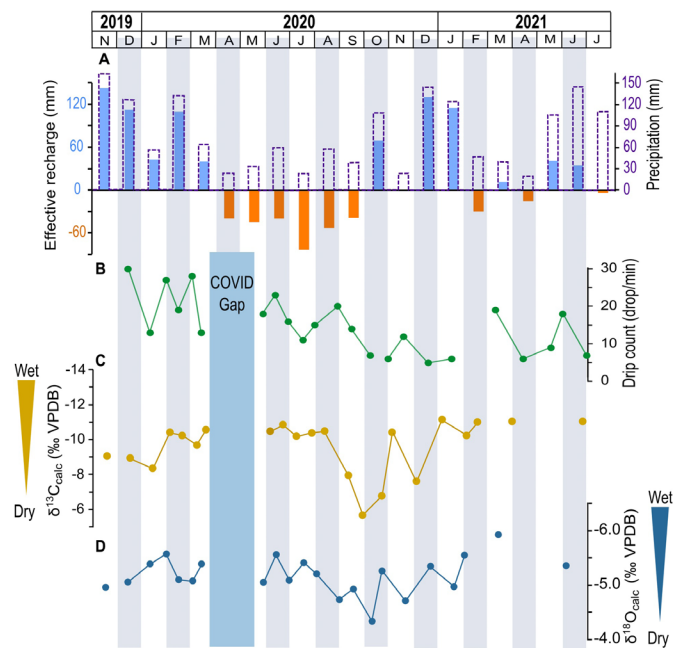


Fig. 6. Monthly precipitation and effective recharge at station Rouen-Boss; Data source: Meteo France (A), compared to CAU-2 drip rate (B), modern carbonate $\delta^{13}\text{C}$ (C), and modern $\delta^{18}\text{O}$ (D). Effective recharge is calculated using the Thornthwaite equation (Palmer & Havens, 1958). The significant increase in $\delta^{13}\text{C}$ values between September and December 2020 might reflect the hydrological (increased PCP) response to the prolonged lack of infiltration over the spring and summer of 2020 (shown in A).

Air temperature measurements at each of the three stations provide information on cave atmosphere and ventilation. Temperature inside the cave and quarry system of Caumont differs between monitoring stations CAU-1, CAU-2, and CAU-3 (Figs. 1B; 3). At station CAU-1 near the Champignonnière entrance, air temperature varies between ca. 4°C in January and 12.5°C in September. Located 50 m from this entrance CAU-1 (Fig. 1B) is more strongly affected by ventilation and direct air exchange with the surface which explains the large air temperature variability (Fig. 4). Temperature recorded at station CAU-2 varies $<1^\circ\text{C}$ between summer and winter seasons over the monitoring period (Fig. 3). At station CAU-3 the temperature range is even more stable, with an amplitude of $<0.1^\circ\text{C}$. The observed reduced temperature range with increase in distance from the entrances reflects the effect of decreasing ventilation at stations CAU-2 and CAU-3 stations.

Our data show that the distal sections, especially station CAU-3, are highly thermally stable and do not respond to surface temperature changes at short timescales (diurnal to seasonal). Such stable conditions make these cave and quarry sections suitable for testing near-equilibrium carbonate deposition.

Drip rate

Drip rate is controlled by surface processes (e.g., precipitation, evaporation, soil moisture capacity) and aquifer characteristics including reservoir capacity and bedrock permeability (Treble et al., 2013; Markowska et al., 2015). The local hydrology feeding the stalagmite determines whether a seasonal signal might be preserved or not. Baldini et al. (2021), divided the bedrock pathways that recharge the drip into diffuse, fracture and conduit flows. Naturally, drip dynamics can also reflect a mix of diffuse and fracture flow. Diffuse flow sites are distinguished by a slow response to precipitation events, while fracture flow sites have a more rapid response. Finally, conduit flow is generally characterized by rather rapid response to individual rainfall events followed by a fast return to baseline flow (Atkinson, 1977; Smart & Friederich, 1986). Very slow drips are also more strongly affected by isotope fractionation, due to prolonged CO₂ degassing and/or evaporation (Mühlinghaus et al., 2007, 2009). Therefore, monitoring drip rates over several seasons helps to characterize the recharge dynamics, lagged response to rainfall, the seasonality signal preserved in the cave hydrology, and isotope fractionation conditions (Atkinson, 1977; Baker & Brunson, 2003; Kaufman et al., 2003; Baker et al., 2019; Baldini et al., 2021).

Seasonal drip rate changes depend on rainfall amount and temporal distribution, evapotranspiration, and recharge dynamics throughout the year. Lower dry season rainfall and high surface temperature result in enhanced impact of evapotranspiration on the epikarst, reducing infiltration and lowering drip rates. In contrast, during winter, evapotranspiration is lower and effective precipitation is enhanced resulting in increased infiltration and higher drip rates (Fig. 6A, B) (Van Rampelbergh et al., 2014).

At station CAU-2 dripwater is available throughout the year. While the station does not reveal clear seasonal cycles, we observed lower drip rates at the end of summer and during the autumn in 2020 (Fig. 6). Decreasing drip rate reflects the drying in the epikarst after lower infiltration summer periods. Both fractures and chalk matrix of the epikarst were less saturated with meteoric water during dry periods, resulting in reduced head pressure and lower drip rates. After a prolonged dry period, like summer 2020, drip rates are likely to respond more slowly to rainfall events due to a partly emptied epikarst reservoir. Following a longer rainfall and refilling of the epikarst, as it occurred during winter season 2020-2021, drip rate increased and might react more directly to rainfall events.

The classification scheme of Baldini et al. (2021) was used with the limited drip rate data obtained at CAU-2. In the scatter plot, our values map above and

parallel to the 1:1 line between monthly maximum vs minimum drip rates. This range has been classified as a “mixture of diffuse and fracture flow” and/or “ideal seasonal flow” by Baldini et al. (2021). Therefore, increasing recharge after December 2020 could be explained by the high level of fractures present in Caumont quarry (Ballesteros et al., 2020; Chevalier, 2022). This is observed especially at station CAU-2, where stalagmite deposition occurs preferentially below fractures (Fig. 7). However, the limited temporal resolution of the drip count at station CAU-2 does not allow us to quantify the lag between rainfall events and drip response, although the available data suggest a response within 4-6 months.



Fig. 7. Stalagmite growth near station CAU-2 aligns with fractures in the gallery's ceiling. This growth pattern strongly suggests that fracture flow is a key characteristic for drips in Caumont Quarry.

Rain and drip water isotopic composition

The degree to which cave dripwater $\delta^{18}\text{O}_{\text{dw}}$ reflects the $\delta^{18}\text{O}$ of meteoric precipitation ($\delta^{18}\text{O}_{\text{prec}}$) is significant for paleoclimate studies and requires detailed monitoring. Numerous publications investigated the relationship between $\delta^{18}\text{O}_{\text{prec}}$ and $\delta^{18}\text{O}_{\text{dw}}$ in order to establish the sensitivity of cave systems to surface hydrology and links between surface environment and speleothem-based $\delta^{18}\text{O}$ time series (Riechelmann et al., 2011; Johnston et al., 2013; Surić et al., 2018; Baker et al., 2019; Nehme et al., 2019; Baldini et al., 2021). $\delta^{18}\text{O}_{\text{dw}}$ values may reflect $\delta^{18}\text{O}_{\text{prec}}$ on timescales ranging from annual (Baker et al., 2019) to individual (intense) recharge events, depending on the site's hydrological characteristics. Other factors such as overburden thickness, lithology, residence time and mixing of the water within the epikarst, soil depth, and aquifer hydraulics can add further complexity.

The oxygen isotopic composition in rainwater is influenced by changes in air temperature, atmospheric circulation (especially dynamics linked to the North Atlantic), precipitation amount and seasonal distribution. The temperature control on $\delta^{18}\text{O}_{\text{prec}}$ at the Orleans-La-Source station in north-western France is ca. +0.23‰/°C. The nearest GNIP station (Fontainebleau), shows cooler $\delta^{18}\text{O}$ values (−4.5‰) during the summer months, and lower values in winter (January-March; −9.6‰) (Fig. 3). The $\delta^{18}\text{O}_{\text{dw}}$

values in Caumont do not show a clear relationship with monthly precipitation, and the very small variation ($\pm 0.4\text{‰}$) results most likely from thorough mixing of waters of different age in the epikarst. When the $\delta^{18}\text{O}_{\text{dw}}$ values are compared to $\delta^{18}\text{O}_{\text{prec}}$ data from Fontainebleau, where a clear seasonal variation is observed around a mean value of $-5.7 \pm 2.7\text{‰}$, the $\delta^{18}\text{O}_{\text{dw}}$ values tend to be more negative with an average of $-7.3 \pm 0.1\text{‰}$ throughout the year (Fig. 3). The observed offset between $\delta^{18}\text{O}_{\text{prec}}$ and $\delta^{18}\text{O}_{\text{dw}}$ implies that dripwater at Caumont is biased towards the high-infiltration season (e.g., winter).

To detect possible secondary evaporation from infiltrating water either in the overburden or in-cave we evaluate the $\delta^{18}\text{O}_{\text{dw}}$ and $\delta^2\text{H}$ values relative to the LMWL (Ayalon et al., 1998; Breitenbach et al., 2015). All dripwater $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values fall close to the GMWL and LMWL (Fig. 5). Samples from stations CAU-1, 2, and 3 cluster around an average value of $-7.3 \pm 0.2\text{‰}$, showing that these waters are well mixed within the epikarst. Furthermore, this indicates that few secondary evaporation occurred and that the dripwater likely retains the (average) precipitation signal (Riechelmann et al., 2011).

The fact that the isotope signatures of all three stations closely overlap at the lower left end of the GMWL suggests that the former reflect wet (winter)

season recharge, rather than annual average infiltration (Genty et al., 2014).

Testing modern carbonate for kinetic isotope fractionation

Information on the importance of kinetic processes is of great relevance for the interpretation of speleothem-based isotope time series in terms of past environmental changes (Mühlinghaus et al., 2009). We therefore tested i) whether $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of samples deposited at the same time vary significantly in response to CO_2 degassing and/or evaporation (effectively a Hendy test on modern carbonate; Hendy, 1971), and ii) whether $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ vary over time in response to changes in temperature and humidity.

A Hendy test on modern carbonate

To test whether kinetic processes affect $\delta^{13}\text{C}$ and/or $\delta^{18}\text{O}$ we collected 13 calcite subsamples from the bottom of a reversed glass flask that was left at station CAU-2 for 20 months (Fig. 8A-D). This test mimics a Hendy test along a speleothem 'growth layer' (here the glass flask) and allows us to estimate the impact of kinetic isotope fractionation at different distances from the central drip spot. SEM microscopy indicates that $\sim 60\text{ }\mu\text{m}$ of blocky calcite was deposited over 20 months (Fig. 8B).

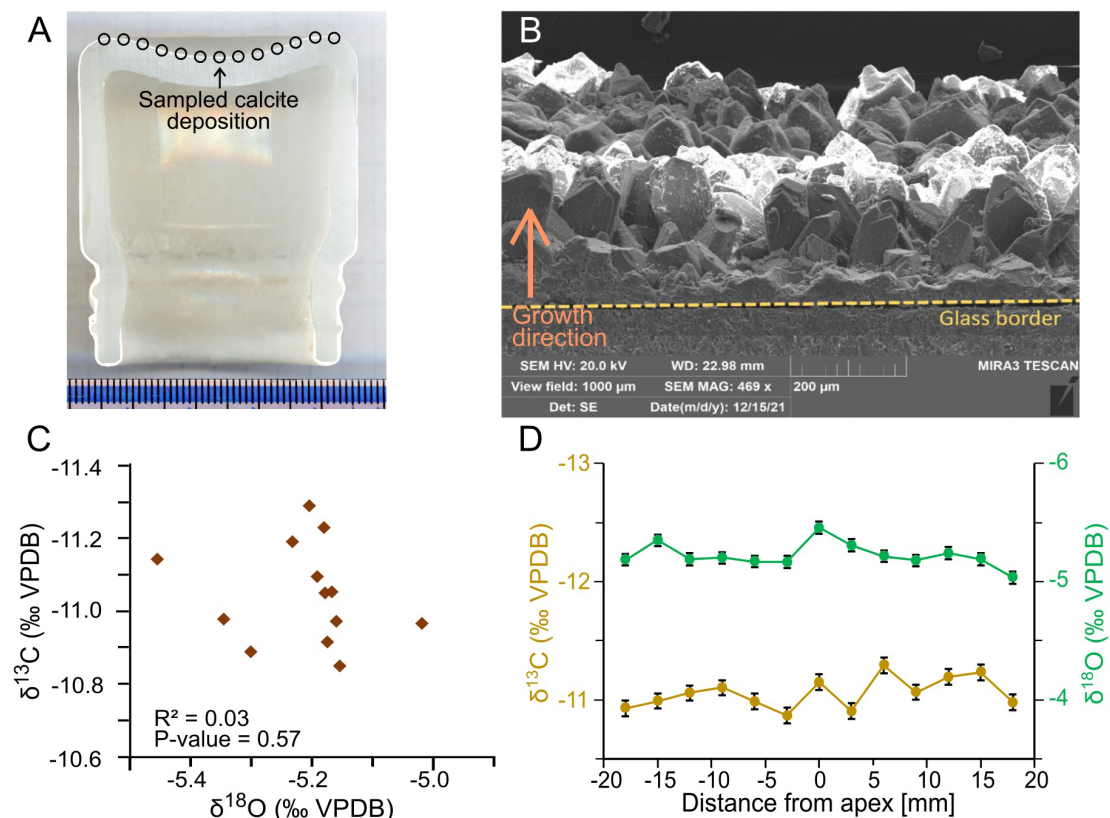


Fig. 8. A) Cross section of the cut glass flask with sample location. B) SEM image of a cut piece from the glass flask. It clearly shows calcite crystals precipitation. C) Cross correlation of $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values. D) Hendy test for the modern calcite vs distance.

The $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values obtained from the 13 subsamples indicate only minimal variability of 0.4‰ , where carbon and oxygen isotope ratios are uncorrelated (R^2 and p -value of 0.03 and 0.57, respectively) (Fig. 8C). Neither isotope ratio shows a trend towards higher values with distance from

the apex (Fig. 8D). These observations indicate that kinetic isotope fractionation through CO_2 degassing or evaporation did not affect the studied isotope system because CO_2 degassing from the water film would lead to increasing $\delta^{13}\text{C}$ values and evaporation would lead to increasing $\delta^{18}\text{O}$ values with distance from the

apex (Hendy, 1971). If both processes were active, the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values should be positively correlated (Hendy, 1971). Our data suggest that the kinetic processes are negligible at this station and that the carbonate contains the original environmental signal of the infiltrating water (i.e., processes in the atmosphere, soil, and epikarst). To evaluate the sensitivity of both isotope systems against environmental processes, we tested their evolution over time with modern carbonate samples, deposited at different times.

Temporal evolution of the isotopic composition of modern calcite

To test the temporal variability of carbon and oxygen isotope ratios we repeated the glass plates calcite growth experiments at station CAU-2 at bi-monthly to monthly intervals (Fig. 6C, D). The $\delta^{18}\text{O}_{\text{calc}}$ of the 22 modern samples ranges from -5.9 to -4.3‰ , with the highest values observed in late summer 2020 (Fig. 6C). When we combine water and carbonate $\delta^{18}\text{O}$ values and cave air temperature to calculate the water-calcite oxygen isotope fractionation factor we find that all samples fall either on or near the regression line proposed by Coplen (2007) and above the one by Tremaine et al. (2011) for (near-)equilibrium conditions (Fig. 9). This finding bolsters our notion that kinetic processes are negligible at station CAU-2 and that $\delta^{18}\text{O}$ could reflect changes in precipitation and humidity above the cave.

The observed $\delta^{18}\text{O}$ changes of ca. -1.6‰ might be related to changes in air temperature, or changes in the dripwater composition (Van Rampelbergh et al., 2014). The minimal temperature variability of $<1^\circ\text{C}$ at station CAU-2 is too small to have a significant influence on the isotopic composition. We conclude that the $\delta^{18}\text{O}_{\text{calc}}$ variation is linked to dripwater changes and by extension to the isotopic composition of the precipitation and (potentially) PCP in the epikarst. Thus, $\delta^{18}\text{O}_{\text{calc}}$ is a sensitive proxy for environmental changes above the Caumont cave/quarry system.

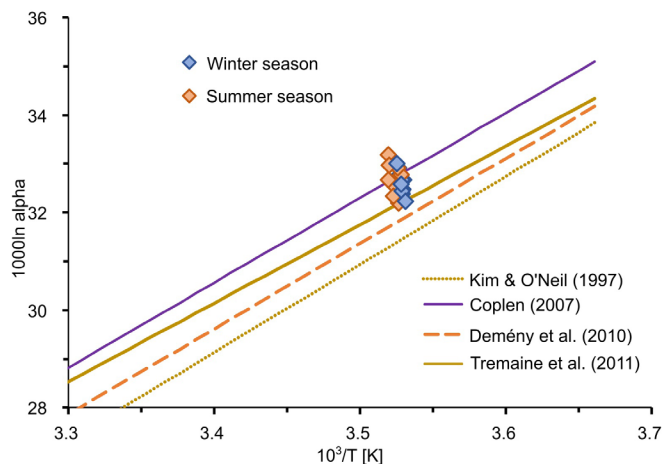


Fig. 9. Plot of calculated water-calcite oxygen isotope fractionation factors, based on the isotope values of modern calcite and water and cave air temperature. All our samples fall either on the regression line of Coplen (2007), or that of Tremaine et al. (2011), strongly suggesting (near-)equilibrium conditions. We show the regressions of Demény et al. (2010) and Kim and O'Neil (1997) for reference. The blue diamonds reflect the samples from winter months and orange from summer months.

The carbon isotopes from the modern samples ($\delta^{13}\text{C}_{\text{calc}}$) vary more significantly around a mean of $-9.7 \pm 1.5\text{‰}$. In late summer and autumn 2002, $\delta^{13}\text{C}_{\text{calc}}$ values increase ca. 2‰ , returning to more negative values around the end of 2020 (Fig. 6). With no noticeable kinetic isotope fractionation is occurring in the cave, this suggests that the isotopic signature of the dissolved inorganic carbon (DIC) of the infiltrating water is affected by processes in the epikarst.

Following Riechelmann et al. (2013), processes that could result in higher $\delta^{13}\text{C}_{\text{calc}}$ values include i) reduced drip rates and longer water residence time on the stalagmite's surface that allow for CO_2 degassing and isotope fractionation, and ii) variations in pCO_2 of the cave atmosphere caused by ventilation changes. As discussed above, the in-cave conditions at station CAU-2 are very stable, rendering either process unlikely candidates for the observed changes. We also do not find a link between drip rate and $\delta^{13}\text{C}$, with a R^2 of <0.02 and p-value of 0.64. (Fig. 6B and C). Therefore, the more plausible explanation for the observed $\delta^{13}\text{C}_{\text{calc}}$ increase is prior carbonate precipitation (PCP) in the epikarst (Fohlmeister et al., 2020). The fact that $\delta^{13}\text{C}$ in the modern carbonate increases during late summer and autumn suggests that higher summertime evapotranspiration above the cave reduced effective infiltration, thereby allowing partial emptying of the epikarst reservoir. Infiltrating water encountering air-filled voids in the epikarst will be subject to PCP, and thus alteration of the $\delta^{13}\text{C}$ signal. This mechanism has also been observed at Han-sur-Lesse Cave in Belgium (Van Rampelbergh et al., 2014) where higher $\delta^{13}\text{C}_{\text{calc}}$ values during the summer months have been associated with intensified PCP.

Thus, the carbon isotope signal in the Caumont cave and quarry is a sensitive proxy for local infiltration changes and speleothems from this cave are likely to record the history of seasonal and longer (multi-annual) droughts in Normandy.

CONCLUSIONS

A monitoring study in the Caumont cave and quarry site (Normandy, NW France) was conducted during the years 2019-2021 to better understand the effects of cave environments on calcite deposition and assess the suitability of the site for paleoclimate study from stalagmites. Cave monitoring was carried out every 2 to 4 weeks in Caumont for 20 months between November 2019 and July 2021. The obtained results display constant internal climatic conditions throughout the year, especially in the natural conduit and deep galleries. The recorded temperature in the CAU-3 (Rivière des Robots) and CAU-2 (Salle des Rois) have a small variability up to 1°C . The $\delta^{18}\text{O}_{\text{dw}}$ values replicate closely the $\delta^{18}\text{O}_{\text{prec}}$ values with most of the samples falling close to GMWL and LMWL, based on Fontainebleau GNIP station data. The $\delta^{18}\text{O}_{\text{dw}}$ signal tends to be more negative compared to Fontainebleau, indicating that it is biased towards the high infiltration season (winter). Furthermore, the low variability in the $\delta^{18}\text{O}_{\text{dw}}$ values throughout 20 months ($\pm 0.1\text{‰}$ for CAU-2 and $\pm 0.2\text{‰}$ for CAU-3), indicate

that the infiltrated waters seem to be well mixed in the epikarst.

A test for kinetic fractionation was done on modern carbonate deposited on a glass flask left at CAU-2 station during the complete monitoring period. The Hendy test performed on the freshly deposited carbonate showed that in the cave no discernible kinetic isotope fractionation is occurring, and hence the $\delta^{18}\text{O}$ signal comprises the original signature of the infiltrating water. The carbonate $\delta^{13}\text{C}$ signal reflects the processes that occurred in the soil and epikarst, which sometimes (whenever effective recharge is negative) includes epikarst PCP but is not affected by in-cave CO_2 degassing.

We tested (near)equilibrium conditions using both a Hendy test on modern carbonate, and the combination of dripwater $\delta^{18}\text{O}$ and modern carbonate $\delta^{18}\text{O}$. The former reveals neither increasing $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ratios with distance from the apex, nor a correlation between both isotope ratios. The calculation of the oxygen isotope fractionation factor based on known cave air temperature, water $\delta^{18}\text{O}$ and carbonate $\delta^{18}\text{O}$ reveals that our modern carbonate maps on or very near the equilibrium regression proposed by Coplen (2007). Together, these tests provide strong evidence for (near)equilibrium isotope fractionation conditions at site CAU-2.

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