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CLIMATIC AND ENVIRONMENTAL VARIATIONS DURING THE LAST INTERGLACIAL RECORDED IN A NORTHERN FRANCE TUF (CAOURS, SOMME BASIN). COMPARISONS WITH REGIONAL RECORDS

Julie DABKOWSKI1,2, Nicole LIMONDIN-LOZOUET2, Julian ANDREWS3, Alina MARCA-BELL3 & Pierre ANTOINE2

ABSTRACT

The geochemical content of calcareous tufa has been recently proven to provide reliable reconstructions of climate variability during Pleistocene interglacials. As one of the best documented, the Last Interglacial (also known as the Eemian in continental area or the MIS 5e in marine records) has been frequently referred to as an analogue for the Holocene. The Caours tufa deposit from northern France provides a long record of the evolution of both climate and environment during the Last Interglacial. Geochemical data in tufa calcite are interpreted to record primarily air temperature (δ18O), biomass productivity (enhanced during wetter – warmer – periods; δ13C) and rainfall intensity (Mg/Ca). The combined data identify a warm and wet climatic optimum followed by oscillations of temperature and humidity conditions. These climatic variations recorded through the Caours profile are strongly consistent with the palaeoenvironmental reconstructions from malacological data. Comparison with those derived from a Grande Pile pollen record suggests that tufa may have precipitated at Caours from ca. 126 to ca. 115 ka (i.e. during the MIS 5e) and that the recorded climatic variations could be related to significant events affecting the Northern Atlantic region. Data comparison with the isotopic record from a speleothem (BDinf) from Bourgeois-Delaunay cave, in south-western France, confirms the relative climatic instability recorded during the Last Interglacial at Caours.

Keywords: calcareous tufa, stable isotopes, Mg/Ca, Last Interglacial, Eemian, palaeoclimate.

1 - INTRODUCTION

Geochemical data from terrestrial carbonates such as lake deposits or speleothems (e.g. Fontes et al., 1996; Xia et al., 1997; Von Grafenstein et al., 1999) is now widely used to reconstruct palaeoclimatic conditions. Tufa deposits are part of this family of continental deposits and result from calcite precipitation at ambient temperature under
open-air conditions in streams, rivers and lake shorelines (Capezzuoli et al., 2014). Their mineralogy (typically >95 % CaCO₃) makes them highly suitable archives of stable isotope (δ¹⁸O and δ¹³C) and trace element (Mg, Ca) information, from which palaeoclimatic conditions can be inferred (Dabkowski, 2014). Recently, scientific interest in Pleistocene tufas as palaeoclimatic archives has increased (Martín-Algarra et al., 2003; Antoine et al., 2006; Limondin-Lozouet et al. 2006; Preece et al., 2007; Ashton et al., 2008; Ortiz et al., 2009; Domínguez-Villar et al., 2011) but to date, most of the geochemical work on tufas has been on recent and Holocene aged material (see review by Andrews & Brasier, 2005; Andrews, 2006; Dabkowski et al., 2015), with just a small number of studies focused on older material (Brasier et al., 2010; Dabkowski et al., 2011, 2012).

This paper presents results from an Eemian tufa from the Somme Basin (Caours, Northern France). The Eemian is known as the interval of climate amelioration during the Last Interglacial (LIG) associated with the spread of temperate mixed forest (Kukla et al., 2002a). In marine sediment cores, the LIG is defined as the Marine Isotopic Stage 5e, the time interval before the Last Glacial period (from 132 to 115 ka) during which temperatures were similar or higher than during the Holocene (Kukla et al., 2002a). In Western Europe, the Eemian interval does not match precisely the MIS 5e and appears diachronous, with a longer duration in Southern Europe, from 126 to 110 ka, than in latitudes above 50°N, where tundra vegetation expanded from 115 ka, while deciduous forest still occupied southern latitudes (Sánchez Goñi et al., 2005).

As one of the best documented Pleistocene interglacial, the LIG has been proposed as an analogue to understand Holocene climate variations (although insolation parameters were slightly different during the LIG; Van Kolfschoten et al., 2003).

Climate stability (or instability) of the Last Interglacial has been widely discussed (Rioual et al., 2001; Kukla et al., 2002a; Shackleton et al., 2002); our detailed study of Caours tufa deposit provide new data to the Eemian palaeoclimatic record in north-western Europe. We here discuss the stable isotope data of the tufa calcite, supported by trace element analyses which can be interpreted in terms of relative changes in palaeotemperature and palaeohumidity. These interpretations are assessed in the context of wider palaeoenvironmental data from mollusc assemblages. We finally attempt to compare our data with those from other regional climatic records of the Last Interglacial, namely Grande Pile and Bourgeois-Delaunay cave.

2 - STUDIED SITE AND MATERIAL

The calcareous tufa sequence at Caours, near Abbeville (Somme Basin, Northern France, fig. 1), was initially mentioned in the 1950s (Breuil, 1952; Patte, 1953; Breuil & Barral, 1955). Early observations concerned a now disused quarry, which, at the time, had yielded Cervid remains associated with “Levalloisian” knapped flints (Agache et al., 1963). Here, F. Bourdier (1969, 1974) swiftly recognised probable evidence of the “Riss-Würm interglacial”. On the basis of these early discoveries, research was resumed in Caours in 2003 by an interdisciplinary team in the context of the SITEP programme (Eclipse II INSU-CNRS). This research resulted in the complete study of the sequence stratigraphy, the confirmation of its chronological attribution to the Last Interglacial, the reconstruction of the palaeoenvironmental dynamics during this period and the discovery of several Palaeolithic levels associated with faunal remains (Antoine et al., 2006). Excavations supervised by Jean-Luc Locht were conducted from 2004 to 2010 as part of a CNRS-INRAP collaboration (Locht et al., 2009).

The Caours tufa formation and the underlying fluvial silts are spread over a surface of several thousand square metres at the confluence of the Scardon and the Drucat stream, with an average thickness of 3 to 4 m. This formation represents an exceptionally well-preserved complex in comparison to the rare Pleistocene tufas known in the Somme valley, which generally only extend over a few square metres (Rousseau et al., 1992; Antoine & Limondin-Lozouet, 2004).

The tufa formation overlies a periglacial alluvial formation corresponding to the penultimate stage of coarse infill of the valley at a relative altitude of +6 m in relation to present valley floor incision. This alluvial formation is attributed to the Étouvie Formation of the Somme Terrace System corresponding broadly to the marine isotopic stage (MIS) 6 (associated to the Saalian glacial; Antoine et al., 2000). Results from the malacological study enabled us to place the transition between the Saalian and the Eemian interglacial within the silt sequence underlying the tufa formation (Limondin-Lozouet in Antoine et al., 2006). This chronostratigraphic attribution is largely confirmed.
by geochronological data (U-series on calcitic stromatolithic tufa, OSL on aeolian or fluvial quartz grains and ESR/U-series on associated herbivorous tooth), which gives an average age for the tufa deposit of 123 ± 3 ka (Antoine et al., 2006; Bahain et al., 2010).

The base of the tufa sequence is separated from the underlying calcareous silts (unit 10, fig. 2) by a thin (ca. 0.5 cm) peat layer (unit 9). The lower part of the tufa sequence shows two thin organic tufa horizons (units 7 and 5; 2 to 10 cm), which contained archaeological remains, alternating with pure calcareous tufa (fig. 2). Units 7 and 5 correspond to drier conditions underlined by a change in the tufa microfacies (« mille feuille » facies, Dabkowski et al., 2010), allowing the formation of incipient “organic soil” horizons associated with bioturbations and a high concentration of rodent remains and forest molluscs (see below). Pure calcareous tufa units 6 and 4 to 1 are granular to sandy, beige to withish or greyish deposits, typical of a perched springline tufa (Pedley, 1990), locally incised by 1 to 2 m large channels. A detailed stratigraphic description was provided in previous publications (Antoine et al., 2006; Dabkowski et al., 2010, 2011; Dabkowski, 2011).

Fig. 2: Summarised geochemical and malacological data regarding Caours stratigraphy (“Caours 2007 - East Profile”, on the left). Boxes labelled with italic numbers show the locations of samples M1 to M55 on the stratigraphic log. Geochemical profiles: Diamonds are stable isotope data (δ¹⁸O is in black on the left and δ¹³C in grey). Black triangles are the Mg/Ca ratio shown with the confidence interval for each sample (grey strips). Additional data from unit 5 (samples M56 to M59 from “Caours 2007 - South Profile”) are shown in the grey square below according to the same scales and keys as data from the main sequence, emphasising the strong δ¹⁸O maximum and δ¹³C minimum. Malacological data (on the right): number of shells (diagram a); distribution of terrestrial molluscs within different ecological groups (diagram b) leading to the identification of several mollusc zones showing the extension of forest habitats (zone 2) followed by a period of more open environments (zones 3 to 5). Upper zones yield faunas of wet soils (zones 3 and 5) but are interrupted by a drier episode (zone 4). Diagram c shows the frequency of aquatic molluscs from which Belgrandia marginata is separated: the occurrence of this gastropod, typical of water table habitats, is a significant indicator of spring activity.

Données malacologiques (à droite) : nombre de coquille (diagramme a) ; répartition des mollusques terrestres au sein des différents groupes écologiques (diagramme b) permettant d’identifier plusieurs zones malacologiques mettant en avant l’expansion des habitats forestiers (zone 2) suivie d’une période où l’environnement est plus fermé (zones 3 à 5). Les malacozones les plus récentes présentent des faunes de sols humides (zones 3 et 5) et sont séparées par un épisode plus sec (zone 4). Le diagramme c présente la distribution des mollusques aquatiques, au sein desquels celle de Belgrandia marginata est mise en exergue : la présence de ce gastéropode typique des eaux froides de sources est un indicateur pertinent de l’activité de ces dernières.
Towards the valley, the calcareous formation is covered by silty colluvial deposits and humic soils following a major phase of erosion truncating the upper part of the tufa sequence. The facies of these soils and their stratigraphic position indicate that they are similar to the Weichselian Early-glacial humic soil complexes of the Somme basin.

3 - OVERVIEW OF THE MALACOLOGICAL RECORD AND OTHER PALAEOENVIRONMENTAL DATA

Earliest fine grained deposits at Caours are composed by non organic calcareous silts (unit 11, underlying unit 10 – not shown) in which mollusc shells are the only bioproxy preserved. Malacological assemblages of this first zone are characterised by low species diversity although abundant snail populations occur. They are composed by taxa of open ground habitats that can withstand humid and cold climatic conditions (Columella columella, Trochulus hispidus, Pupilla muscorum, Oxytoma elegans; Antoine et al., 2006). The best analogue to malacozone 1 at Caours is the mollusc assemblage recovered from the Younger Dryas calcareous silts in the Somme basin, which is typical of humid and cold environmental conditions and include the same marshy taxa, C. columella, today restricted to a Boreal-alpine European range (Limondin-Lozouet & Antoine, 2001; Limondin-Lozouet, 2011). The following mollusc zone 2 of Caours begins in the upper part of the silt accumulation (unit 10; fig. 2). Diversity increases dramatically and mollusc faunas are dominated by thermophilous species indicating the occurrence of forest environments and thus temperate climatic conditions. The abrupt transition between these two mollusc zones suggests a hiatus corresponding to the very early beginning of the interglacial period (Limondin-Lozouet, 2011). The earliest Neanderthal occupation (archaeological level N4; Antoine et al., 2006; Locht et al., 2009) occurs in unit 10. The associated mammal fauna (aurochs, red deer, roe deer and meadow rhinoceros) confirms the full interglacial conditions emphasized by molluscs (Auguste in Antoine et al., 2006). The absence of the fallow deer is a good indication of an open forest. The mollusc zone 2 extends up to within unit 4, covering the first part of the tufa accumulation and reveals the progressive development of the forest with optimal interglacial conditions recorded in unit 5 by both maximal diversity and number of shells of shade-loving mollusc species (fig. 2). It is noteworthy that each organic layer of this succession, corresponding to slow accumulation rate, is marked by a new arrival of forest snail species indicating the extension of shaded biotopes and yields Neanderthal settlements (Antoine et al., 2006; Limondin-Lozouet & Preece, 2014). Associated mammal and micromammal faunas are consistent with the malacological record and typical of a full temperate climate (Antoine et al., 2006). In the large mammal associations from unit 5, contemporaneous with the last Neanderthal occupation at Caours (archaeological level N1; Antoine et al., 2006; Locht et al., 2009), the rhinoceros and the elephant disappear while the fallow deer is predominant. This indicates a more closed forest environment in agreement with the ecological context of the Eemian optimum (Antoine et al., 2006).

The organic tufa of units 5 and 7 are separated by granular tufa levels (units 8 – not preserved here, 6, 4) yielding malacofaunas including high numbers of aquatic and ostracod assemblages dominated by benthic species and with some taxa typical of cold spring waters (Antoine et al., 2006). Those invertebrate faunas reveal episodes of rapid tufa accumulation in the bottom of the valley of both river and spring origin. In the middle part of the tufa (units 3 and 2), both molluscs and ostracods reveal the dominance of aquatic conditions. Belgrandia marginata, a gastropod living in water tables, shows an important peak while ostracod assemblages appear mostly composed by spring taxa (Carbonel in Antoine et al., 2006). More dynamic flows in the valley are emphasized by biological proxies and suggest that tufa accumulated at higher rates in a wetter floodplain. Terrestrial habitats are represented by mesophilous snails of humid environments (mollusc zone 3) or open ground mollusc species more tolerant of drier soils (mollusc zone 4), which inhabit grassland covering tiny islands between small channels created by waters flowing from springs. Some leaf prints of humid biotopes such as incrusted fern (Phyllitis scolopendrium) have also been recovered from these layers (Antoine et al., 2006). Fewer thermophilous gastropods occur discontinuously indicating that forest habitats are still not very far from the river banks. Those witnesses of interglacial conditions persist in the upper part of the tufa (unit 1) although in lesser extent suggesting a decline of temperate conditions. In fact, the mollusc zone 5 appears mostly characterized by the development of wet grassland species; in the same level, B. marginata peaks again and benthic species reappear within ostracod assemblages indicating more open conditions.

4 - METHODS

4.1 - SCREENING FOR DIAGENETIC ALTERATION

Before geochemical analysis the tufa was examined under petrographic microscope as impregnated thin sections searching for evidence of diagenetic alteration. The Caours tufa precipitation is attributable to cyanobacteria and microalgae that can be taxonomically determined from crystal morphology (Freytet, 1997). The morphotaxa Brouinella sp. and Ponsinella sp. are predominant; filaments and fascicules are well preserved allowing identification to the species level (in some cases B. arvenensis and B. variegata; Dabkowski et al. 2010). Tufa precipitation in Caours is generally arranged around fragments of plants (leaves and stems, for which only the shape is preserved) but may also occur around fragments of shells, bones or flint. Quartz grains have also been observed. The detrital component is more abundant in organic tufa units 5 and 7 (up to 15 %, mostly
quartz grains) but aragonite mollusc shells remain scarce (< 2 %) and do not constitute a significant amount of carbonate compared to tufa calcite precipitated in situ.

No significant recrystallisation is observed. There is no evidence of sparry calcite cementation of void spaces except in units 3 to 1, where sparry phreatic cements and crusts of fungal-related needle-fibre calcite (Verrecchia, 2002) are present around the pore edges: these do not constitute a significant mass of calcite (< 3 %). Additionally, all units also contain microbial micritic fabrics, either clotted or around tufa grains but they represent only a small amount of calcite (< 5 %). The observed “diagenetic” fabrics are mostly syndepositional forming under similar environmental and climatic conditions as the tufa precipitates. For these reasons, we believe the Caours tufa geochemical data largely records palaeoenvironmental signals preserved at the time of tufa formation.

4.2 - GEOCHEMISTRY

The Caours sample was collected continuously every 5 cm for malacology (samples M1 to M55; fig. 2) on “Caours 2007 - East Profile”. Four additional samples were collected on another profile, a few metres away, where unit 5, contemporaneous of the Eemian optimum according to malacological data (see above), is better developed (samples M56 to M59; fig. 2).

Geochemical analyses were performed on the same samples, which were sub-sampled (about 5-10 g), crushed when required, and sieved to < 250 μm to obtain a few grams of fine tufa material for geochemical analyses. Stable isotope and trace element analyses were performed according to protocols described below.

4.2.1 - Stable isotopes

Approximately 1 g of each geochemical sample was cleaned of volatile organic matter by low temperature (< 80°C) oxygen plasma etching for 6 h at 300 W forward power in a Bio-Rad PT 7300 plasma barrel etcher. Stable isotope analyses were performed on CO2 derived from 100 ± 10 μg of plasma ashed sample reacted with anhydrous H2PO4 at 90 °C. Isotope ratios were measured on a Europa SIRA isotope ratio mass spectrometer, coupled on-line with an in-house built automatic preparation system, at the Stable Isotope Laboratory of the University of East Anglia (UK). Replicate analyses of the laboratory in house standard (n = 75) gave 2σ precision of 0.09 ‰ for δ18O and 0.3 ‰ for the δ13C. These data are plotted in figure 2.

4.2.2 - Trace elements

About 500 mg of sieved tufa were dissolved in 25 mL of 10 % acetic acid for 1 h. After filtration, the acid was diluted with ultra pure water up to 100 mL. Sr, Mg and Ca concentrations were measured in these solutions on a Fast Sequential Atomic Absorption Spectrometer (AA240FS, Varian), at the Laboratoire de Géographie Physique, CNRS-Meudon (France). Confidence intervals were calculated according to the ISO 11929 standard for each element and then converted (with error propagation) to Mg/Ca and Sr/Ca. Due to large analytical errors in the measurement of Sr concentrations, Sr/Ca variations are lower than its confidence intervals (Dabkowski, 2011), which do not allow us to discuss them. Consequently, only Mg/Ca data are discussed in this paper and shown in figure 2. Some ratios fell outside the confidence limits after error propagation and were rejected, explaining the data gaps in figure 2.

5 - RESULTS AND INTERPRETATION

5.1 - STABLE ISOTOPES

Caours stable isotope data has been previously published and discussed by Dabkowski et al. (2011) but major features are detailed in this paper. The δ18O values range between -5.67 to -4.76 ± 0.09 ‰ (mean value: -5.35 ± 0.09 ‰; n = 59) while δ13C values range between -11.1 to -9.5 ± 0.3 ‰ (mean value: 10.1 ± 0.3 ‰; n = 59; tab. 1). These stable isotope means and ranges are consistent with those from late Quaternary temperate tufas from NW Europe where continentality and aridity/evaporation effects are low (Andrews, 2006).

Regarding to δ13C variations, the Caours tufa sequence is partitioned in two phases: the lowest units show more negative δ13C values (units 7-3 partial mean: -10.3 ± 0.3 ‰; n = 35), then a shift occurs at the boundary between units 3 and 2 toward higher δ13C values (units 3-1 partial mean: -9.9 ± 0.3 ‰; n = 24). However, this change in carbon isotopic values corresponds to a sedimentary hiatus and should not be interpreted as a rapid climatic change. In the so-defined lower part, the uppermost samples in unit 6 and additional samples from unit 5 record the lowest δ13C values (respectively -10.4 and -11.1 ± 0.3 ‰; fig. 2 and tab. 1). This minimum in carbon isotopic data is synchronous with maximal δ18O values (-4.96 in the main profile and -4.76 ± 0.09 ‰ in unit 5 additional samples; fig. 2 and tab. 1). It is followed by significant variations in the oxygen stable isotope values: the δ18O decreases in unit 4, then increases in unit 3, and decreases again in unit 2 (fig. 2).

At decadal-scale sampling resolution, the tufa calcite δ18O has been shown to record the isotopic composition of regional rainfall, which in turn reflects mainly the mean annual air temperature, in modern to Pleistocene tufas (Andrews et al. 1997; Dabkowski et al., 2011). In North-Western Europe, the empirical relationship between δ18O in meteoric water and surface air temperature is known to be +0.58 ‰/°C (Rozanski et al., 1993). The air temperature record in tufa is however “smooth” by the opposing signal of temperature dependence of calcite precipitating in water, which is described by the Craig’s thermodynamic equation, where a 1 °C increase in water temperature results in a -0.24 ‰ change in δ18O of the precipitating calcite (Craig, 1965). Combining
these mechanisms, Andrews (2006) suggests that a 1‰ change in tufa δ¹⁸O actually corresponds to about a 3.5 °C change in mean annual air temperature (i.e. +0.58 ‰/°C - 0.24 ‰/°C). A trend toward higher δ¹⁸O values thus corresponds to increasing temperatures (Andrews, 2006).

Variations in tufa δ¹³C are known to mainly reflect the relative contribution of carbon from sources of dissolved inorganic carbon (DIC) of the depositing waters (Janssen, 2000; Andrews, 2006). The main sources are: (1) DIC derived from soil organic matter with negative δ¹³C values (Cerling et al., 1989; Amundson et al., 1998), (2) dissolution of the aquifer marine carbonate by groundwater (Hudson, 1977), in this case Upper Cretaceous Chalk, with δ¹³C values close to 0 or slightly positive; (3) equilibration of the aquifer and spring water with atmospheric CO₂ (Usdowski et al., 1979). Soil biological activity, associated with vegetation type and abundance, would be mainly promoted by increasing moisture supply under wet environmental conditions, although warmer periods can also favour plant and soil development producing a strong component of isotopically light carbon (source 1 above). Limestone dissolution and water equilibration with atmospheric CO₂ (encouraging “prior calcite precipitation” - PCP, Fairchild et al., 2000) in the aquifer are mainly controlled by recharge characteristics. In-aquifer processes (source 2 and 3 above) would be thus mainly influenced by the rainfall amount: during drier periods, limestone dissolution, equilibration with atmospheric CO₂ and PCP increase and lead to isotopically heavier remaining DIC in the aquifer water which will be transferred to the precipitating tufa (Andrews, 2006). Consequently, lower tufa calcite δ¹³C values would indicate wetter (and warmer) environmental conditions and/or higher rainfall intensity (Garnett et al., 2004; Andrews, 2006; Dabkowski et al., 2011).

On this basis, the Caours sequence shows two distinct climatic phases: in the lower half, wetter conditions (lower δ¹³C) are recorded, then an abrupt transition to relatively drier conditions occurs between unit 3 and 2. However, this transition is related to a hiatus, observed in the stratigraphy, not to an abrupt climate event. Maximal moisture conditions are especially recorded in unit 5 and clearly correlated with the warmest episode (highest δ¹⁸O). According to isotopic data, unit 5 would be thus contemporaneous of the Eemian climatic optimum. After the optimum, oxygen isotopic data show successive trends toward cooler (in unit 4), warmer (in unit 3), then cooler conditions again (in unit 2; fig. 2).

<table>
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<tr>
<th></th>
<th>Main sequence only (n = 55)</th>
<th>Mean sequence with unit 5 additional samples (n = 59)</th>
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<tbody>
<tr>
<td>δ¹⁸O (%) V-PDB</td>
<td>Range -5.67 to -4.96</td>
<td>Range -5.67 to -4.76</td>
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<tr>
<td></td>
<td>Mean -5.38</td>
<td>Mean -5.35</td>
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<td></td>
<td>Confidence interval ± 0.09</td>
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<td></td>
<td></td>
<td>± 0.3</td>
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<td></td>
<td>Estimated maximal warming</td>
<td>+1.5 °C</td>
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<tr>
<td>during the optimum</td>
<td></td>
<td>+2.0 °C</td>
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</tbody>
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Tab. 1: Caours stable isotope data summary.
Significant differences between the main sequence (samples M1-M55) and unit 5 (climatic optimum; samples M56-M59; fig. 2) are in bold. Underlined data are those used to estimate the range between the thermal optimum and the mean recorded temperature (last entry of this Table, see text for detailed).

Tab. 1 : Synthèse des données isotopiques du tuf de Caours. Les différences les plus marquées entre les données de la séquence principale (échantillons M1 à M55) et celles de l’unité 5 (optimum thermique ; échantillons M56 à M58) sont en gras. Les données soulignées sont celles prises en coup dans l’estimation de l’écart de température entre l’optimum thermique et la moyenne des température enregistrée Caours (dernière entrée de ce tableau : voir détails dans le texte).
If a 1 % change in tufa δ¹⁸O corresponds to about a 3.5 °C change in air temperature as suggested by Andrews (2006), the δ¹⁸O maxima would indicate temperatures about 1.5 °C to 2.0 °C warmer during the climatic optimum compared to the average temperature recorded at Caours (for main sequence only and when unit 5 is included, respectively; tab. 1). The following variations, in units 4-1, suggest temperature changes with maximal amplitudes of about 1.5-2 °C. These temperature ranges and their relevance are discussed in section 6.2.

5.2 - TRACE ELEMENTS

Concentration of Mg ranges between 400 and 623 ppm (mean value: 463 ppm, n = 40) and are plotted in stratigraphic order as Mg/Ca ratio on figure 2. Calcium concentrations close to 40 % (tab. 2) confirms that, after dissolution and filtration, the analysed samples were mostly comprised of calcite and validate our laboratory protocol.

Mg/Ca exhibits little variations relative to confidence interval. However, a significant decrease of the ratio occurs at units 6-5 boundary (fig. 2): Mg/Ca is higher that the mean value for the sequence (21.10⁴, tab. 2) in unit 6 then appears generally lower than the mean value from unit 5 to 1. Data from additional unit 5 samples clearly records this transition toward lower Mg/Ca values (fig. 2).

The so-called “trace element” ratios (Mg/Ca and Sr/Ca) have been shown in tufa calcite to mainly depend on rainfall availability through various processes in the aquifer. They are thus valuable complementary data to tufa δ¹³C which is itself controlled by the combined and hard to differentiate parameters including biomass productivity (Garnett et al., 2004; Dabkowski et al., 2012). In-aquifer processes controlling Mg, Sr and Ca concentrations in groundwater are mainly: (1) the higher dissolution rate of calcite compared to dolomite, which allows a possible increase of Mg/Ca once groundwater is saturated with Ca (Chou et al., 1989; Fairchild et al., 2000); (2) prior calcite precipitation (PCP) by degassing in air-pockets leading to Ca concentration decreases relative to Mg and Sr (Fairchild et al., 2000; 2006); and (3) selective leaching of Mg and Sr with respect to Ca even from non-magnesian limestones (Fairchild et al., 1994). These processes are favoured by longer residence time and higher occurrence of air-pockets during low recharge periods (low rainfall), leading to increasing Mg/Ca and Sr/Ca in groundwater (Fairchild et al., 2000; Garnett et al., 2004).

At Caours, a significant increase of rainfall intensity is thus recorded at the units 6-5 boundary (decreasing Mg/Ca) and then rainfall conditions are stable. A closer look at the additional samples from unit 5 shows that the transition from less wet to more intense rainfall conditions is actually contemporaneous with the deposition of this unit (fig. 2).

6 - DISCUSSION

6.1 - COMBINING PALAEOENVIRONMENTAL AND PALAEOCLIMATIC DATA

As geochemical analyses were performed on subsampled malacological samples, direct comparisons between palaeoenvironment and climate dynamics can be achieved with strong confidence at Caours.

In the first part of the tufa accumulation, as mollusc assemblages show the progressive expansion of forest environments (mollusc zone 2; fig. 2), stable isotopes record warm and wet conditions. The two organic tufa layers, units 7 and 5, are both characterised by new arrival of forest snail species and temperature maxima. They were described in thin section as a « mille-feuille » facies characterised by micro-laminations underlined by organic matter and oriented fragments of shells or encrusted vegetation fragments (Dabkowski et al., 2010). Warmer conditions may have led the tufa surface to dry up, at least seasonally, and so favoured the development of microsoils and allowed Neanderthal settlements. In unit 7, mollusc and mammal fauna are characteristic of a forest environment which reaches its maximal development in unit 5, indicating the interglacial optimum: mollusc assemblages are highly diversified and dominated by closed forest species (fig. 2) and the mammal fauna (archaeological level N1) mainly comprise fallow deer remains with no occurrence of the rhinoceros and

<table>
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<th>Mg (ppm)</th>
<th>Ca (%)</th>
<th>Mg/Ca (10⁻⁴ molar)</th>
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<tr>
<td>Range</td>
<td>400-623</td>
<td>33-40</td>
</tr>
<tr>
<td>Mean (n = 40)</td>
<td>463</td>
<td>37</td>
</tr>
<tr>
<td>Mean confidence interval</td>
<td>According to ISO 11929 standard</td>
<td>²σ/₄</td>
</tr>
<tr>
<td>Largest confidence intervals</td>
<td>²σ/₃</td>
<td></td>
</tr>
</tbody>
</table>

Tab. 2: Caours Mg and Ca data summary for the whole sequence (main sequence and additional unit 5 samples, i.e. M1-M59).

Mg/Ca precision intervals were calculated by errors propagation on Mg and Ca measurements; they are shown for each sample on figure 2.
the elephant. In this unit 5, stable isotopes record the warmest and wettest conditions of the sequence (fig. 2). Regarding rainfall, this optimum appears to be a transition interval toward more intense precipitations as shown by the significant shift toward lower Mg/Ca ratios explicitly recorded within unit 5 (fig. 2).

Intense rainfalls persist then, up to the top of the record, and should have fed springwater precipitating tufa as suggested by the high number of aquatic molluscs and ostracods in units 4, 3 and 2 and especially of the spring gastropod Belgrandia marginata (fig. 2).

Following the unit 5 optimum, molluscs of forest environment sharply decrease and mesophilous taxa of humid environments become predominant within terrestrial snails (mollusc zone 3; fig. 2). During this interval, temperatures decrease but humidity conditions remain close to those prevailing during the optimum (no significant δ13C variation; fig. 2). An important shift toward relatively drier conditions occurs at the unit 3-2 transition; the stepped shape of the δ13C signal must reflect a sedimentary hiatus rather than a sudden climatic/environmental variation. In the malacological succession, this transition corresponds to the strong decrease of the spring taxa B. marginata observed at the limit of units 3 and 2. Land snails from open-ground environment and tolerant of drier soils consequently expand in unit 2 (mollusc zone 4; fig. 2). Finally, significant climate variations occurred during unit 1 deposition: a relatively warm and wetter episode is recorded, contemporaneous with the re-expansion of mesophilous mollusc taxa and the development of wet grassland species, as interglacial temperate conditions decline (mollusc zone 5; fig. 2).

6.2 - COMPARISON WITH OTHER REGIONAL CLIMATIC RECORDS

Long-term reconstructions of continental climatic conditions can be achieved mainly on speleothems and long lacustrine sequences. The closest lacustrine sequence that allowed environmental and climatic reconstructions for the last climatic cycle, including the Eemian, is the Grande Pile long palynological record in the Vosges, north-eastern France (Woillard, 1978). Applying the inverse mode to a process-based biosphere model on pollen data from a Grande Pile core, Rousseau et al. (2006) reconstructed the most probable climate at that site from ca. 130 to 100 ka. We attempt here to compare geochemical data from Caours tufa to model derived mean annual temperature and precipitation from Grande Pile (fig. 3). The comparison between these climatic records is strongly supported by that of palaeoenvironmental data from Grande Pile (pollen data: total arboreal pollen - AP - and temperate deciduous arboreal pollen – DAP – percentages are shown in figure 3) and Caours (malacological assemblages; fig. 2). This comparison allows us to position Caours tufa record into the general

Fig. 3: Comparison between geochemical data from Caours (see fig. 2 for details), model derived climatic reconstructions (mean annual temperature and precipitation) from pollen data from Grande Pile XXI core (Rousseau et al., 2006) and the isotopic record from Bourgeois-Delaunay speleothem (BDinf; Couchoud et al., 2009).

On the left, total arboreal pollen (AP, dotted line) and temperate deciduous arboreal pollen (DAP, plain line) percentages from Grande Pile XXI and classic European pollen biostratigraphy (according to Woillard, 1978; Sánchez Goñi et al., 2005), compared to the LIG isotopic stratigraphy (MIS 5) derived from planktonic δ18O records in marine core MD 90-2042 (Shackleton et al., 2002; after Rousseau et al., 2006 modified). Timescales according to authors (see text for details).
Malacological data suggest that tufa started precipitating at Caours shortly after the beginning of the interglacial when reconstructed temperature at Grande Pile increased significantly from ca. 126 ka (fig. 3). The progressive development of the interglacial forest revealed by mollusc zone 2 at Caours can be correlated to the increase of deciduous arboreal pollen (DAP; fig. 3) at Grande Pile up to a maximum corresponding to the *Quercetum-mixtum* warm temperate forest maximum (Woillard, 1978; Rousseau et al., 2006). Within this interval, model derived climate reconstructions suggest that maximal mean annual temperature was reached ca. 124.5 ka at Grande Pile while mean annual precipitations strongly increased. Similar climatic changes are recorded in unit 5 at Caours by the δ¹³C of Mg/Ca signals, respectively (fig. 2 & 3). Additionally, *Quercus* is best represented at this time in the Grande Pile pollen record (Rousseau et al., 2006) which is consistent with the maximal development of thermophilous molluscs in unit 5 (fig. 2). This warm episode is contemporaneous in Caours with the δ¹³C minimum, suggesting strong biomass productivity under wet (and warm environmental conditions). The wettest conditions are similarly recorded in BDinf by a period of more negative δ¹³C around 126.5 ka (fig. 3). However, BDinf δ¹⁸O suggests a decrease of the rainfall amount at this time, after a maximum (low δ¹⁸O values) about 1000 years earlier.

The following maximum in the reconstructed mean annual temperature at Grande Pile (around 123 ka) is questionable and was rejected by the authors as an anomaly with no climatic significance (Rousseau et al., 2006). Otherwise, they described a cooling starting just after the interglacial optimum and up to 122-121 ka

(fig. 3), also indicated by the decrease of DAP. However, warm temperate forest persists during this interval (*Corylus* forest; Woillard, 1978; Rousseau et al., 2006), which is consistent with the predominance of mesophilous mollusc taxa of humid environments in mollusc zone 3, following the optimum at Caours (fig. 2). The persistence of warm forest environments also results in the relatively low δ¹³C values recorded at Caours during this interval, although they increase after the optimum (fig. 2). A similar tendency to higher δ¹³C values is observed in BDinf with millennial-scale variations that might be contemporaneous with those observed at Caours: lower δ¹³C values are recorded around 124 ka and the highest around 125 and 122 ka at Bourgeois-Delaunay (fig. 3). In the meantime, BDinf δ¹⁸O variations suggest the succession of periods of reduced and increased rainfall amount (Couchoud et al., 2009) with no equivalent recorded in Caours Mg/Ca ratio variations, while reconstructions of mean annual precipitation from Grande Pile show no significant variation (fig. 3). Due to error propagation, the Mg/Ca record for this period at Caours is sparse (fig. 2), which does not allow us to discuss further the variability of rainfall intensity after the optimum.

Reconstructions from Grande Pile report a cold interval between 123 and 121 ka, marked by low percentages of DAP, during the *Taxus* peak (fig. 3). It was already observed in temperature reconstructions derived from pollen and beetles assemblages from another Grande Pile core (Guiot et al., 1993). Rousseau et al. (2006) suggest that this event could be contemporaneous with increasing IRD deposition recorded in the Greenland Sea at ca. 122 ka indicating some freshwater input into the North Atlantic (Fronval et al., 1998) and impacting climate and vegetation in NW Europe (Fronval & Jansen, 1997). As tufa deposition is very sensitive to climatic parameters, such a significant cold event is likely to result in the reduction or even the stop of tufa accumulation. We thus assume that the hiatus observed at Caours between units 3 and 2 is contemporaneous of the coolest conditions reconstructed from the Grande Pile record (fig. 3). The BDinf speleothem also stopped growing at this time: the most recent radiometric age for this stalagmite is 121.1 ± 1.4 ka but a 2-5 cm growth segment is missing suggesting that BDinf may have stopped growing between 121 and 120 ka (Couchoud et al., 2009; fig. 3).

At Caours, the hiatus is marked by an important shift toward higher δ¹³C values that mainly reflects less developed soil and vegetation (lower productivity), as rainfall intensity shows no significant variation (no significant Mg/Ca variation). In Grande Pile pollen record, the *Corylus* warm temperate forest declines resulting in a DAP about 10 % lower than during the optimum (Rousseau et al., 2006 and fig. 3), which is consistent with the more open, locally drier, environment reflected by the mollusc assemblage from Caours zone 4 (fig. 2). This period is marked by a warm interval at Grand Pile, from ca. 120 to ca. 118 ka, while temperature similarly increased at Caours after the main hiatus (increasing δ¹⁸O; fig. 3).
The last Eemian warm interval at Grande Pile occurs between ca. 118 ka and ca. 116 ka, while reconstructed mean annual precipitation slightly increased. The warm event recorded in the last part of Caours record (higher δ¹⁸O) is likely to be contemporaneous with this interval. At this time indeed, the Mg/Ca record (rainfall intensity) is scarce but the δ¹³C signal shows a significant increase suggesting wetter conditions, related to the development of wet grassland as suggested by molluscs from the last zone, at the top of the sequence (fig. 2 & 3). In the meantime, molluscs indicate the decline of temperate climatic conditions in the upper part of Caours deposit (Unit 1, mollusc zone 5; fig. 2). At Grande Pile, a dramatic cooling is recorded at about 115 ka, as the percentage of deciduous trees clearly decreases, marking the end of the interglacial sensu stricto. Regarding the sensitivity of tufa deposits to climatic and environmental variations, the decline of interglacial conditions is likely to have affected tufa deposition at Caours and led to its end (fig. 4).

At Grande Pile, Rousseau et al. (2006) suggest that the main changes in the reconstructed climatic parameters would be controlled by major events recorded in the North Atlantic cores. According to our correlations with Grande Pile, temperature and humidity variations recorded in the geochemical content of Caours tufa would not be simply locally relevant but similarly related to regional significant climate events within the MIS 5e.

At Grande Pile, the warmest episode could correspond to the migration of the polar front toward the north in the Greenland Sea at 126-125 ka, as described by Fronval & Jansen (1996, 1997; Rousseau et al., 2006). Maximal δ¹⁸O values in unit 5 at Caours suggest that the climatic optimum was about 1.5-2.0 °C warmer than the average temperature recorded in the whole sequence (tab. 1). This is similar to the range observed at Grande Pile between maximal temperature at ca. 124.5 ka and those recorded in average during the MIS 5e (if the coldest temperature, during the cold event around 122 ka, probably not recorded at Caours are excluded; see fig. 3). Other mean annual temperature changes at Grande Pile during the MIS 5e also seem of similar amplitude as those observed at Caours (ca. 1.5-2 °C). For the same time interval, temperature variations of similar amplitude (as MTCO and/or MTWA) are generally reconstructed from European continental records and off the Iberian margin with a much marked cool and dry event after the optimum at some sites (e.g. Guio et al., 1993; Zagwijn, 1996; Cheddadi et al., 1998; Sánchez Goñi et al., 2012). A dry (and cool) spell is also recorded in BDinf speleothems after a significantly wetter episode at 127.9-126.3 ka (Couchoud et al., 2009; fig. 3). Similar variations are observed in the δ¹³C record at Caours. On the other hand, the scarcity of our Mg/Ca data during this period does not allow discussion of the significant variations of rainfall intensity recorded in BDinf δ¹⁸O.

The occurrence of a cold event at ca. 122-121 ka has been documented in several marine cores, where it is marked by an IRD peak, minimal SST and low benthic δ¹⁸O (McManus et al., 1994; Fronval & Jansen, 1997; Fronval et al., 1998; Shackleton et al., 2002). This event seems to result in a slightly cooler spell in Europe, that was not strong enough to induce a complete, abrupt, change in the environment as at the MIS 6/5 or MIS 5e/5d transitions (Cheddadi et al., 1998; Sánchez Goñi et al., 2005), but would have stopped the growth of BDinf speleothem and induced a sedimentary hiatus at Caours. This may reflect significantly cooler or drier conditions, while malacological data from Caours suggest the progressive development then, retreat of the interglacial forest, with no evidence of a dramatic return to cold environment conditions.

Finally, the decline of interglacial conditions, marked by a strong temperature decrease around 115 ka at Grande Pile, may be related to the C26 cold event in marine cores, linked to an important sea surface cooling in the North Atlantic (Chapman & Shackleton, 1999; Rousseau et al., 2006), which would be related to the stop of tufa development at Caours.

6.3 - TUFAM ACCUMULATION RATE

As no age model based on radiometric dating can be achieved so far for Pleistocene tufa deposits, the period of tufa accumulation at Caours can only be assumed though comparison with other records. To evaluate the inferred chronology, calculation and discussion of the resulting accumulation rate may be valuable.

According to the above comparison with the Grande Pile sequence, we suggest that the tufa deposit developed at Caours from ca. 126 to ca. 115 ka, with a gap contemporaneous with a 1-2 ka long cold event around 122 ka. The average tufa accumulation rate at Caours is thus of roughly 2-4 mm/year. Additionally, this rate appears faster during the first half of Caours deposit, around the climatic optimum, than during the latter part of the Interglacial, as illustrated by the adaptation of height scale according to time length in figure 3. At Bourgeois-Delaunay, the authors assume that BDinf speleothem was inactive during the preceding Termination II and U-series dating show it started growing around 128 ka (Couchoud et al., 2009), which may be simultaneous with the onset of tufa accumulation at Caours. The lower half of Caours deposit would have thus developed from ca. 128 to ca. 121 ka, suggesting an average accumulation rate of 2 mm/year.

Caours tufa deposition is mainly related to the activity of cyanobacteria (Dałkowski et al., 2010), so it is expected to have a slow accumulation rate: rates of 1-3 mm/year are the most commonly observed on modern tufa of this kind in Western Europe. However, higher rates (5.5 mm/year on average) are commonly reported for long Quaternary tufa sequences presenting a succession of various tufa facies as Caours (Pentecost, 2005). Tufa deposits are highly sensitive to climatic parameters (Pentecost & Viles, 1994). They are also better developed when forest environments spread, mainly because they prevent soil erosion and turbidity in rivers and streams,
which in turn enhance aquatic photosynthesis and reduce the abrasion of nucleation sites at the tufa surface. Under forest environments, CO₂-enriched soils leading to CO₂-enriched groundwaters infiltrating through them, also lead to higher dissolution of the aquifer limestone and consequently, to increasing tufa deposition (Goudie et al., 1993; Pentecost, 2005). Indicative accumulation rates derived from the correlation of our data with those from Grande Pile and Bourgeois-Delaunay are thus consistent with rates observed in other modern and Quaternary tufa deposits, faster accumulation being expected during the first part of the Caours record as it is contemporaneous with both the climatic optimum and the maximal forest development. Additionally, these correlations assign Caours tufa deposit to the interval of maximal formations of speleothems and tufas observed worldwide between 125 and 115 ka and correlated with the MIS 5e (Hennig et al., 1983).

7 - CONCLUSION

Combined stable isotopes (δ¹⁸O and δ¹³C) and trace elements (as Mg/Ca ratio) from Caours tufa show a clear record of climate variability during the Last Interglacial. Calcite δ¹⁸O mainly depends on air temperature and δ¹³C is controlled by biomass productivity (enhanced by wetter – and warmer – environmental conditions). Both temperature and humidity reach a clear maximum in unit 5. An alternation of warmer and cooler periods follows this climatic optimum while a significant shift toward relatively drier conditions is observed at mid-sequence, between units 3 and 2, contemporaneous with a sedimentary hiatus. Mg/Ca is interpreted to be mainly controlled by rainfall intensity and during the climatic optimum, in unit 5, it records a significant increase of rainfall intensity, which then apparently remains stable.

Stable isotope and Mg/Ca data from Caours tufa are strongly concordant with palaeoenvironmental reconstructions. Mollusc assemblages from the same profile show the progressive development of the temperate forest (mollusc zone 2), with an unambiguous optimum in unit 5, which is consistent with environmental conditions suggested by mammal faunas from archaeological levels. The following mollusc zones (3-5) indicate a more open environment with the alternation of wet and drier episodes.

Comparison of temperature and rainfall intensity variations recorded at Caours with model derived climatic reconstructions from Grande Pile pollen data are strengthened by the coherence of palaeoenvironmental data from both sites (mollusc and pollen, respectively), and the realistic timing and rates thus inferred for Caours tufa accumulation. The Caours climatic record is likely to cover the MIS 5e (from ca. 126 to ca. 115 ka) and to show responses to regionally significant climatic events recorded at Grande Pile, Bourgeois-Delaunay (BDinf speleothem) and in North Atlantic cores. Variations in the δ¹⁸O and δ¹³C at Caours, together with the successive stops in tufa accumulation (at ca. 122 and ca. 115 ka), suggest a relative climatic instability during the Last Interglacial even if does not lead to dramatic changes in the environment (as shown by malacological data).

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