



HAL
open science

Reappraisal of the chronology of Orgnac 3 Lower-to-Middle Paleolithic site (Ardèche, France), a regional key sequence for the Middle Pleistocene of southern France

Jean-Jacques Bahain, Norbert Mercier, Hélène Valladas, Christophe Falguères, Hassan Masaoudi, Jean-Louis Joron, Laurence Froget, Anne-Marie Moigne, Jean Combier, Moncel Marie-Hélène

► To cite this version:

Jean-Jacques Bahain, Norbert Mercier, Hélène Valladas, Christophe Falguères, Hassan Masaoudi, et al.. Reappraisal of the chronology of Orgnac 3 Lower-to-Middle Paleolithic site (Ardèche, France), a regional key sequence for the Middle Pleistocene of southern France. *Journal of Human Evolution*, 2022, 162, pp.103092. 10.1016/j.jhevol.2021.103092 . hal-03471882

HAL Id: hal-03471882

<https://hal.science/hal-03471882v1>

Submitted on 19 Oct 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Reappraisal of the chronology of Orgnac 3 Lower-Middle Palaeolithic site (Ardèche, France), a regional key sequence for the Middle Pleistocene of southern France

Jean-Jacques Bahain^{a,*}, Norbert Mercier^b, Hélène Valladas^c, Christophe Falguères^a, Hassan Masaoudi^a, Jean-Louis Joron^d, Laurence Froget^c, Anne-Marie Moigne^a, Jean Combier^e, Marie-Hélène Moncel^a

^a *UMR 7194 HNHP MNHN-CNRS-UPVD, Département Homme et Environnement du Muséum national d'Histoire Naturelle, Institut de Paléontologie Humaine, 1 rue René Panhard, 75013 Paris, France*

^b *UMR 5060 IRAMAT CNRS-Université de Bordeaux, Centre de Recherche en Physique Appliquée à l'Archéologie, Maison de l'archéologie, 33607 Pessac cedex, France*

^c *UMR 8212 LSCE/IPSL CEA-CNRS-UVSQ, Bât. 12, Avenue de la Terrasse, 91198 Gif-sur-Yvette Cedex, France*

^d *Groupe des Sciences de la Terre, Laboratoire Pierre Süe, CEN, Saclay, 91191 Gif-sur-Yvette, France, France*

^e *5, rue Saint-Jean, 71000 Mâcon, France*

***Corresponding author.**

Email address: bahain@mnhn.fr (J.-J. Bahain)

Reappraisal of the chronology of Orgnac 3 Lower-Middle Paleolithic site (Ardèche, France), a regional key sequence for the Middle Pleistocene of southern France

Abstract

Previous studies have suggested that the Lower-to-Middle Paleolithic transition was associated with the earliest Neanderthals, but recent research has established that the oldest Neanderthal fossils and the first signs of their technologies and behavior appear from MIS 11 or possibly earlier. To understand these changes, re-evaluation of the evidence is necessary to determine if this transition corresponds to a progressive evolution rather than abrupt change. Orgnac 3 is a key and appropriate site to study this research context. Located in southern France, it yields a long stratigraphic sequence testifying the evolution of technical and subsistence behaviors of pre-Neanderthal human groups during a Middle Pleistocene interglacial-glacial cycle. In this paper, a new chronological framework is provided for the sequence based on results of dating methods applied to various types of geological materials. Speleothems and volcanic minerals, dated in previous studies by U-series and $^{40}\text{Ar}/^{39}\text{Ar}$ respectively, show periods of calcitic crystallization and regional volcanic activity. Other materials, such as heated flints and herbivore teeth, are directly related to evidence of anthropogenic activities and are analyzed in the present work by trapped-charge dating methods such as thermoluminescence and electron spin resonance combined with uranium series (ESR/U-series). The new thermoluminescence and ESR/U-series dates confirm the attribution of the Orgnac 3 stratigraphic sequence to the MIS 10–MIS 8 period and are discussed in relation to paleoenvironmental data derived from bioarchaeological studies. The paleoanthropological levels, including the emergence of Levallois technology, are dated to ca. 275 ka (early MIS 8), and appear coeval to a wet and

temperate period recorded locally, the Amargiers interstadial, defined in the regional palynological records. The implications of this reassessed chronology for the archaeological assemblages are discussed in the wider context of behavioral innovations from MIS 11 onwards and their establishment in subsequent periods.

Keywords: Thermoluminescence dating; ESR/U-series dating; Heated flints; Teeth; Multi-disciplinary geochronological approach

1. Introduction

The transition from the Lower to the Middle Paleolithic is rarely observed in a continuous stratigraphic sequence and is therefore difficult to date accurately. Our knowledge of this transition from a chronological point of view often reflects data obtained from sites scattered over a relatively large area and varied geological contexts. The dating of these Middle Pleistocene archaeological sites is often complicated by the lack of materials suitable for geochronological analyses. If radioisotopic methods based on radioactive decay of elements such as ^{238}U or ^{40}K can provide precise and accurate dates, their use is limited to specific materials, such as speleothems for U-series analyses and volcanic minerals for $^{40}\text{Ar}/^{39}\text{Ar}$ dating. When such radioisotopic methods can be applied to Middle Pleistocene archaeological or paleontological sites, they provide a chronological framework of reference, in which evidence of human activities can be easily contextualized (e.g., Jaubert et al., 2016 or Pereira et al., 2018). Unfortunately, these materials are not always available in the stratigraphic sequences of archaeological sites and the obtained chronology often reflects geological events, such as karstic calcite crystallization or volcanic events, rather than human activities.

Other dating techniques, less precise than the aforementioned radioisotopic methods, can also be directly applied to archaeological remains, thus providing a direct chronology of the human activities at the studied sites. This is the case of trapped-charge methods, such as luminescence or electron spin resonance, which consider the analyzed samples as dosimeters that have recorded the total radiation dose received since the geological or archaeological events being dated. These methods are currently used to clarify the chronology of Middle Pleistocene archaeological sites as they allow the dating of materials that bear witness to past human activities. Thermoluminescence can hence be applied to estimate the date of the last heat affecting mineral pieces, such as flint or quartz artifacts, often recovered from Paleolithic sites in association with hearths or other anthropic structures (Aitken, 1985; Valladas, 1992; Mercier et al., 1995a, Richter et al., 2017). Teeth, another material frequently recovered from archaeological levels, can be dated using coupled electron spin resonance and U-series (ESR/U-series) method directly linking age to hominin subsistence activities (Grün et al., 1988; Falguères et al., 1999; Bahain et al., 2015; Shao et al., 2015). These paleodosimetric methods have consequently been widely used to date Paleolithic archaeological sites since the 1980s (see for example Aitken, 1985; Grün and Stringer, 1991; Roberts et al., 2015; Falguères, 2020). Unfortunately, the established chronologies are often constructed with a single method or the results are contradictory, leading to debates and discussions illustrated for example by the chronology of the Tabun site, Israel (Grün et al. 1991; Mercier et al., 1995b; Grün and Stringer, 2000; Mercier and Valladas, 2004).

Although such comparison and combination of archaeological data with geochronological (radioisotopic and trapped-charge) results are crucial for discussing the chronology of human evolution, these two different dating approaches are rarely used

because it is quite unusual to find speleothems, volcanic minerals, heated flints, and paleontological teeth at the same site. When applied, this multi-methods approach greatly improves our knowledge of the chronology of Paleolithic sites and more generally of human evolution (e.g., Sutkina et al., 2016; Dirks et al., 2017; Richter et al., 2017; Ingicco et al., 2018). Unfortunately sites offering such possibilities are few in number. Orgnac 3, in southeastern France, in addition to being an important archaeolocal site, is a good locality to conduct such a multi-disciplinary dating approach. Here we present results obtained by thermoluminescence (TL) and coupled ESR/U-series dating methods on burnt flints and teeth respectively, and compare these results with data previously obtained by U-series and $^{40}\text{Ar}/^{39}\text{Ar}$ dating on speleothems and volcanic minerals, respectively. In association with available paleoclimatic data, these results permit a reappraisal of the geochronological framework for the site and allow a discussion of the timing and context of the transition between the Lower Paleolithic and Middle Paleolithic in Western Europe.

1.1. Orgnac 3 site and the Lower-Middle Paleolithic transition in southeastern France

Orgnac 3, located near the Rhone and Ardèche valleys (Fig. 1), is a key Middle Pleistocene archaeological site that records the transition between the Lower and Middle Paleolithic through a relatively thick and apparently quickly deposited karstic sequence. Discovered by local searchers in 1956, it was mainly studied by Jean Combier who dug several pits between 1959 and 1962, then directed annual excavations between 1964 and 1972 (Combier, 2005). At present, the site appears like a doline within the Cretaceous limestone constituting the local bedrock of the Bourg-Saint-Andéol plateau. However, during part of the Middle Pleistocene human occupation it was initially structured as a cave, as evidenced by the presence of speleothems crystallized in situ into the deposits. Then, due

to the progressive collapse of the limestone ceiling, it evolved successively into a sinkhole, then into a rock shelter and finally into an open air site (Combier, 1967).

The site today presents a 7-meter thick stratigraphic sequence studied in detail by Debard (1988) and Khatib (1989, 1994), who subdivided it into four main geological complexes allowing the development of the cave to be reconstructed (Fig. 2). Eight archaeological levels were also identified in the sequence (archaeological levels AL 1–8), some of them (AL 5 and AL 4) being subdivided into sub-levels (Combier, 1967; Debard, 1988), which, through lithic and faunal studies, made it possible to show the parallel development of geological change with environmental and technical evolution in terms of lithic technology, subsistence behavior and land-use patterns (Figs. 2 and 3; Gauthier, 1992; Aouraghe, 1999; Moigne and Moncel, 2005; Moncel et al., 2005, 2012, 2020; Valensi et al., 2005).

Complex I, which was not observed during the present study, consists of alternating sands and coarse rounded limestone clasts. It is characterized by a fauna mainly composed of carnivores associated with some herbivores. The presence in the faunal record of reindeer and collared lemming remains indicates a cold and dry period coeval to a glacial stage. The top of this complex provided the first traces of human occupation of the deposit (level AL 8), without any evidence of use of Levallois method, the small quantity of material indicating occasional human occupation of the cave.

Complex II contains a greater proportion of silt and is marked by the presence of several stalagmitic formations, having grown in situ, and of numerous limestone blocks, indicating the progressive collapse of the cave vault. The faunal spectrum of the archaeological levels of this complex (AL 7 and lower part of AL 6) is dominated by deer and horse, and includes forest species such as fallow deer and roe deer. The palynological data,

derived from the analysis of speleothems, also suggest a forest environment, initially closed and then increasingly open, with a humid temperate climate (Gauthier, 1992). These levels do not contain any evidence of use of Levallois debitage.

Complex III, consisting mainly of limestone clasts and blocks embedded in a clayey matrix, bears witness to the opening of the cavity, which was gradually transformed into a rock shelter. The faunal record of the corresponding archaeological levels is dominated first by cervids (upper part of AL 6, AL 5b) and then by bovids (*Bos* and *Bison*) in association with equids and cervids (AL 5a, AL 4b, AL 4a and AL 3). Both geological data and biomarkers indicate a gradual cooling of the climate, which nevertheless remains humid. Seven human teeth attributed to *Homo heidelbergensis* (de Lumley, 1981) were recovered in levels AL 6, AL 5b and AL 5a. Levallois technology appears fleetingly in AL 5b and 5a, becomes a significant part of the debitage in AL 4b and 4a (40% of the cores) and becomes prevalent in AL 3 (90% of the cores).

Finally, Complex IV is characterized by fine sediments (silt then clay) with scattered gravels. It has yielded the most recent traces of human occupation of the site (AL 2 and AL 1). The available paleoenvironmental data with the dominance of equids and the decrease of cervids, coinciding with the replacement of the forest by steppe, indicate a cold and dry climate and an open landscape. Levallois core technology is dominant (90% of the cores), associated with a mosaic of changes in land-use patterns and subsistence behaviors appearing in comparison to the underlying levels.

1.2. *Former geochronological studies at Orgnac 3*

Different dating methods have been applied in the past to Orgnac 3 samples: U-series on speleothems (Shen, 1985; Falguères et al., 1988; Michel et al., 2011, 2013; Ayliffe,

unpublished data), fission tracks and $^{40}\text{Ar}/^{39}\text{Ar}$ on volcanic minerals (Khatib, 1994; Michel et al., 2011, 2013) and ESR/U-series on teeth and bones (Masaoudi, 1995). The first results published in the 1980–1990s seemed to indicate a rapid deposition of the stratigraphic sequence and placed the onset of the Levallois technique at Orgnac 3 as progressing by phases during the MIS 9–MIS 8 interglacial/glacial cycle, among the earliest evidence known in Europe (Moncel et al., 2012, 2020). Indeed, speleothems and volcanic minerals highlight a relatively short depositional history mainly during MIS 9 and early MIS 8 (Shen, 1985; Falguères et al., 1988; Khatib, 1994; Michel et al., 2011, 2013). By contrast, a recent chronological synthesis (Michel et al., 2013) noted that the U-series ages, which range between 320 and 260 ka for AL 7 to AL 5, were in disagreement with the $^{40}\text{Ar}/^{39}\text{Ar}$ ages of around 300 ka obtained on volcanic minerals extracted from AL 2, and argue for a longer depositional history (data summarized in Fig. 2).

ESR/U-series was also attempted at Orgnac 3 by Masaoudi (1995). He analyzed eight teeth from AL 7 to AL 1, revealing that the ‘uranium-series (US) model’ (Grün et al., 1988), classically used for ESR/U-series age calculation, cannot be used for some teeth in relation to high $^{230}\text{Th}/^{234}\text{U}$ ratios indicating probable U-leaching (Supplementary Online Material [SOM] Table S1). The chronology obtained for the other teeth yielded ages ranging from 132 ± 19 ka to 294 ± 44 ka, without regard for the stratigraphic order (SOM Table S2). Although this chronology was considered by Combier (2005) to be in general agreement with the other available geochronological data, this series of dates was considered by Michel et al. (2013) as too scattered to be useful from a geochronological point of view.

1.3. *Current study*

In the present study, new analyses were conducted using TL on heated flint pieces from AL 6 and AL 5 selected from the collection. The ESR/U-series age estimates were also recalculated using in situ dosimetry data (AL 6, AL 5 and AL 4) and paleodosimetric environment reconstruction (AL 7, AL 3, AL 2 and AL 1). We compare these results to the former geochronological dates and available paleoenvironmental data mainly derived from bioarchaeological studies. We then discuss the implications of this reassessed chronology for the archaeological assemblages in the wider context of behavioral innovations from MIS 11 onwards and their establishment in subsequent periods.

2. Material and methods

2.1. Sampling and associated fieldwork

The location of the samples analyzed in the present study by TL and ESR/U-series dating methods is shown in Figure 1.

Eleven burnt flints were selected for TL analyses from the collection of materials recovered during the various archaeological excavations, more specifically from two archaeological levels (AL 6 and AL 5) located in the middle part of the stratigraphic sequence. The study included a dosimetric survey of the site through 11 CaSO₄:Dy dosimeters left for several months (from 365 to 1057 days depending on the levels) at different locations in these two archaeological levels as well as in the upper level AL 4 (SOM Table S3). The in situ dosimetric data also enabled a recalculation of the ESR/U-series ages of the teeth analyzed by Masaoudi (1995; one tooth per archaeological level from AL 7 to AL 1; see Fig. 2). A reconstruction of the paleodosimetric environment of the teeth carried out from the other levels (AL 1, AL 2, AL 3 and AL 7) was also attempted (Fig. 4) and the corresponding ESR/U-series ages recalculated.

2.2. *Thermoluminescence*

Following Valladas (1992), the outer part of the selected flint pieces irradiated by the alpha and beta particles originating from the sediments was mechanically removed with a diamond saw. The remaining core was then crushed, sieved and the 100–160 μm grain-size part was extracted and washed in dilute HCl to eliminate eventual carbonates. Once the samples were prepared, all the analyses aimed at determining the TL ages followed the analytical protocol given in Mercier et al. (1995a). The analysis of the luminescence signals produced by the grains during heating in the TL reader indicated that each flint had been heated in the past to a sufficiently high temperature to ensure that its geological TL was fully reset before it was abandoned. For this purpose, the grain powder (100–160 μm) was dispersed on stainless steel cups and aliquots received increasing artificial doses. The measurement of the TL signals was detected with a UV/blue filter and identified at around 380°C (for a heating rate of 5°/second) allowing verification (the ‘plateau test’; Aitken, 1985). Equivalent doses were then determined by applying an additive dose technique: for each sample, three fractions of the natural (not artificially irradiated) powder (60 mg each) were irradiated with a calibrated gamma source (^{137}Cs source delivering 1.33 Gy/minute) and their signals, as well as those of the natural fraction, were analyzed with a TL reader optimized for the analysis of flint samples (Valladas et al., 1994). The internal dose-rates (alpha and beta) induced by the radioisotopes (U, Th, K) present in the flints were computed from their contents (Table 1) as determined by Neutron Activation Analysis (Joron, 1974). The external dose-rates were computed from 11 dosimeters inserted in the site for measuring the present gamma and cosmic dose-rates in the vicinity of the flint findspots: 1 in AL 4 (square E17), 6 in AL 5 (F12, 13 and B13, 15, 16) and 3 in AL 6 (B12, 15 and D17; SOM Table S3).

2.3. ESR/U-series

The ESR and U-series data used for the ESR/U-series age calculation in the present study were acquired by Masaoudi (1995), following the analytical protocol then classically used to provide ESR/U-series dates using the ESR-US model from Grün et al. (1988; see details of the protocol in Bahain et al., 1992 and Falguères et al., 1997). Nine aliquots of enamel powder (100–200 μ m) from each sample were irradiated using a ^{60}Co source at doses ranging from ca. 20 to ca. 2100 Gy (radiation dose steps: 19, 36, 76, 140, 239, 323, 690, 1397 and 2102 Gy) and one tenth was kept as a natural reference. ESR spectra of the 10 aliquots were measured at room temperature using a E-109 X-band Varian spectrometer with the following conditions: 10 mT scan range, 0.1 mT modulation amplitude, 10 mW microwave power, 0.25 seconds time constant, 4 mn scan time, and 100 KHz modulation frequency. Each ESR measurement was repeated twice for every dose on different days. Equivalent doses were determined from the growth of the $g = 2.0018$ ESR signal of tooth enamel using single exponential fitting using 'ESRSAT', a custom program written by Yuji Yokoyama and used in the Muséum national d'histoire naturelle (MNHN) until the 2000s.

U-series analyses were performed on each dental tissue by α -rays spectrometry according to the standard methods described by Bischoff et al. (1988; SOM Table S1). Briefly, the samples were completely dissolved in a boiling mixture of HNO_3 and hydrogen peroxide. FeCl_2 carrier and $^{232}\text{U}/^{228}\text{Th}$ spike were added to equilibrate for 12 hours. The samples passed through an anion exchange resin equilibrated with 7N HNO_3 to eliminate all the impurities and Fe carrier. U and Th isotopes were separated by successive passages of the obtained solution into anion exchange columns under specific acid conditions (Th is eluted from the resin with 8N HCl while U, which is strongly absorbed by the resin, is eluted

with 0.1N HCl). After drying in separate teflon beakers, U and Th were taken in a small volume of 0.1N HNO₃ and pH was adjusted to 1.0. Th and U were extracted into thenoyltrifluoroacetone in benzene and deposited in separate stainless steel planchets and placed into alpha spectrometers for counting.

The main difference between Masaoudi's and the present work concerns the determination of the annual dose. The gamma contributions of the dose rate were determined by Masaoudi (1995) exclusively from the radioelement contents of the sediments associated with the teeth (SOM Table S4) whereas in the present work doses measured in situ with dosimeters were used for levels AL 4, AL 5 and AL 6 (SOM Table S3). These measurements indeed revealed that the γ dose contributions estimated by Masaoudi (1995) from the sediments associated with the teeth were not fully representative of the doses to which the samples were actually exposed in the corresponding levels. Due to this observation, the ESR/U-series ages were recalculated taking into account the gamma doses measured in situ when available (AL 4, AL 5 and AL 6), or when not available by using dosimetric reconstruction considering overlying and underlying levels (AL 1, AL 2, AL 3 and AL 7; Fig. 4).

This age estimate was obtained using the US model for the main part of the analyzed teeth. In addition, the 'accelerating uptake (AU) model' (Shao et al., 2012) was used to estimate the age of the teeth for which the U-series isotopic ratios in some dental tissues indicate that uranium loss may have occurred after the initial U-uptake. The use of this model can then produce an age estimate when the US model is not usable

ESR/U-series ages were recalculated by either the US or AU models using the following updated parameters, impacting the final estimates: radioelement contents-to-doses conversion factors from Guérin et al. (2011), rather than Yokoyama et al. (1982);

cosmic dose rate estimated from the Prescott and Hutton's equations (1994) and not from Yokoyama et al. (1982); α efficiency (k-value) of 0.13 ± 0.02 (Grün and Katzenberger-Appel, 1994), rather than the k-value of 0.15 used by Masaoudi, in accordance with de Canniere et al. (1986); β -dose rate calculated taking into account the initial and removed enamel thicknesses during sample preparation and the associated β -rays attenuations in the enamel by Monte Carlo approach (Brennan et al., 1997), rather than using Grün's equations (1986); and a zero water content ratio for enamel was used compared to 3 ± 3 wt% used by Masaoudi (1995). Finally, the ESR/U-series ages were calculated using the 'ESR US' and 'ESRAU' custom software programs written by Qingfeng Shao (Shao, 2011), and not with the 'ESREMAIL' custom program written by Yuyi Yokoyama and historically used in MNHN. Note, however, that in both studies some identical parameters were used: potential radon losses were determined for each tissue by combining α -rays and γ -rays spectrometry data (Bahain et al., 1992) and the water content was estimated to be 7 ± 5 wt% in the dentine and 15 ± 5 wt% in the sediment.

3. Results

3.1. Thermoluminescence

Eleven burnt flints, collected in archaeological levels AL 6 and AL 5, were dated using TL. Equivalent doses were then computed following Mercier et al. (1992) by comparing the TL growth curves of the natural and additive dose signals with those induced only by artificial irradiations, after the original TL signal was reset in the laboratory. The equivalent dose values range from 189 ± 34 to 318 ± 20 Gy (Table 1). The external dose rates recorded by the dosimeters were found to vary both between levels (vertically) but also horizontally due to the heterogeneity of the sediments (irregular presence of limestone fragments): the

values range from 526 to 850 $\mu\text{Gy/a}$ for AL 5 and from 316 to 610 $\mu\text{Gy/a}$ for AL 6. For each flint, the external dose rate was then calculated by averaging the doses recorded by the closest dosimeters. However, because of the observed spatial variations, we cannot rule out the possibility that such a mean value does not represent the dose actually received by the sample during burial. To avoid this, the dose would have needed to be measured at the exact location of the sample before the excavation took place, which was not possible. Depending on the radioelement contents of the samples and considering the estimated external dose rates, the internal dose rate represents between 26% and 43% of the total dose rate.

These ages range from 210 ± 20 ka to 332 ± 35 ka for AL 5, and from 244 ± 21 ka to 337 ± 29 ka for AL 6, but these two time intervals overlap. Despite some scatter of the individual age results, potentially due to gamma dose rate heterogeneities and the inadequacy of the external dose rate, the average age values (262 ± 36 ka for AL 5 and 286 ± 30 ka for AL 6), consistent with the stratigraphy, are coeval to the glacial marine isotopic stage (MIS) 8 (considering the curves of Lisiecki and Raymo, 2005).

3.2. *ESR/U-series age recalculation*

The new ESR/U-series ages obtained on the eight mammalian teeth from Orgnac 3 (Table 2) using the TL and reconstructed dosimetric data (SOM Table S3; Fig. 4) range from 311 ± 30 ka to 236 ± 20 ka and are in better agreement with the stratigraphic succession than the age estimates previously calculated by Masaoudi (1995; SOM Table S2). The external dose is the main contribution to the dose rate (48% for the Orgnac 4a tooth to 70% for Orgnac 2 and Orgnac 7 teeth) and explains these differences. For the main part of the analyzed teeth, the US model, describing post-burial U-uptake in the dental tissues, can be

used, and indicates an incorporation first into the dentine and then into the enamel. This process is commonly observed in ESR/U-series dating. For two teeth from AL 4a and AL 5, the US model cannot be applied and the U-uptakes and age estimates were calculated using the AU model. U-leaching was then modeled in both dentine and enamel, but the obtained results are in good agreement with the ESR/US ages of the other levels.

4. Discussion

4.1. Reappraisal of the Orgnac 3 chronostratigraphical framework

The entire set of geochronological data now available for the Orgnac 3 site is compiled in Figure 5, presented alongside the global paleoclimatic curve (Lisiecki and Raymo, 2005) and the regional paleoenvironmental curve (Reille et al., 1998; Roger et al., 1999). Supported by paleoenvironmental data derived from both local paleontological (Moigne et al., 2002; Moncel and Moigne, 2005; Moncel et al., 2012) and regional palynological (Reille et al., 1998; Roger et al., 1999) studies, we propose the following chronostratigraphical framework.

These new results confirm the attribution of the deposition of the Orgnac 3 stratigraphic sequence to MIS 10–8, between 350 and 240 ka. Although some of our dating results could correspond chronologically to MIS 7 (e.g. the ages obtained by TL for AL 5 samples ORG13, ORG19 and ORG40), consideration of the complete dataset, including the paleoenvironmental record, yields no indication of any return to interglacial conditions in the upper part of the stratigraphic sequence (Complexes III and IV). The scattered TL results obtained for AL 5 samples could be related to a poor dosimetric reconstruction for the corresponding samples—although we have no definitive arguments to support such interpretation—but the overlap of both MIS 9 and MIS 7 interglacial records in the AL 7–AL

6–AL 5 succession seems very unlikely, as does a failure to record a complete glacial cycle of about 100 ka in a stratigraphic sequence that appears to be fairly continuous overall. At the same time, the U-series data indicate that the calcitic crystallization of the speleothems occurred for Complex II of the sequence during MIS 9 and the onset of MIS 8, and for AL 5 during the first part of MIS 8. In addition, the ESR/U-series dates obtained for the teeth recovered from these upper levels (Complexes III and IV) are also mainly coeval to MIS 8, and we suggest as a first approximation that the following interglacial stage (MIS 7) is not recorded stratigraphically at Orgnac 3. This interpretation allows the stratigraphic complexes of Orgnac 3 to be assigned to the MIS record as follows.

Stratigraphic Complex I = MIS 10 (ca. 375–340 ka) Although this complex, including AL 8, has not been directly dated by the present study, both geological and paleontological data indicate that its deposition was coeval with a glacial stage. The ESR/U-series age of 311 ± 30 ka obtained on a tooth from overlying AL 7, as well as the first period of stalagmitic flow crystallization (PL 2), place AL 7 within MIS 9, and thus suggest as a first approximation the attribution of Complex I to MIS 10. No evidence for the use of the Levallois method was found in this complex, which corresponds to a phase during which the site was only occasionally occupied by humans.

Complex II = MIS 9 and MIS 9–MIS 8 transition (up to Amargiers interstadial; ca. 340–270 ka)

As summarized by Michel et al (2013), the U-series dates obtained on the speleothems of this complex indicate crystallization corresponding to MIS 9 and early MIS 8 interstadials. The TL and ESR/U-series ages obtained on AL 7 and AL 6 confirm this attribution and place the deposition of these levels contemporary with MIS 9 (AL 7) and the transition between MIS 9 and MIS 8 (AL 6). The paleoenvironmental reconstructions indicate a forested

environment and a temperate humid climate for this complex in which the use of Levallois debitage is still not observed.

Complexes III and IV = MIS 8 (post Amargiers interstadial; ca. 270–240 ka) These complexes record a progressive climatic degradation, with first a decrease in temperature (Complex III) followed by a decrease in precipitation (Complex IV), probably during the same glacial isotopic stage. The ESR/U-series chronology of the upper complexes (from AL 5 to AL 1) allows the corresponding sedimentation to be assigned to the second part of MIS 8, between 270 and 230 ka. It agrees with the U-series age of the calcitic crystallization phase recorded in AL 5 but does not agree with the $^{40}\text{Ar}/^{39}\text{Ar}$ age (303 ± 6 ka; Michel et al., 2013) obtained on the volcanic minerals extracted from the AL 2 sediments, confirming that this $^{40}\text{Ar}/^{39}\text{Ar}$ age estimate does not date the archaeological level but rather, dates an older volcanic event, as previously proposed by Michel et al. (2013). Levallois debitage, rare in AL 5, represents a significant part of the lithic production in AL 4 and becomes quite exclusive in AL 3 and archaeological levels of Complex IV (AL 2 and AL 1). The proposed chronological framework makes it possible to place the deposition of AL 6 and AL 5 as contemporary with the Amargiers interstadial highlighted in the Velay palynological sequences (Reille et al., 1998). The coincidence of the regional climatic optimum of this interstadial phase with a volcanic event allowed it to be dated precisely to 275 ± 5 ka (Roger et al., 1999), i.e. during the first part of MIS 8 according to the global isotopic curve of Lisiecki and Raymo (2005). This is in agreement with the paleoenvironmental reconstruction of Roger et al. (1999) indicating mainly a forest environment during this interstadial. This interpretation dates the appearance of Levallois technology at Orgnac 3 to around 275 ka.

4.2. *Archaeological implications of the new Orgnac 3 chronological framework*

This study demonstrates that the chronology established from geological materials is very useful for site history reconstruction, but can be disconnected from the archaeological history. At Orgnac 3, the speleothems dated by U-series were mainly formed during MIS 9 and early MIS 8, between 320 and 280 ka (AL 7 and AL 6) and between 280 and 260 ka (AL 6 and AL 5), respectively (Michel et al., 2011, 2013). However, some of the dated samples seem to be out of this time range and could correspond to reworked materials (e.g., the PL1–6 sample of Michel et al., 2013). On the other hand, while the $^{40}\text{Ar}/^{39}\text{Ar}$ age of the volcanic minerals from layer AL 2 (Michel et al., 2013) is analytically accurate, it likely corresponds to reworked minerals in the sediments associated with an older volcanic event disconnected from the archaeological record, and it should be considered with caution as the dated minerals are probably reworked into the Orgnac 3 sequence.

Therefore, this reassessment of the Orgnac 3 chronology makes it possible to place the appearance of Levallois debitage at this site to around 280–260 ka, during early MIS 8. This new lithic technology is associated with significant changes in subsistence behaviors and land-use patterns that did not occur at the same pace through the sequence, as observed in other parts of Western and Central Europe. These gradual changes indicate that the transition from the Lower to the Middle Paleolithic is not a threshold, but results from a long evolutionary process that is rooted in the past since at least MIS 11. Orgnac 3 is unique in the sense that through the successive phases of occupation, we can observe the processes and the rhythm of the onset of a new core technology associated with other behavioral changes.

Most of the time, the record of Levallois core technology only allows for the dating of its use and diffusion, not the way in which it was developed, such as in Western Europe as early as MIS 12 and 11 (Cagny-la-Garenne in France or Guado San Nicola in Italy; Moncel

et al. 2020). In Central Europe, this technology has not been found before MIS 8, suggesting an abrupt introduction by dispersal of new populations (Wiśniewski, 2014). In Moncel et al. (2020), the review of European collections and new sites indicated that this technology probably did not come from the Levant, contra Adler et al. (2014), where there is no evidence of Levallois before MIS 9. Only the recent discoveries of the site of Juljulia (Israel) deliver some proto-Levallois cores dated between 500 and 300 ka (Zupancich et al., 2021), possibly pushing back the age of the earliest evidence of Levallois in the Levant penecontemporaneous with the earliest European sites with this technology.

Recent discoveries of an early Levallois technology associated with *H. sapiens* remains at the site of Misliya Cave in Israel, and dated between 240 and 150 ka (Zaidner and Weinstein-Evron, 2020), indicate different trajectories depending on the region. Compared to the earlier Acheuleo-Yabrudian, the Levallois discoveries at Misliya Cave suggest an abrupt introduction of this technology (production of triangular products) at the site by new populations of *Homo* sp., arriving in the Levant later than expected. Early (i.e., MIS 9–8) Middle Stone Age (MSA) sites known in East Africa could be a source of this technology for the Levantine area, (i.e., Tyron and Faith, 2013; Douze and Delagnes, 2016). In Europe, despite the revision of the Apidima hominin skull in Greece (Harvati et al., 2019) suggesting possible early incursions of *H. sapiens* into Eastern Europe as early as MIS 7, current data indicate for Europe at least a plausible relationship between pre-Neanderthals and European hominins with the appearance and spread of Levallois (Moncel et al., 2020, 2021). A gradual evolution from the Early Stone Age to Early MSA is observed in East Africa, as in Western Europe from the Lower to the Middle Paleolithic, suggesting two parallel and similar histories towards more complex behaviors and regional diversity, albeit in different ways and by different hominins.

At Orgnac 3, we observe three time-spaced phases with gradual transition of behavioral changes in lithic technology and subsistence strategies, each feature following its own rhythm (Moncel et al., 2012). The first phase concerns the lower part of the archaeological record with no (AL 7–6) or little evidence (AL 5b–5a) of Levallois debitage. The tool kit was diverse, with various flake-tools including scrapers, denticulates, notches, bifaces (on slabs, pebbles and large flakes), and pebble tools. Hominin occupation was spread over several seasons at the site (Moncel et al., 2012). We also see a decrease in the carnivore occupation of the site. Hominin occupation was more persistent with the opening of the Aven d’Orgnac (aven), collecting raw materials from both the local territory and from nearby rivers (the Ardèche and Céze rivers a few kilometers away and the Rhône more than 15 kilometers distant) in the form of slabs, pebbles and nodules. Six bone retouchers are present (Moigne et al., 2016). According to the proposed chronology, it was coeval with the MIS 9 interglacial and the beginning of the following MIS 8 glacial period, corresponding to a gradual shift from a closed forest environment to a more open landscape under an increasingly cold but relatively humid climate.

The second phase records a mixture of old and new behaviors (AL 4b–4a) during the same early glacial stage, associated with an open environment during transitional climate. Hominins came to the site on a regular basis for repeated short-term occupations devoted to hunting of several species of large herbivores, mainly during a given time of the year, while the aspect of the site changed from a cave to an open air site. Butchery procedures seem to indicate changes in social management and cooperation patterns (with a decrease in the number of cutmarks on the faunal remains). Bifaces are less numerous, perhaps in relation to shorter occupations or greater use of flakes and flake-tools for butchery processes.

The third phase, under colder and drier climatic conditions (AL 3–1), was marked by the predominance of Levallois core technology with a succession of methods on the same core (from mainly centripetal to uni-bipolar and/or preferential removal). There were also changes in the function of the site with specialized horse hunting during the autumn and by more localized land use, with raw material acquisition directly from the southern Tertiary flint formations at the site (Moncel et al., 2012, 2020). It was no longer occupied throughout the year and, after the roof collapse, hominins settled in an open air site protected by the remains of the aven walls. Shorter occupations and the onset of Levallois technology predate the opening of the cave roof and changes in environmental conditions. Phases of the butchery processes became more standardized, with fewer cut marks on bones compared to the lower occupations, indicating a more careful treatment of the carcasses. Cores on flakes became numerous, indicating the recycling of large flakes for knapping and segmentation of the debitage process typical of Neanderthal technology. Cores on flakes produced a large quantity of very small flakes left unretouched. The tool kit became more standardized as well, with the predominance of scrapers with fine retouch and fewer other categories of flake-tools. Most of the Levallois products were unretouched, some perhaps being transported outside suggesting a new form of tool-kit management (Moncel et al., 2020). Bifaces in AL 1 are rare and look more like partial bifacial tools, while the bifaces of AL 3, despite the prevalence of Levallois core technology, are more numerous and varied including some pieces with a general volume management.

Although the earliest Levallois core technology is sporadically recorded earlier than at Orgnac 3, at other Western European sites from MIS 12 onwards, for example at Cagny-la-Garenne (beginning of MIS 12, Northern France), Guado san Nicola (MIS 11/10, Italy) or Frossinone-Ceprano Basin (MIS 11/10, Italy; Peretto et al., 2016; Moncel et al., 2020),

Orgnac 3, at the transition of an interglacial/glacial period (MIS 9/8), marks the first persistent and prevalent use of Levallois technology (90% of the cores for AL 3 to AL 1). This site constitutes an early phase for the general diffusion of new strategies that are typical of the Middle Paleolithic and that subsequently characterize Neanderthal occupations during MIS 5–3.

5. Conclusions

Orgnac 3 in southeastern France is a key site for studying the transition between the Lower and Middle Paleolithic. Its 7-meter thick stratigraphic sequence has recorded the evolution of technical and subsistence behaviors of pre-Neanderthal human groups during a Middle Pleistocene interglacial-glacial cycle. The new TL and ESR/U-series data presented in this study confirm the attribution of the archaeological sequence to MIS 10–8, in agreement with the previous chronology based on U-series dates on speleothems. The overall chronological results make it possible to assign a reliable age to each archaeological level. Our results also make it possible to date the paleoanthropological levels (AL 6 and AL 5) to ca. 270–280 ka and the corresponding human occupations during a temperate and humid period evidenced by the biomarker record. Considering these results, this temperate humid period probably corresponds to the Amargiers interstadial, highlighted in the nearest palynological records from Velay, and mainly characterized first by the regional development of a forest landscape that gradually became more open. In this context of an early glacial stage, the appearance of Levallois debitage in the Orgnac 3 sequence appears to have been progressive and is probably the result of a long evolution rather than an abrupt event.

Acknowledgments

This geochronological study of Orgnac 3 was initiated in the late 1980s by Yuji Yokoyama at the request of Henry de Lumley and Jean Combier and we have benefited from their help and experience throughout our study. This article is dedicated to our colleague Jean Combier who died in 2020, victim of the Covid-19 epidemic. The excavations at Orgnac 3 were realized with the financial support of the Service Régional de l'Archéologie Rhone-Alpes. We would also thank the Andrea B. Taylor, Co-Editor-in-Chief, and the guest editors of this special issue, for their help and support, with special thanks to Nick Ashton for greatly improving the English. The present study was financially supported by the Service Régional de l'Archéologie Rhone-Alpes. The ESR and two gamma-ray spectrometers used in the present study were purchased with the financial support of the Sesame Île-de-France program, the Région Centre' and the ANR 'BcDIV program.

References

- Adler, D. S., Wilkinson, K. N., Blockley, S., Mark, D. F., Pinhasi, R., Schmidt-Magee, B. A., Nahapetyan, S., Mallol, C., Berna, F., Glauberman, P. J., Raczynski-Henk, Y., Wales, N., Frahm, E., Jöris, O., MacLeod, A., Smith, V. C., Cullen, V. L., Gasparian, B., 2014. Early Levallois technology and the Lower to Middle Paleolithic transition in the Southern Caucasus. *Science* 345, 1609-1613.
- Aitken, M.J., 1985. *Thermoluminescence Dating*. Academic Press, London.

Aouraghe, H. 1999. Reconstitution du paléoenvironnement les grands mammifères par les faunes du Pléistocène Moyen d'Orgnac 3 (Ardèche, France). *L'Anthropologie* 103, 177-184.

Bahain, J.-J., Yokoyama, Y., Falguères, C. Sarcia, M.N., 1992. ESR dating of tooth enamel: A comparison with K-Ar dating, *Quat. Sci. Rev.* 11, 245-250.

Bahain, J.-J., Falguères, C., Laurent, M., Dolo, J.-M., Shao, Q., Auguste, P., Tuffreau, A., 2015. ESR/U-series dating of faunal remains from the paleoanthropological site of Biache-Saint-Vaast (Pas-de-Calais, France). *Quat. Geochronol.* 30, 541-546.

Bischoff, J. L., Rosenbauer, R. J., Tavano, A., Lumley, H. de, 1988. A test of uranium-series dating of fossil tooth enamel: Results from Tournal cave, France. *Appl. Geochem.* 3, 135–141.

Brennan, B.J., Rink, W.J., McGuirl, E.L., Schwarcz, H.P., Prestwich, W.V., 1997. Beta doses in tooth enamel by “One Group” theory and the Rosy ESR dating software. *Radiat. Meas.* 27, 307–314

Combiér, J., 1967. Le Paléolithique de l'Ardèche dans son cadre paléoclimatique. Publications de l'Institut de Préhistoire de l'Université de Bordeaux, Mémoire n° 4.

Combiere, J., 2005. Quelle présence humaine dans l'espace rhodanien avant le stade 5 (135 ka) ? In 150 ans de Préhistoire autour de Lyon, Cahiers scientifiques du Muséum d'histoire naturelle de Lyon. Hors-série, 3, 71-99.

Debard, E., 1988. Le Quaternaire du Bas-Vivarais d'après l'étude des remplissages d'ovens, de porches de grottes et d'abris sous roche. Dynamique sédimentaire, paléoclimatologie et chronologie. Travaux et Documents des Laboratoires de Géologie de Lyon, n° 103.

DeCanniere, P., Debuyst, R., Dejehet, F., Apers, D., Grün, R., 1986. ESR dating: A study of ²¹⁰Po-coated geological and synthetic sample. Nuc. Tracks Rad. Meas. 11, 211–220.

de Lumley, M.A., 1981. Les restes humains d'Orgnac 3. In: Les Premiers Habitants de l'Europe (1,500,000–100,000 ans). Laboratoire de Préhistoire du Musée de l'Homme, Paris, pp. 143-145.

Dirks, P.H.G.M., Roberts, E.M., Hilbert-Wolf, H., Kramers, J.D., Hawks, J., Dosseto, A., Duval, M., Elliott, M., Evans, M., Grün, R., Hellstrom, J., Herries, A.I.R., Joannes-Boyau, R., Makhubela, T.V., Placzek, C. J., Robbins, J., Spandler, C., Wiersma, J., Woodhead, J., Berger, L.R., 2017. The age of *Homo naledi* and associated sediments in the Rising Star Cave, South Africa. eLife, 6, e24231.

Douze, K., Delagnes, A., 2016. The pattern of emergence of a Middle Stone Age tradition at Gademotta and Kulkuletti (Ethiopia) through convergent tool and point technologies. *J. Hum. Evol.* 91, 93-121.

Falguères, C., 2020. The first human settlements out Africa into Europe: A chronological perspective. *Quat. Sci. Rev.* 247, 106551.

Falguères, C., Shen, G., Yokoyama, Y., 1988. Datation de l'Aven d'Orgnac III: Comparaison par les méthodes de la résonance de spin électronique (ESR) et du déséquilibre des familles de l'uranium. *L'Anthropologie* 92, 727-730.

Falguères, C., Bahain, J.-J., Saleki, H., 1997. U-series and ESR dating of teeth from Acheulian and Mousterian levels at La Micoque (Dordogne, France). *J. Archaeol. Sci.* 24, 537-545.

Falguères, C., Bahain, J.-J., Yokoyama, Y., Arsuaga, J.L., Bermúdez de Castro, J.M., Carbonell, E., Bischoff, J.L., Dolo, J.M., 1999. Earliest humans in Europe: The age of TD6 Gran Dolina, Atapuerca, Spain. *J. Hum. Evol.* 37, 343-352.

Gauthier, A., 1992. Paléoenvironnement du Pléistocène moyen dans le sud de la France. Apport et limite de l'analyse pollinique de trois sites préhistoriques: Caune de l'Arago, Orgnac 3, grotte du Lazaret. Ph.D. Dissertation, Muséum National d'Histoire Naturelle.

Grün, R., 1986. Beta-dose attenuation in thin layers. *Ancient TL* 4, 1-8.

Grün, R., Katzenberger-Apel, O., 1994. An alpha irradiator for ESR dating, *Ancient TL* 12, 35–38.

Grün, R., Stringer, C.B., 1991. Electron spin resonance dating and the evolution of modern humans. *Archaeometry* 33, 2, 153-199.

Grün, R., Stringer, C.B., 2000. Tabun revisited: Revised ESR chronology and new ESR and U-series analyses of dental material from Tabun C1. *J. Hum. Evol.* 39, 601–612.

Grün, R., Schwarcz, H.P., Chadam, J.M., 1988. ESR dating of tooth enamel: Coupled correction for U-uptake and U-series disequilibrium. *Nuc. Tracks Rad. Meas.* 14, 237-241.

Grün, R., Stringer, C.B., Schwarcz, H.P., 1991. ESR dating of teeth from Garrod's Tabun cave collection. *J. Hum. Evol.* 20, 231–248.

Guérin, G., Mercier, N., Adamiec, G., 2011. Dose-rate conversion factors: Update. *Ancient TL* 29, 5–8.

Harvati, K., Röding, C., Bosman, A. M., Karakostis, F. A., Grün, R., Stringer, C., Karkanas P., Thompson N.C., Koutoulidis V., Moulopoulos L. A., Gorgoulis V.G., Kouloukoussa, M., 2019. Apidima Cave fossils provide earliest evidence of *Homo sapiens* in Eurasia. *Nature* 571, 500-504.

Ingicco, T., Van Den Bergh, G.D., Jago-On, C., Bahain, J.-J., Chacón, M. G., Amano, N., Forestier, H., King, C., Manalo, K., Nomade, S., Peirera, A., Reyes, M., Sémah, A-M., Shao, Q., Voinchet, P., Falguères, C., Albers, P., Lising, M., Lyras, G., Yurnaldi, D., Rochette, P., Bautista, A., De Vos, J., 2018. Earliest known hominin activity in the Philippines by 709 thousand years ago. *Nature* 557, 233–237.

Jaubert, J., Verheyden, S., Genty, D., Soulier, M., Cheng, H., Blamart, D., Burlet, C., Camus, H., Delaby, S., Deldicque, D., Edwards, R. L., Ferrier, C., Lacrampe-Cuyaubère, F., Lévêque, F., Maksud, F., Mora, P., Muth, X., Régnier, É., Rouzaud, J.-N., Santos, F., 2016. Early Neanderthal constructions deep in Bruniquel Cave in southwestern France. *Nature* 534, 111-114.

Joron, J.-L., 1974. Contribution à l'analyse des éléments en traces dans les roches et les minéraux par activation neutronique. Application à la caractérisation d'objets archéologiques. Ph.D. Dissertation, Université Paris-Sud.

Khatib, S., 1989. Le site d'Orgnac 3 (Ardèche, France). Étude sédimentologique et géochimique. Cadre chronologique et évolution paléoclimatique. Ph.D. Dissertation, Museum national d'histoire naturelle.

Khatib, S., 1994 Datations des cendres volcaniques et analyses géochimiques du remplissage d'Orgnac 3 (Ardèche, France). *Quaternaire* 5, 13-22.

Lisiecki, L.E., Raymo, M.E., 2005. A Plio-Pleistocene stack of 57 globally distributed benthic ^{18}O Records. *Paleoceanography* 20, Pa1003.

Masaoudi, H., 1995. Application des méthodes du déséquilibre des familles de l'uranium ($^{230}\text{Th}/^{234}\text{U}$) et de la résonance de spin électronique (ESR) à la datation des sites d'Ornac 3, Payre et de l'abri des Pêcheurs (Ardèche). Ph.D. Dissertation, Muséum national d'Histoire naturelle.

Mercier, N., Valladas, H., 2003. Reassessment of TL age estimates of burnt flints from the Paleolithic site of Tabun Cave, Israel. *J. Hum. Evol.* 45, 401–409.

Mercier, N., Valladas, H., Valladas, G., 1992. Observations on palaeo-dose determination with burnt flints. *Ancient TL* 10, 28–32.

Mercier, N., Valladas, H., Valladas, G., 1995a. Flint thermoluminescence dates from the CFR laboratory at Gif: Contributions to the study of the chronology of the Middle Palaeolithic. *Quat. Sci. Rev.* 14, 351-364.

Mercier, N., Valladas, H., Valladas, G., Reyss, J.-L., Jelinek, A., Meignen, L., Joron, J.-L., 1995b. TL dates of burnt flints from Jelinek's excavations at Tabun and their implications. *J. Archaeol. Sci.* 22, 495–509.

Michel, V., Shen, G., Shen, C., Fornari, M., Verati, C., Gallet, S., Sabatier, D., 2011. Les derniers *Homo heidelbergensis* et leurs descendants les néandertaliens: Datation des sites d'Orgnac 3, du Lazaret et de Zafarraya. C.R. Palevol 10, 577-587.

Michel, V., Shen, G., Shen, C., Wu, C., Vérati, C., Gallet, S., Moncel, M.-H., Combier, J., Khatib, S., Manetti, M., 2013. Application of U/Th and ⁴⁰Ar/³⁹Ar Dating to Orgnac 3, a Late Acheulean and Early Middle Palaeolithic Site in Ardèche, France. PLoS One, 8, e82394.

Moigne, A.-M., Moncel, M.-H., 2005. Données nouvelles sur les restes fauniques et lithiques dans les différents niveaux d'occupation du site d'Orgnac 3 (Ardèche, Sud-Est France): Types d'occupation. In: Molines, N., Moncel, M.-H., Monnier, J.-L. (Eds.), Les Premiers Peuplements en Europe. British Archaeological Reports, International Series, S1364, 215-226.

Moigne, A.-M., Auguste, P., Valensi, P., Solano, J.G., Tuffreau, A., Lamotte, A., Barroso, C., Moncel, M.-H., 2016. Bone retouchers from Lower Palaeolithic sites. A focus on Terra Amata, Orgnac 3, Cagny-l'Épinette and Cueva del Angel. Quat. Int. 409, 195-212.

Moncel, M.-H., Moigne, A.-M., Combier, J., 2005. Pre-Neandertal behavior during isotopic stage 9 and the beginning of stage 8: New data concerning fauna and lithics in the different occupation levels of Orgnac 3 (Ardèche, South-East France): Occupation types. J. Archaeol. Sci. 32, 1283-1301.

Moncel, M.-H., Moigne, A.-M., Combier, J., 2012. Towards the Middle Palaeolithic in Western Europe: The case of Orgnac 3 (southeastern France). *J. Hum. Evol.* 63, 653-666

Moncel, M.-H., Ashton, N., Arzarello, M., Fontana, F., Lamotte, A., Scott, B., Muttillo, B., Berruti, B., Nenzioni, G., Tuffreau, A., Peretto, C., 2020. Early Levallois core technology between Marine Isotope Stage 12 and 9 in Western Europe. *J. Hum. Evol.* 139, 102735.

Moncel, M.-H., Garcia Medrano, P., Despriée, J., Arnaud, J., Voinchet, P., Bahain, J.-J. 2021. Tracking behavioral persistence and innovations during the Middle Pleistocene in Western Europe. Shift in occupations between 700 ka and 450 ka at la Noira site (Centre, France). *J. Hum. Evol.* 156, 103009

Pereira, A., Nomade, S., Moncel, M.-H. Voinchet, P., Bahain, J.-J., Biddittu, I., Falguères, C., Giaccio, B., Manzi, G., Parenti, F., Scardia, G., Scao, V., Sottili, G., Vietti, A., 2018. Integrated geochronology of Acheulian sites from the southern Latium (central Italy): Insights on human-environment interaction and the technological innovations during the MIS 11-MIS 10 period. *Quat. Sci. Rev.* 187, 112-129.

Prescott, J.R., Hutton, J.T., 1994. Cosmic ray contributions to dose rates for luminescence and ESR dating: Large depths and long-term time variations. *Radiat. Meas.* 23, 497-500,

Reille, M., Andrieu, V., De Beaulieu, J.-L., Guenet, P., Goeury, C., 1998. A long pollen record from Lac du Bouchet, Massif Central, France: For the period ca. 325 to 100 ka (OIS 9c to OIS 5e). *Quat. Sci. Rev.* 17, 1107–1123.

Richter, D., Grün, R., Joannes-Boyau, R., Steele, T.E., Amani, F., Rué, M., Fernandes, P., Raynal, J.P., Geraads, D., Ben-Ncer, A., Hublin, J.-J., McPherron, S.P., 2017. The age of the hominin fossils from Jebel Irhoud, Morocco, and the origins of the Middle Stone Age. *Nature* 546, 293-296.

Roberts, R.G., Jacobs, Z., Li, B., Jankowski, N.R., Cunningham, A.C., Rosenfeld, A.B., 2015. Optical dating in archaeology: Thirty years in retrospect and grand challenges for the future. *J. Archaeol. Sci.* 56, 41-60.

Roger, S., Feraud, G., De Beaulieu, J.-L., Thouveny, N., Coulon, C., Cocheme, J.-J., Andrieu, V., Williams, T. 1999. $^{40}\text{Ar}/^{39}\text{Ar}$ dating on tephra of the Velay maars (France): Implications for the Late Pleistocene proxy-climatic record. *Earth Planet. Sci. Lett.* 170, 287-299.

Shao, Q., 2011. Combined ESR/U-series dating of fossil teeth from Middle Pleistocene sites in Northern Europe and Mediterranean Area: Contributing to the chronology of the Acheulian settlement of Europe. Ph.D. Dissertation, Museum national d'histoire naturelle.

Shao, Q., Bahain, J.-J., Falguères, C., Dolo, J.-M., Garcia T., 2012. A new U-uptake model for combined ESR/U-series dating of tooth enamel. *Quat. Geochronol.* 10, 406-411.

Shao, Q., Bahain, J.-J., Wang, W., Jin, C., Wang, Y., Voinchet, P., Lin, M., 2015. Combined ESR and U-series dating of early Pleistocene *Gigantopithecus* faunas at Mohui and Sanhe Caves, Guangxi, southern China. *Quat. Geochronol.* 30, 524-528.

Shen, G., 1985. Datation des planchers stalagmitiques de site acheuléens en Europe par les méthodes des déséquilibres des familles de l'uranium et contribution méthodologique. Ph.D. Dissertation, University Paris VI.

Sutikna, T., Tocheri, M.W., Morwood, M.J., Wahyu Saptomo, E., Jatmiko, Due A., R., Wasisto, S., Westaway, K. E., Aubert, M., Li, B., Zhao, J., Storey, M., Alloway, B.V., Morley, M.W., Meijer, H.J.M., van den Bergh, G.D., Grün, R., Dosseto, A., Brumm, A., Jungers, W.L., Roberts, R.G., 2016. Revised stratigraphy and chronology for *Homo floresiensis* at Liang Bua in Indonesia. *Nature* 532, 366–369.

Tryon, C.A., Faith, J.T., 2013. Variability in the Middle Stone Age of eastern Africa. *Curr. Anthropol.* 54, S234-S254.

Valladas, H., 1992. Thermoluminescence dating of flint. *Quat. Sci. Rev.* 11, 1–5.

Valensi, P., Aouraghe, H., Baillon, S., Cauche, D., Combier, J., Desclaux, E., Gagnepain, J., Gaillard, C., Khatib, S., de Lumley, H., Moigne, A.-M., Moncel, M.-H., Notter, O., 2005. Les peuplements préhistoriques dans le Sud-Est de la France à la fin du Pléistocène Moyen: 400

- 120 000 ans. Terra Amata, Orgnac 3, Baume Bonne, Lazaret. Cadre géochronologique et biostratigraphique, paléoenvironnements et évolution culturelle des derniers anténéandertaliens. In: Tuffreau, A. (Ed.) Peuplements humains et variations environnementales au Quaternaire, British Archaeological Reports, International Series, S1352, 33-37.

Wiśniewski, A., 2014. The beginnings and diversity of Levallois methods in the early Middle Palaeolithic of Central Europe. *Quat. Int.* 326, 364-380.

Yokoyama, Y., Nguyen, H.V., Quaegebeur, J.-P., Poupeau, G., 1982. Some problems encountered in the evaluation of annual dose rate in the electron spin resonance dating of fossil bones. *PACT* 6, 103–115.

Zaidner, Y., Weinstein-Evron, M., 2020. The emergence of the Levallois technology in the Levant: A view from the Early Middle Paleolithic site of Misliya Cave, Israel. *J. Hum. Evol.* 144, 102785.

Zupancich, A., Shemer, M., Barkai, R., 2021. Biface use in the Lower Paleolithic Levant: First insights from late Acheulean Revadim and Jaljulia (Israel). *J. Archaeol. Sci.: Rep.* 36, 102877.

Figure legends

Figure 1. Location map of the Orgnac 3 site (Ardèche, France).

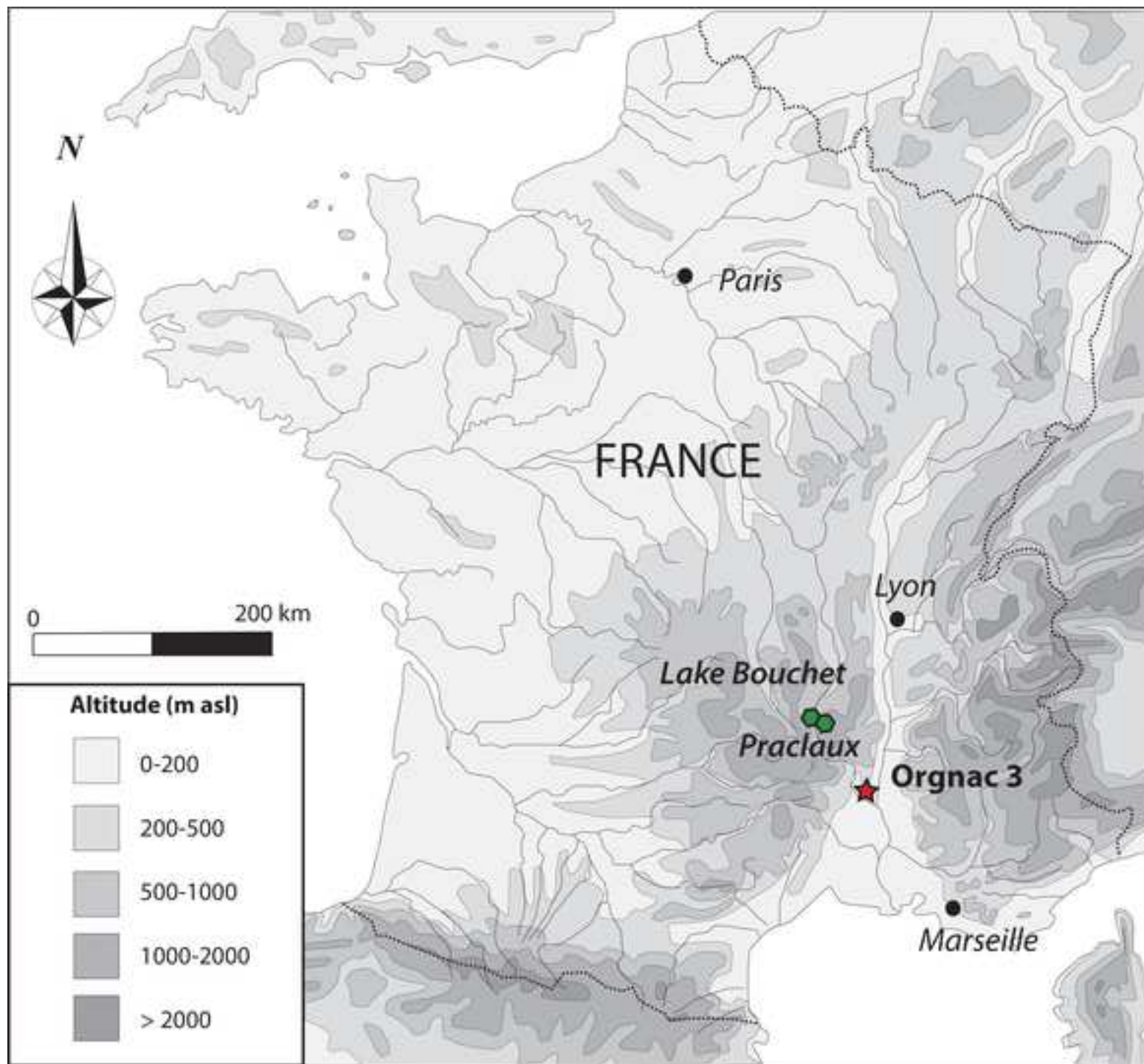
Figure 2. Stratigraphic, geochronological, paleoenvironmental, and archaeological data available on the Orgnac 3 site prior to the present study (modified after Michel et al., 2013), indicating sample location.

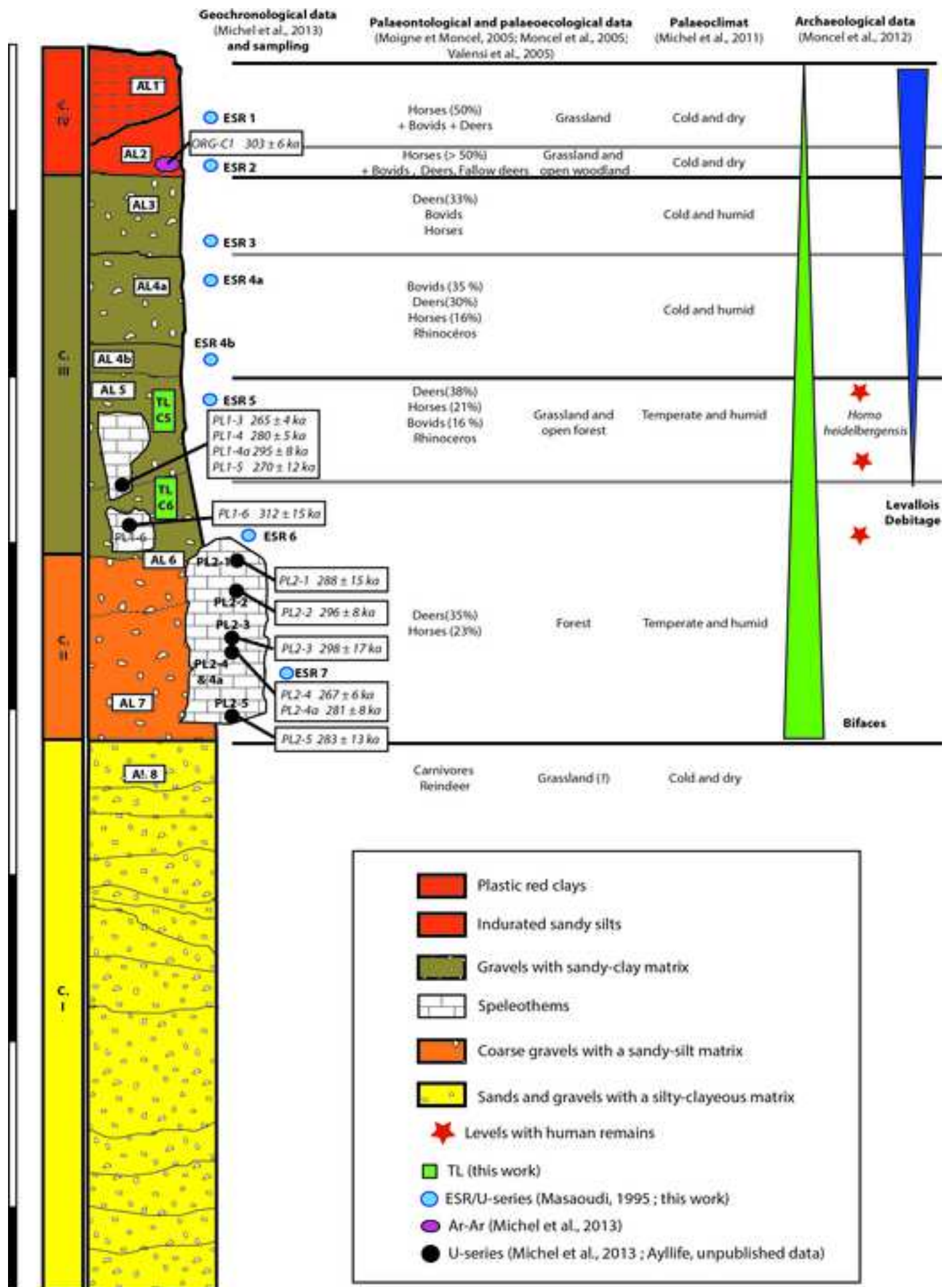
Figure 3. Lithic material from Orgnac 3. 1. Biface in Tertiary flint (level 5b); 2. Levallois flake in Tertiary flint (level 1); 3. Bone retoucher (level 5b); 4. Levallois core in Tertiary flint (level 1). Photos 1, 2 and 4 from M.-H. Moncel; photo 3 from A.-M. Moigne.

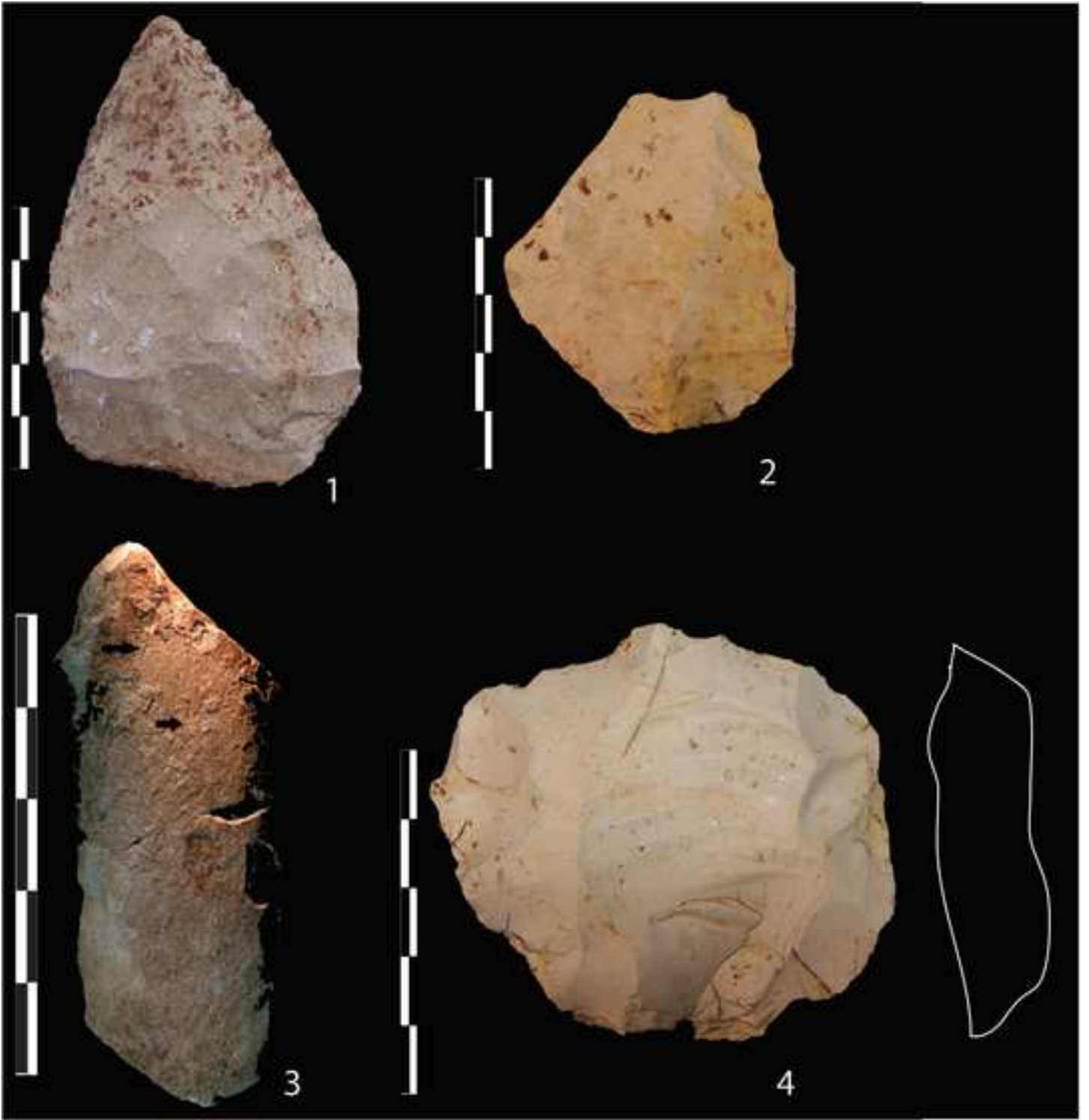
Figure 4. Gamma dose dosimetry and reconstruction proposed for the Orgnac 3 teeth. A sphere of 70 cm in diameter is considered for reconstruction using available radioelement contents or in situ dosimetric measurements to assess the relative contributions of the levels surrounding the sample under consideration.

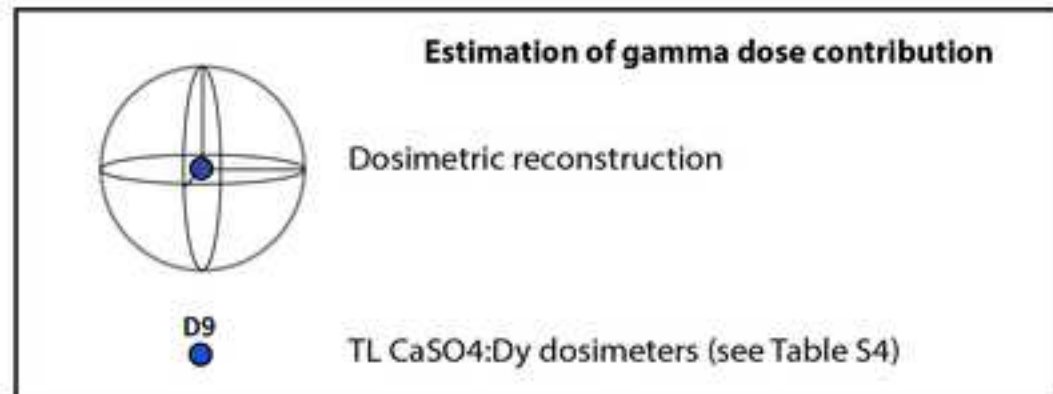
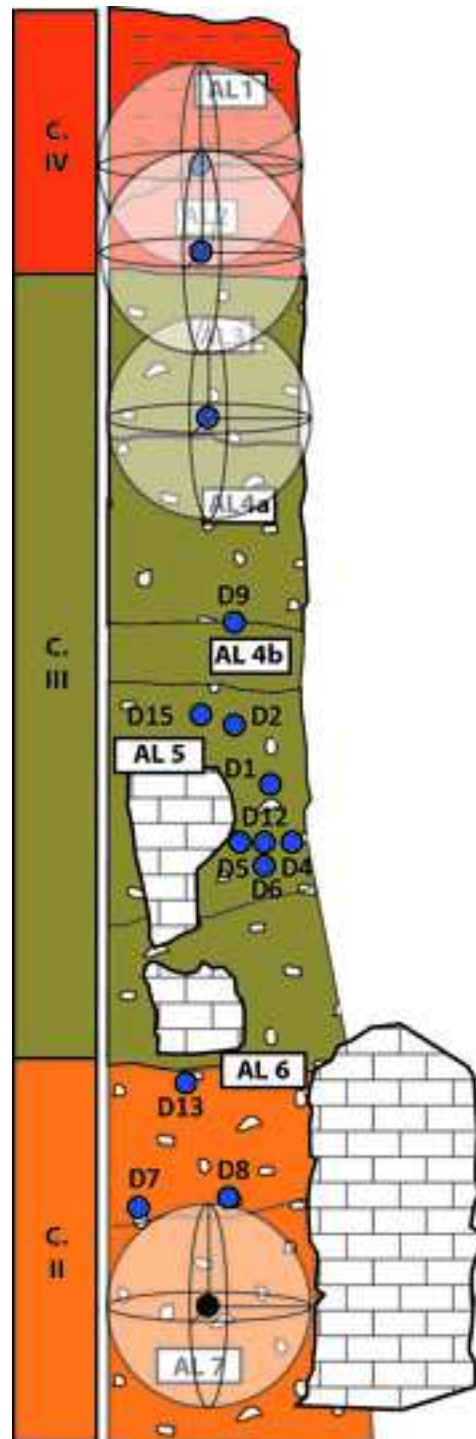
Figure 5. Synthesis of the geochronological data available on the Orgnac 3 site including from the current study and proposed paleoclimatic and paleoenvironmental correlation.

Figure 1









Estimation of gamma dose contribution by dosimetric reconstruction

D_{γ} AL 1 = 50% D_{γ} sed AL 1 + 50% D_{γ} sed AL 2

D_{γ} AL 2 = 10% D_{γ} sed AL 1 + 60% D_{γ} sed AL 2 + 30% D_{γ} sed AL 3

D_{γ} AL 3 = 50% D_{γ} sed AL 3 + 50% D_{γ} dos AL 4

D_{γ} AL 7 = 10% D_{γ} sed AL 6 + 90% D_{γ} sed AL 7

Lithology

-  Plastic red clays
-  Indurated sandy silts
-  Gravels with sandy-clay matrix
-  Speleothems
-  Coarse gravels with a sandy-silt matrix

Figure 5

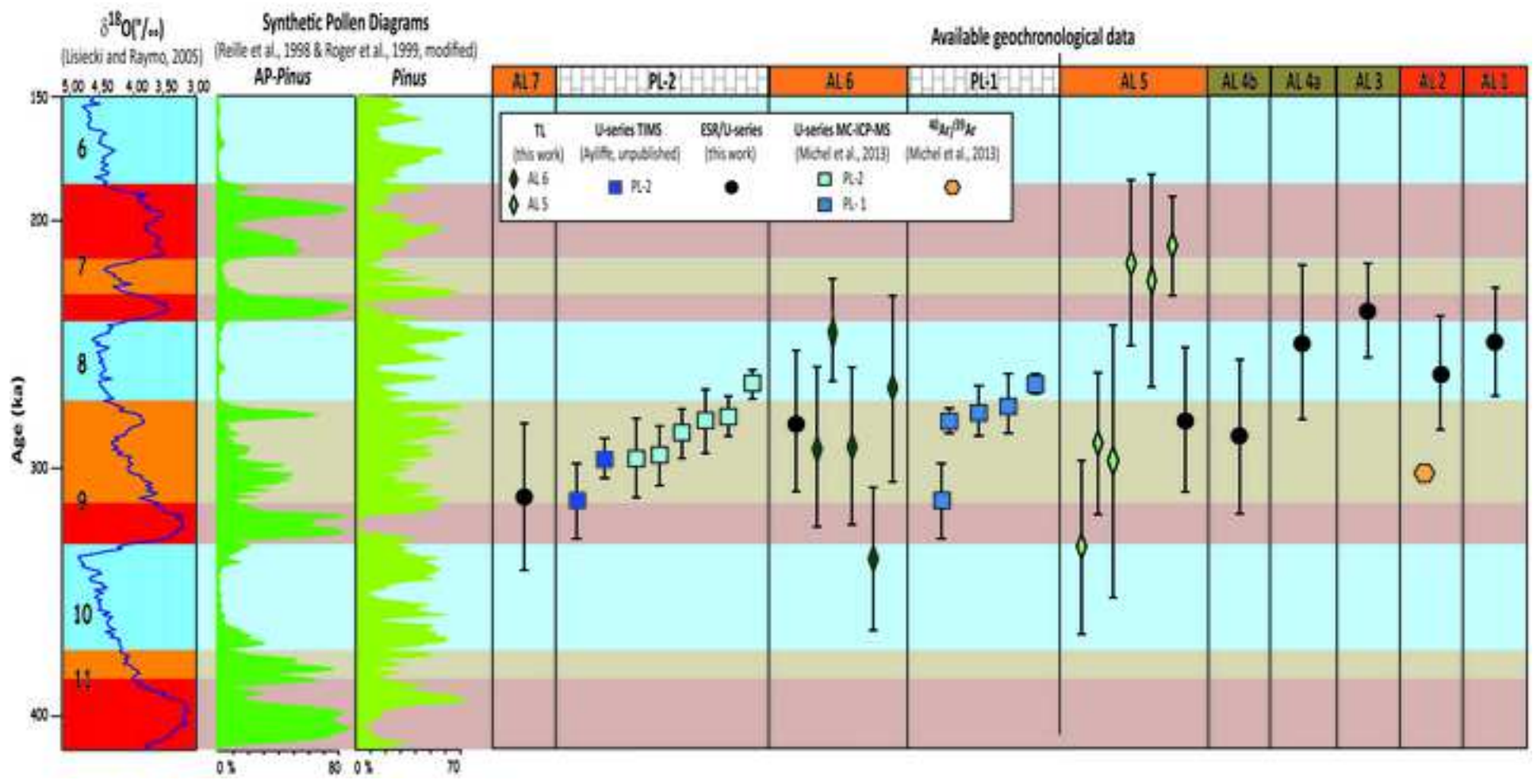


Table 1

Radiometric data and ages of burnt flints from Orgnac 3 analyzed by thermoluminescence (TL).

Archaeological level	Sample	Depth (cm)	U ^a (ppm)	Th ^a (ppm)	K ^a (%)	α -sens.	Internal (μ Gy/a)	External (μ Gy/a)	Annual (μ Gy/a)	Equivalent dose (Gy)	Age (ka)
AL 5	ORG13	261	0.490	0.086	0.028	24.0	303 \pm 28	539 \pm 46	842 \pm 54	189 \pm 34	224 \pm 43
	ORG19	253	0.500	0.089	0.027	22.8	300 \pm 28	706 \pm 61	1007 \pm 67	218 \pm 31	217 \pm 34
	ORG22	251	0.475	0.062	0.008	16.7	217 \pm 26	570 \pm 47	788 \pm 53	234 \pm 40	297 \pm 55
	ORG25	253	0.378	0.033	0.007	16.4	170 \pm 16	716 \pm 62	886 \pm 64	257 \pm 18	290 \pm 29
	ORG38	250	0.523	0.075	0.014	13.2	212 \pm 18	561 \pm 46	773 \pm 49	234 \pm 40	332 \pm 35
	ORG40		0.650	0.120	0.045	23.7	406 \pm 38	699 \pm 89	1105 \pm 97	214 \pm 15	210 \pm 20
AL 6	ORG30	305	0.271	0.092	0.0187	22.5	172 \pm 15	438 \pm 67	609 \pm 69	164 \pm 14	268 \pm 38
	ORG31	322	0.921	0.113	0.0231	13.9	172 \pm 33	771 \pm 45	943 \pm 56	318 \pm 20	337 \pm 29
	ORG35	322	0.795	0.05	0.0119	20.7	172 \pm 40	691 \pm 38	863 \pm 73	251 \pm 17	291 \pm 32
	ORG36	-	0.620	0.222	0.0454	25.8	172 \pm 39	600 \pm 30	772 \pm 50	188 \pm 11	244 \pm 21
	ORG39	350	0.271	0.049	0.0102	15.0	172 \pm 10	685 \pm 46	685 \pm 47	199 \pm 17	291 \pm 32

Abbreviation: α -sens. = α -sensitivity (expressed in μ Gy for 1000 α/cm^2).

^aContents of these samples were determined by neutron activation analysis and were assigned a systematic error of 10% (for each element). The α -sensitivity parameter quantifies the efficiency of alpha particles relative to beta rays to produce a TL signal. The internal dose rate was computed from the radioelement contents, the α -sensitivity and by using the conversion factors of Guérin et al. (2011). The annual dose includes the internal dose rate and the

external dose rate deduced from the dosimeter measurements. The external dose rate was computed from these last values, taking into account the past cosmic dose rate. The mean water content of the sediments during the past was estimated at 25%. For each sample, the closest dosimeters were used for assessing a representative external dose rate. The equivalent doses include a 3% systematic error related to the gamma source calibration used for applying the additive dose protocol. Errors on the ages were computed by combining the different errors quadratically. The ages are calculated with 1 σ error range.

Table 2Radiometric data and ages of Orgnac 3 teeth analyzed by ESR/U-series.^a

Samples	Depth (cm)	Tissue	U (ppm)	Equivalent dose (Gy)	U-uptake parameters (a. u.)		D_{α} ($\mu\text{Gy/a}$)	D_{β} ($\mu\text{Gy/a}$)	$D_{(\gamma + \text{cosm})}$ ($\mu\text{Gy/a}$)	ESR/U-series ages (ka)	
					p (US model)	n (AU model)				US model	AU model
Orgnac 1	-50	enamel	1.40 ± 0.14	494 ± 14	-0.45 ± 0.10		315 ± 99	538 ± 149	1139 ± 53		248 ± 22
		dentine	64.42 ± 6.40		-0.97 ± 0.04						
Orgnac 2	-80	enamel	0.48 ± 0.05	471 ± 11	0.10 ± 0.15		71 ± 31	479 ± 153	1253 ± 49		261 ± 23
		dentine	64.86 ± 6.49		-0.72 ± 0.07						
Orgnac 3	-120	enamel	0.55 ± 0.05	422 ± 13	-0.72 ± 0.07		166 ± 22	518 ± 131	1102 ± 70		236 ± 20
		dentine	63.46 ± 6.35		-0.87 ± 0.05						
Orgnac 4a	-160	enamel	1.19 ± 0.10	375 ± 6			405 ± 154	371 ± 90	728 ± 62		249 ± 31
		dentine	45.91 ± 4.59								

0.0009

Orgnac 4b	-180	enamel	0.46 ± 0.05	411 ± 17	-0.25 ± 0.14	84 ± 38	482 ± 159	865 ± 62	287 ± 31
		dentine	74.97 ± 7.50		-0.40 ± 0.12				
Orgnac 5	-210	enamel	0.69 ± 0.07	406 ± 8	1.91 ± 0.38	46 ± 19	494 ± 139	911 ± 62	280 ± 29
		dentine	111.05 ± 11.11		0.36 ± 0.20				
Orgnac 6	-295	enamel	1.21 ± 0.12	276 ± 5	0.72 ± 0.23	124 ± 55	251 ± 56	607 ± 62	281 ± 28
		dentine	66.57 ± 6.66		1.05 ± 0.27				
Orgnac 7	-370	enamel	0.48 ± 0.05	546 ± 37	-0.0037 ±	137 ± 89	402 ± 181	1219 ± 49	311 ± 30
		dentine	47.01 ± 4.70		0.0006				

^aEquivalent doses were determined using single exponential fitting with the additive dose method by Masaoudi (1995), who also performed U-series analyses using alpha spectrometry. Dose rates were determined from radioelement contents of the associated sediments using conversion factors from Guérin et al. (2011), except γ -dose rate of archaeological levels AL4, AL5 and AL6, measured in situ using CaSO₄:Dy dosimeters (1, 6 and 3 measurement points respectively). For the other levels, gamma dose reconstruction was made for the teeth according to their depth from sediments of the corresponding layer and, if necessary, overlying or underlying layers. The cosmic dose rate was determined from the equations given by Prescott and Hutton (1994). A k-value of 0.13 ± 0.02 was used in the α -dose rate calculation (Grün and Katzenberger-Appel, 1994). The β -dose rate was calculated taking into account Monte Carlo β -ray attenuations in the enamel (Brennan et al., 1997), and taking into account the initial and removed enamel thicknesses during the sample

preparation. The water content was estimated to be 0 wt % in the enamel, 7 ± 5 wt % in the dentine and 15 ± 5 wt % in the sediment. The ESR-U/Th ages are given with ± 1 sigma. U-uptake parameters ('p' for the US model and 'n' for the AU model) are expressed in arbitrary units (a. u.).

Supplementary Online Information (SOM):

Reappraisal of the chronology of Orgnac 3 Lower-Middle Palaeolithic site (Ardèche, France), a regional key sequence for the Middle Pleistocene of southern France

Jean-Jacques Bahain^{a,*}, Norbert Mercier^b, Hélène Valladas^c, Christophe Falguères^a, Hassan Masaoudi^a, Jean-Louis Joron^d, Laurence Froget^c, Anne-Marie Moigne^a, Jean Combier^e, Marie-Hélène Moncel^a

^a *UMR7194 HNHP MNHN-CNRS-UPVD, Département Homme et Environnement du Muséum national d'Histoire Naturelle, Institut de Paléontologie Humaine, 1 rue René Panhard, 75013, Paris, France*

^b *UMR 5060 IRAMAT CNRS-Université de Bordeaux, Centre de Recherche en Physique Appliquée à l'Archéologie, Maison de l'archéologie, 33607 Pessac cedex, France*

^c *UMR8212 LSCE/IPSL CEA-CNRS-UVSQ, Bât. 12, Avenue de la Terrasse, 91198 Gif-sur-Yvette Cedex, France*

^d *Groupe des Sciences de la Terre, Laboratoire Pierre Süe, CEN, Saclay, 91191 Gif-sur-Yvette, France*

^e *5, rue Saint-Jean, 71000 Mâcon, France*

***Corresponding author.**

Email address: bahain@mnhn.fr (J.-J. Bahain)

SOM Table S1

U-series data (α spectrometry) of the Orgnac 3 teeth analyzed by Masaoudi (1995) using standard methods described by Bischoff et al. (1988).

Samples	Tissue	U content				
		(ppm)	$^{234}\text{U}/^{238}\text{U}$	$^{230}\text{Th}/^{232}\text{Th}$	$^{230}\text{Th}/^{234}\text{U}$	$^{222}\text{Rn}/^{230}\text{Th}$
Orgnac 1	enamel	1.398 ± 0.072	1.178 ± 0.052	42	0.715 ± 0.042	1.000
	dentine	64.423 ± 1.033	1.201 ± 0.015	95	0.919 ± 0.018	0.258
Orgnac 2	enamel	0.480 ± 0.020	1.096 ± 0.039	94	0.607 ± 0.029	1.000
	dentine	64.864 ± 1.153	1.119 ± 0.015	>100	0.813 ± 0.018	0.311
Orgnac 3	enamel	0.548 ± 0.026	1.252 ± 0.047	52	0.792 ± 0.045	1.000
	dentine	63.462 ± 1.315	1.219 ± 0.015	100	0.858 ± 0.025	0.249
Orgnac 4a	enamel	1.190 ± 0.056	1.187 ± 0.056	46	0.971 ± 0.045	1.000
	dentine	45.913 ± 0.935	1.262 ± 0.020	>100	0.964 ± 0.024	0.096
Orgnac 4b	enamel	0.460 ± 0.022	1.053 ± 0.044	>100	0.701 ± 0.039	1.000
	dentine	74.965 ± 1.666	1.091 ± 0.016	60	0.739 ± 0.022	0.232
Orgnac 5	enamel	0.690 ± 0.026	1.108 ± 0.027	68	0.422 ± 0.022	1.000
	dentine	111.046 ± 2.949	1.041 ± 0.016	>100	0.583 ± 0.022	0.351
Orgnac 6	enamel	1.208 ± 0.079	1.025 ± 0.061	28	0.535 ± 0.038	1.000
	dentine	66.574 ± 1.251	0.970 ± 0.013	>100	0.496 ± 0.012	0.317
Orgnac 7	enamel	0.480 ± 0.023	1.184 ± 0.054	>100	0.893 ± 0.045	1.000
	dentine	47.007 ± 1.422	1.205 ± 0.024	>100	1.084 ± 0.044	0.412

SOM Table S2

ESR/U-series ages obtained for the Orgnac 3 teeth as given in Masaoudi (1995).^a

Archaeological level	Depth (cm)	D _E (Gy)	Initial enamel thickness (μm)	Removed enamel thickness (μm) on each side	ESR/U-series ages US-model (ka)
1	-50	494 ± 14	1125 ± 56	100 ± 5	294 ± 44
2	-80	471 ± 11	1281 ± 64	100 ± 5	212 ± 31
3	-120	422 ± 13	1406 ± 70	100 ± 5	Not calculable
4a	-160	375 ± 6	1281 ± 64	100 ± 5	Not calculable
4b	-180	411 ± 17	1156 ± 56	100 ± 5	169 ± 25
5	-210	406 ± 8	1156 ± 56	100 ± 5	132 ± 19
6	-295	276 ± 5	1438 ± 72	100 ± 5	162 ± 24
7	-370	546 ± 37	1531 ± 76	100 ± 5	Not calculable

^a Unfortunately, the detailed dose contributions and U-uptake parameters are not given in the original document by Masaoudi (1995). Note also that the removed enamel thickness on each side was estimated by Masaoudi (1995), while it was measured accurately in more recent work.

SOM Table S3Gamma dose contribution measured in situ by CaSO₄ dosimeters.^a

Layer	Dosimeter	Depth (cm)	Duration of exposure (days)	In situ γ dose ($\mu\text{Gy/a}$)
4	D9	190	1057	558.5
4b/5a	D15	215	365	634.5
4b/5a	D2	220	693	774.0
4b/5a	D1	237	693	849.7
Mean value				752.7
5b	D5	255	693	669.0
5b	D12	255	365	526.5
5b	D4	255	693	577.1
5b	D6	260	693	562.1
Mean value				583.7
6	D13	315	365	472.7
6	D8	338	693	315.7
6	D7	340	693	609.6
Mean value				466.0

^a The statistical error on a dose rate recorded by each dosimeter is generally small (because the count is high). Due to the uncertainty associated with the calibration of the source used to estimate the dose rate, an error of $\pm 5\%$ is attributed to each individual measurement.

SOM Table S4

Radioelement contents of the Orgnac 3 sediments measured by γ spectrometry (Masaoudi, 1995).

Sediment samples	U (ppm)	Th (ppm)	K (%)
Orgnac 1	1.650 \pm 0.075	5.553 \pm 0.046	0.343 \pm 0.01
Orgnac 2	4.014 \pm 0.189	11.982 \pm 0.114	1.034 \pm 0.031
Orgnac 3	5.604 \pm 0.159	10.450 \pm 0.084	1.750 \pm 0.028
Orgnac 4a	3.550 \pm 0.132	12.176 \pm 0.080	1.199 \pm 0.020
Orgnac 4b	4.180 \pm 0.185	10.610 \pm 0.111	1.183 \pm 0.027
Orgnac 5	4.243 \pm 0.168	11.685 \pm 0.103	1.366 \pm 0.260
Orgnac 6	5.483 \pm 0.132	9.718 \pm 0.072	1.010 \pm 0.018
Orgnac 7	5.233 \pm 0.193	9.154 \pm 0.103	1.149 \pm 0.026

SOM References

Bischoff, J. L., Rosenbauer, R. J., Tavano, A., de Lumley, 1988. A test of uranium-series dating of fossil tooth enamel: Results from Tournal cave, France. *Appl. Geochem.* 3, 135–141.

Masaoudi, H., 1995. Application des méthodes du déséquilibre des familles de l'uranium ($^{230}\text{Th}/^{234}\text{U}$) et de la résonance de spin électronique (ESR) à la datation des sites d'Ornac 3, Payre et de l'abri des Pêcheurs (Ardèche). Ph.D. Dissertation, Muséum national d'Histoire naturelle.