

New ESR and U-series dating at Caune de l'Arago, France: A key-site for European Middle Pleistocene

C Falguères, Q Shao, F Han, J J Bahain, M Richard, C Perrenoud, A M Moigne, H Lumley De

▶ To cite this version:

C Falguères, Q Shao, F Han, J J Bahain, M Richard, et al.. New ESR and U-series dating at Caune de l'Arago, France: A key-site for European Middle Pleistocene. Quaternary Geochronology, 2015, 30, pp.547 - 553. 10.1016/j.quageo.2015.02.006 . hal-03739357

HAL Id: hal-03739357 https://cnrs.hal.science/hal-03739357

Submitted on 13 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.

New ESR and U-series dating at Caune de l'Arago, France: a key-site for European Middle Pleistocene

Falguères C.1*, Shao Q.1,2, Han F.3, Bahain J.J.1, Richard M.1, Perrenoud C.1,4, Moigne A.M.1,4, Lumley de H.1,

Abstract

The Caune de l'Arago, located at Tautavel in the southern part of France, is one of the best documented Middle Pleistocene sites allowing a good understanding of human evolution in Europe. Since its discovery in 1829, the cave yielded more than 140 human remains associated with abundant lithic industries and thousands of faunal remains in a 10 meter thick stratigraphic sequence divided in three complexes (Lumley et al., 2014). The Lower stratigraphic complex is only known from cores while the Middle and Upper ones can be divided into about 17 main archaeological levels indexed from the bottom to the top: from level Q to level A. Since 1981, a number of dates were done using practically all the methods available for dating Quaternary period up to 700 ka. U-series dates performed on the upper stalagmitic floor yielded a minimum age of 400 ka for human remains found in the underlying level G (Falguères et al., 2004). This age range confirmed the direct non-destructive gamma-ray age published more than 30 years before (Yokoyama et Nguyen, 1981). Recently, a methodological work was published on herbivorous teeth coming from different parts of the G level highlighting the difficulties to get reliable radiometric dates on a level so rich in bones and partly weathered by guano deposits or issues associated with carbonate accumulation (Han et al., 2010).

This study presents new ESR/U-Series dates obtained on herbivorous teeth coming from the lowest excavated layers (Q-P levels) which yield an age range for the entire thick archaeological infilling. The new results suggest that the sedimentological levels containing lithic and faunal artefacts began to be deposited at 550 ka in the P-Q levels and at a younger age of 350-400 ka for the F level. The P-Q levels which have yielded bifaces are one of the oldest lines of evidence for the Acheulian culture in the southern part of Western Europe, while the dates obtained on G level suggest contemporaneity with la Sima de los Huesos and TD10 Dolina level at Atapuerca.

Key-words: combined ESR/U-Series, Middle Pleistocene, Arago, Acheulian.

Highlights

ESR/U-series on teeth within the entire sequence of Arago Comparison of the radiometric results with biochronological data Importance of a chronological framework of Arago for human evolution in Europe during Middle Pleistocene

¹ Département de Préhistoire, Muséum national d'histoire naturelle, UMR7194, 1, rue René Panhard, 75013, Paris, France

² Department of Geography Sciences – Nanjing Normal University, Nanjing, Jiangsu, China

³ State Key Laboratory of Earthquake Dynamics, Institute of Geology, China Earthquake Administration, Beijing 100029, China

⁴ Département de Préhistoire, Muséum national d'histoire naturelle, UMR7194, Centre Européen de Recherches Préhistoriques, avenue Grégory, 66720, Tautavel, France

^{*} Corresponding author: falguere@mnhn.fr

Introduction

The Caune de l'Arago, located in southern France, is one of the most important Middle Pleistocene archaeological sites in Europe. The stratigraphic sequence, and essentially the Middle Complex, has yielded 148 human remains associated with around 120 faunal species (herbivorous, carnivorous, rodents, birds, lagomorphs...) and a number of lithic artefacts documenting the major part of the Middle Pleistocene period.

The chronology of European humans is one of the major issues in human evolution. It is essential, in terms of chronology, to understand if there is a correlation between the Early Pleistocene hominins at Elefante (Carbonell et al., 2008), Barranco Leon (Toro Moyano et al., 2013) and Gran Dolina (Carbonell et al., 1995) and later populations present in Europe, particularly those at Arago and la Sima de los Huesos (SH) with an intermediate site represented by Isernia la Pineta, Italy (Galotti and Peretto, 2014). This topic was discussed in Wagner et al., (2010) including references therein. Arago and SH are being excavated today and many fossils continue to be unearthed rendering this problem complex. Arago Man is one of the main actors since, together with SH hominins and Mauer, they are chronologically the followers of *Homo antecessor*, which they probably replaced during Middle Pleistocene according to Carbonell et al., (2005). On the other hand, these fossils could be considered of utmost importance regarding the emergence of Neandertals before 430 ka (Arsuaga et al., 2014). The recent discoveries at northern sites such as the Happisburgh footprints site (Ashton et al., 2014) and La Noira (France), including handaxes at more than 650 ka (Moncel et al., 2013), contribute to narrow the gap between the settlements making industries with core and flake tools (Mode 1) and those who brought Acheulian handaxes (Mode 2) like SH and Arago which are possibly associated with Boxgrove finds.

Arago cave is also important for its wide faunal spectrum providing important climatic and palaeoenvironmental information (Moigne et al., 2006; Rivals and Deniaux, 2003). For instance, large herbivores found (around 180,000 remains) form a typical Middle Pleistocene faunal assemblage known to be present at the beginning of this period in the Mediterranean context: *Hemitragus bonali, Palaeoloxodon antiquus, Ursus deningeri, Cervus elaphus, Dama* sp., but mainly a number of animals coming from several dispersal phases in bonds with the significant cooling associated with MIS12 and 14, especially in the Pyrenees area. This faunal assemblage is marked by a cold and dry environment which is unique in Mediterranean zone and is similar to that of Northen sites which have provided records during interglacial periods.

Arago cave is the most representative site documenting a huge dispersal episode with *Praeovibos priscus, Rangifer tarandus, Bison priscus, Equus mosbachensis, Stephanorhinus hemitoechus, Cuon priscus* up to the Northern part of Pyrenees and the migration paths followed also by South-Asiatic carnivorous species coming from mountainous areas of Central Asia, or representatives of mammal families originated from Siberia (Palombo, 2007).

Since 1981, an incredible number of chronometric analyses were performed on bones, teeth, quartz, calcite which first yielded a chronostratigraphical scheme that was hotly debated (Lumley et Labeyrie, 1981). Many analyses using different methods were applied for the first time adding to the difficulties for having a clear vision of the age of Arago Man. ESR and U-series methods, using alpha and gamma ray spectrometry, were independently applied on

calcite, bones and teeth, particularly on the G level in which many human remains were found mixed with animal remains and lithic artefacts. Additionally, the G soil is partly altered by guano bat deposits complicating the dating interpretations.

In this work, herbivorous teeth were analyzed, some of them being already published (Han et al, 2010), and ages calculated using ESR/U-series (US or AU) models. The samples come from the top, F level, to the bottom, P/Q level of Middle Complex corresponding to the oldest hominid-bearing occupation levels found in the cave and where recent excavations have provided a number of lithic artefacts attesting the presence of Acheulian technology at an early stage in the Middle Pleistocene. For the first time, a chronological scheme is proposed for the entire archaeological sequence providing an idea of the human occupation time span.

Stratigraphy

The stratigraphical infill exhibits a 10 meters thick sequence revealing the alternation of humid-temperate and drycold phases. Three complexes have been formally defined:

The Lower stratigraphical Complex is known through analysis of 5 cores. It is composed of silty sandy clays deposited by dripping water, on top of a stalagmitic floor at the bottom. This speleothem was dated by ESR method at about 700 ka (Yokoyama et al., 1982).

The Middle stratigraphical Complex is constituted by three distinct ensembles. At the bottom, Ensemble I is made mainly with stratified sands suggesting a cold and dry period in agreement with the presence of the reindeer, *Rangifer tarandus*. The Ensemble II is composed of a clayey sandy silt which was deposited during a more humid and temperate period. The top of this Complex is made by the Ensemble III constituted by coarse stratified sands accumulated by wind during a cold and dry period.

The Upper stratigraphical Complex, 1 meter thick, is composed of alternating of stalagmitic floors and silty-clayey sands suggesting climatic variations.

The stalagmitic floor at the base of Upper Complex was dated by U-series, yielding a minimum age of 350-400 ka for the underlying levels containing human remains while the most recent deposits are probably contemporaneous with the MIS 5 (Falguères et al., 2004).

Material and methods

The combined ESR/U-series dating approach takes into account both ESR and U-series data and, including radioelement contents, isotopic ratios, palaeodoses and external gamma-dose rate. The relevance of the method was discussed in Grün (2009a). The application of this combined approach allows dating of the whole Middle Pleistocene period (Falguères et al., 2010; Bahain et al., 2007). The US-ESR model allows the reconstruction of the uranium uptake history in each dental tissue using a specific U-uptake parameter (p-value) (Grün et al., 1988), but cannot account for uranium loss. A new model, called accelerating uptake model, AU-ESR, allows an extension to samples exhibiting a slight uranium leaching (Shao et al., 2012). An improved version, called CAM, Combined Age

model, allows calculation of ages for teeth constituted by tissues which display uranium leaching while other tissues in the same tooth have experienced uptake (Shao, personal communication).

Sample preparation and measurements

16 herbivorous teeth, all taken in the Middle Complex from levels F, G (Ensemble III), J (Ensemble II), and L, P/Q (Ensemble I), were selected and prepared in the Paris MNHN laboratory for analyses (Figure 1). Teeth coming from level G were already published (Han et al., 2010) and here the ages are calculated using CAM model when it was necessary. Enamel, dentine and cement were separated mechanically and their radioisotope contents were measured by ICPQMS (LSCE, Gif-sur-Yvette, France). A part of the enamel, after cleaning of its surface on both sides (inner and outer sides) to eliminate the effect of external alpha radiation, was ground, sieved and the 100-200 μ m fraction split into 10 aliquots. Nine of the ten were irradiated with a calibrated ⁶⁰Co gamma-ray source from 200 to 8,000 Gy.

ESR measurements were performed at room temperature on an EMX-6 Bruker spectrometer (X band, 9.82 GHz) with a microwave power of 10 mW and modulation amplitude of 0.1 mT. A scan range of 10 mT and a scan time of 4 minutes with a modulation frequency of 100 kHz were used for each spectrum. Each ESR measurement was repeated three times for each dose in order to minimize the anisotropic effect. The equivalent doses (D_E) were determined from the asymmetric enamel T1-B2 signal at g = 2.0018 (Grün et al., 2008) by fitting a single saturating exponential plus linear function which better describes the experimental data points (Duval et al., 2009) (see supplement data). ESR age calculations were carried out with the ESR-DATA program of Grün, (2009b) which uses an alpha efficiency of 0.13 \pm 0.02 (Grün and Katzenberger-Apel, 1994) and Monte-Carlo beta attenuation factors (Marsh, 1999) based on the thickness of the tooth enamel and outer layers removed. In some cases, ages were calculated using the CAM program. In addition, the following parameters were used:

- The water content was estimated to be 3 ± 1 wt% in the enamel, 7 ± 3 wt% in the dentine and in the cement, and 15 ± 5 wt% in the sediment (an assumed value).
- Gamma-ray spectrometry was used to determine the sediment radioisotopes U, Th and K in where the teeth were collected. The dose rate was calculated according to Adamiec and Aitken (1998). In addition, TL dosimeters were inserted in the layers and left *in situ* for nearly one year.

Results and discussion

Table 1 shows isotopic ratios, U content and other data necessary to the age calculation. U content in enamel varies from 0.1 to 1 ppm with a sample at 3 ppm, and from 3 to 30 ppm in dentine and cementum.

 D_E values range between 210 and 330 Gy for F and G levels and increase from J to P levels (430-500 Gy) (see supplementary information).

Three dosimeters were inserted in G level in which teeth were sampled in carbonated, phosphated and non-altered areas (one dosimeter in each area) providing a 350 to 660 microGy/a dose rate. For non altered (NA) samples, the external dose represents 80-90% of the total dose. In the phosphated (P) samples, the external dose is ranging

between 60 and 85 %. For the carbonated (C) teeth, the external dose is 70-80 % of the total dose. This point is emphasized by the low uranium content in teeth which renders the external dose extremely dominant in the total dose and in the age calculation. In order to minimize this dependence, a mean value of the dose rate recorded in three dosimeters (436 μ Gy/a) has been used for age calculation (Table 2).

A mean age of 438 ± 31 ka for G level is in agreement with the age obtained for F level (392 ± 43 ka). The range for the G level 407-469ka corresponds to the MIS12 (424-478 ka).

In J and L levels, results are obtained using US model and p-values suggest a post-burial uranium uptake leading to small internal dose rates. These levels have the highest U, Th, K contents observed in the whole sequence and the external dose in these two levels represents about 70-80% of the total dose. The increase of clay and loamy sands in J level probably contributed strongly to retain water and also a lot of organic elements and phosphates rich in uranium coming from upper layers. A similar phenomenon was found in the lower archaeological levels of Ambrona, Spain, where the analyzed horse teeth yielded Holocene U-series age while their D_E and archaeological data suggested a much older age (see Falguères et al., 2007).

The teeth coming from the P level have undergone a complex geochemical history with ²³⁰Th/²³⁴U greater than 1 in more than half of the tissues. ARA0101 and ARA0102 have thorium/uranium ratios far beyond unity and in this case, thus the models reach their limits. ARA0301 for which only one tissue exhibits an isotopic ratio slightly greater than 1 (cementum tissue) underwent less leaching than the two other samples. The obtained age is probably closer to the real age than the two other esimates coming from P level.

Figure 2 shows D_E, ages and a mean external dose per level. For G level, there is a clear correlation between the three data. Below the G level, in spite of an increase of D_E, an inverse correlation between ages which get younger and an external dose which increases. This is particularly true in the J and L levels for which the values are multiplied by a factor 2 or 3. This fact could be linked to an increase of the clay and loamy sands proportion due to "bioturbation" beginning with higher degree of humidity in these levels and contributing to retain water particularly rich in phosphate phasis and organic matter coming from upper human bearing occupation (level G). These intense fluid movements have probably contributed to enrich J and L sediments in uranium, thorium and potassium. On the other hand loams and clays have probably a different origin than sands which can explain this radioactive content variation.

P level presents an external dose more similar to that of upper F and G levels, in agreement with the similarity of their sedimentological characteristics.

The ESR/U-series ages range from 300 to 530 ka in these heterogeneous levels full of bones, teeth, lithic artefacts, limestones blocks, sands and clays rendering very difficult accurate geochronological measurements.

They are compared with faunal spectra and sedimentological data in order to obtain a better constrained frame (Figure 3).

The F and G levels contain cold faunas represented by the reindeer, the muskox, *Preovibos priscus* and rodents such as *Discrotonyx torquatus*. The wolf evolutive degree confirms an age older than 300 ka in agreement with the dates older than 350 ka obtained on the oldest part of the stalagmitic formation located at the top of Middle Complex

(Falguères et al., 2004). By contrast, palaeoenvironmental data suggest that J level has been deposited under humid conditions since occurrences of faunas such as *Dama* and *Cervus* live in a forest environment. The slight increase of rhinoceros remains also seems to confirm the increase of humidity in this level. From a chronostratigraphical point of view, the rhinoceros remains (*Stephanorhinus hemitoechus*) suggest an age between 600 and 400 ka (Guérin, 1981), in agreement with radiometric ages from P to G levels.

The P/Q levels provided also cold faunas such as *Lagopus*, *Ovis ammon antiqua* and *Rangifer tarandus* traducing a cold climate.

At the bottom of the considered sequence, the fauna contains several carnivores (*Canis lupus mosbachensis* and *Ursus deningeri*) associated with large ungulates (*Equus mosbachensis*, *Rangifer tarandus*, *Praeovibos priscus*) that seem to correspond to a transition between Cromerian faunas and those that arrived at the beginning of the Upper Pleistocene. The Arago cave sequence has also yielded one of the oldest occurrences of *Ursus arctos* in the Ensemble I, one Asiatic panther, big reindeer of the beginning of Pleistocene and a number of *Ovis ammon antiqua* in the Ensemble III. The abundance of *Praeovibos priscus* marks a dispersive phase through Europe during the MIS 12. The microfaunal fossils confirm the chronological position of archaeological levels of Arago cave, with particularly the spread of some taxa coming from northern areas earlier than expected (Moigne et al., 2006). The association of some taxa like *Ochotona pusillus*, *Citellus* sp. *Chionomys nivalis*, *Microtus gregalis* suggests the appearance of Central Asian and Siberian species, while *Dicrostonys torquatus* and *Microtus oeconomus* refer to Northern species (Hanquet and Desclaux, 2011). Micromammal (rodent) studies suggested correlation of levels H to D (Ensemble III and top of Ensemble II) to layer H of Orgnac III, primarily by the presence of *Microtus gregalis* (Chaline, 1981).

Conclusions

In the Middle Complex of the Arago cave, the exceptional faunal spectra associated with radiometric and sedimentological data, as shown in Figure 3, highlight the record of an alternation between dry-cold periods and humid-temperate phases. P, and F and G soils are marked by cold conditions with a major climatic pejoration in the lowest part of Middle Complex as suggested by the presence of primitive muskox and of reindeer which becomes the oldest occurrence for this species observed in Western Europe.

F and G levels can be assigned to MIS12 using the mean age which correlates with this period. Palaeontological and environmental data confirm a cold period. The F and G levels are also constrained by the presence of stalagmitic floor above them which provided age of more than 350 ka for the older calcitic layers. The G level is also coeval with a cold phase which could be attributed to MIS 12 though the larger biodiversity represented helps to conclude that G level was less dry and cold than the P/Q levels.

If we consider the alternation of cold and dry periods and humid and temperate periods, the P level, despite its large error range, could be contemporaneous with MIS 14 though this glacial period was less cold than MIS 12 or MIS 16 according to the data published in Lisiecki and Raymo, (2005). The J level, which contains more clay percentage,

corresponds to a more humid period as indicated by faunas living in forest environment, could be attributed to MIS 13. These data assignments remain speculative since the radiometric data are not conclusive.

Notwithstanding these caveats, our data suggest that Acheulian industry at Arago cave, and especially in P level, was probably present as soon as early as 500 ka. In the southern part of Western Europe, only Arago and Venosa Notarchirico, Italy, can yield human remains associated with Mode 2 industry at ca 600 ka (Lefèvre et al., 2010), with Isernia being considered as intermediate between Mode 1 and Mode 2 industries in Western Europe for the same period range (Galotti and Peretto, 2014).

In terms of human evolution, the mean age of 438 ± 31 ka for G level which yielded many human remains is concordant with that published recently by Arsuaga et al., (2014) for Sima de los Huesos human fossils. However, there is a clear difference between the fossils of both sites, Sima fossils showing more Neandertal features than Arago fossils. The contemporaneity of Arago and SH fossils raises the issue of whether one or two human species existed around 400-450 ka in Western Europe.

Acknowledgements

The ESR dating studies at Caune de l'Arago were initiated in 1978 by Y. Yokoyama and represent the first work in this way in France. We are grateful to him. This work is a collective task and we would like to thank all participants in Arago excavations and field studies. We thank warmly the reviewer who contributed to improve this paper.

Figure captions

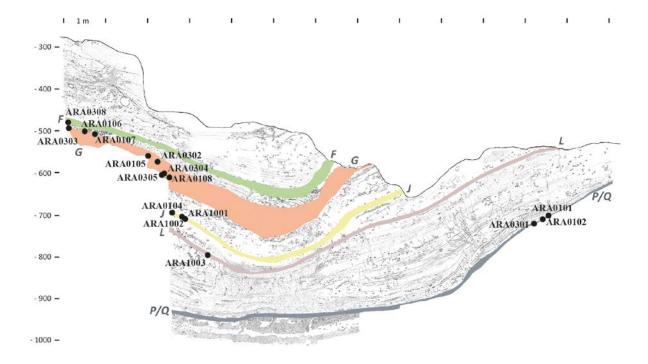


Figure 1: Stratigraphical sequence of the Arago cave showing the human bearing occupation levels of Ensemble II of Middle Complex. All the analyzed samples dated by ESR/U-series combined method are located.

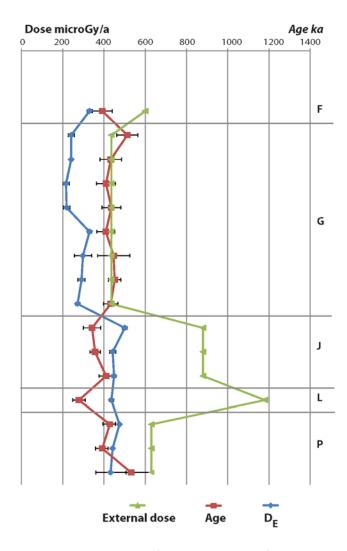


Figure 2: Evolution of external doses (green triangles), DE (blue lozenges) and ages (red squares) along the stratigraphical sequence of Middle Complex.

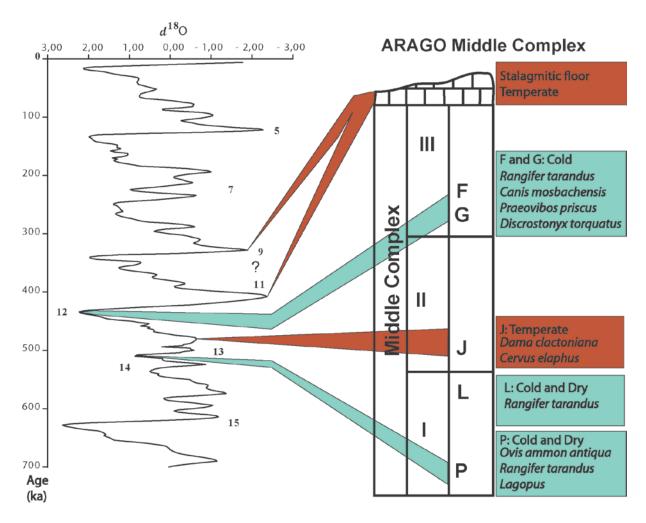


Figure 3: Middle Complex Arago sequence versus MIS curve according Lisiecki and Raymo data (2005).

Sample	species	Tissue	U (ppm)	²³⁴ U/ ²³⁸ U	²³⁰ Th/ ²³⁴ U	²²² Rn/ ²³⁰ Th	T enamel (μm)	removed enamel* (μm)		
F level	0,000.00	110000	(PP)	<u> </u>	1111	,	(μ)	σταττοι (μ.τ.)		
1 10101			29.8	0.978 ± 0.02	0.590 ±	1.00				
ARA0308	Equus molar	D			0.03 0.892 ±	1.00				
		Е	0.46	1.030 ± 0.05	0.06	0.50	1460	50/190		
		С	32.45	0.856 ± 0.03	0.604 ± 0.03	0.60				
G level										
	Equus molar		2.97	1.139 ± 0.05	0.691 ±	0.40				
		D	0.10	1.309 ± 0.15	0.03 0.754 ±		1400	30/70		
ARA0108		Е	0.10	1.509 ± 0.15	0.12 0.691 ±	0.40	1400	30/70		
		С	2.97	1.139 ± 0.05	0.03	0.40				
		D	4.76	1.133 ± 0.04	0.848 ± 0.04	0.30				
ARA0304	Rhino	E	0.05	1.56 ± 0.40	0.329 ± 0.11	0.30	2050	40/120		
711010004	TTIIIIO		26.02	0.898 ± 0.02	0.265 ±	0.30				
		D			0.01 0.259 ±		2540	50/120		
ARA0305	Rhino	E	0.15	0.736 ± 0.10	0.05 0.866 ±	0.40	2540	50/120		
		D	6.90	1.216 ± 0.06	0.05	0.60				
ARA0303	Equus molar	Е	0.42	1.806 ± 0.28	0.435 ± 0.08	0.20	1490	60/100		
		С	2.93	1.08 ± 0.05	0.804 ± 0.05	0.60				
			12.67	1.119 ± 0.04	0.964 ±	0.50				
	Equus molar	D			0.05 1.190 ±	0.00	4400	50/400		
ARA0107		Е	0.19	0.791 ± 0.18	0.30 0.964 ±	0.40	1160	50/120		
		С	12.67	1.119 ± 0.04	0.05	0.50				
ARA0106	Equus molar	D	11.39	1.220 ± 0.03	0.995 ± 0.04	0.60				
			0.49	1.232 ± 0.07	1.059 ± 0.08	0.70	1480	80/130		
		E	1.81	1.257 ± 0.06	$0.783 \pm$	0.70 0.70				
		С			0.04 0.899 ±					
	Ovis molar	D	4.80	1.209 ± 0.04	0.09	0.20				
ARA0302		E	0.08	1.25 ± 0.42	0.923 ± 0.33	0.40	870	70/110		
		D	5.40	1.379 ± 0.03	1.015 ± 0.03	0.30				
A D A 04 0 F	Ovis		0.19	1.304 ± 0.14	0.35 ± 0.06	4.00	1110	20/180		
ARA0105	molar	E				1.00				
J level			17.02	4 200 + 0.02	0.686 ±					
	Dama	D	17.83	1.288 ± 0.02	0.02					
ARA0104	molar	E	3.10	1.22 ± 0.02	0.66 ± 0.02		940	40/70		
ARA1001	Equus molar	D	21.31	1.365 ± 0.04	0.799 ± 0.05	0.35				
		E	0.68	1.435 ± 0.04	0.745 ± 0.16	0.83	1344	102/21		
					$0.796 \pm$		1044	104/41		
		С	20.52	1.449 ± 0.04	0.07 0.714 ±	0.58				
ARA1002	Equus molar	D	16.46	1.30 ± 0.05	0.07 0.685 ±	0.38				
		Е	0.62	1.410 ± 0.06	0.01	0.83	1330	30/76		
		С	16.91	1.386 ± 0.04	0.758 ± 0.04	0.38				
L level										
ARA1003		D	4.70	1 174 : 0.06	0.892 ±	0.44				
	Ovis molar	D	4.73	1.174 ± 0.06	0.07 0.922 ±	0.44				
		molar	molar E	E	0.30	1.210 ± 0.06	0.02 0.733 ±	0.33	893	37/42
		С	7.16	1.235 ± 0.02	0.04	0.59				

P level								
	_	D	14.4	1.574 ± 0.04	1.218 ± 0.04	0.40		
ARA0101	Equus molar	Е	1.08	1.584 ± 0.06	0.85 ± 0.04	0.60	1360	50/50
		С	11.75	1.637 ± 0.06	1.137 ± 0.05	0.30		
		D	9.14	1.577 ± 0.04	1.188 ± 0.04	0.30		
ARA0102	Bison molar	E	1.01	1.445 ± 0.06	0.859 ± 0.05	0.60	1020	30/40
		D	10.25	1.533 ± 0.04	0.932 ± 0.05	0.40		
ARA0301	Equus molar	E	0.55	1.536 ± 0.07	0.741 ± 0.05	0.30	1110	40/80
		С	6.35	1.621 ± 0.04	1.058 ± 0.03	0.40		

Table 1: ESR and U-series data on fossil herbivorous teeth from Arago cave, Middle Complex. E = enamel, D = dentine, C = cementum. Uncertainties for isotopic ratios are given with ± 1 sigma. The initial (T) and removed enamel thickness are used for the age calculation

^{*} the first number corresponds to the enamel subtracted in the dentine-enamel side; the second number to the cementum(sediment)-enamel side

Samples	(β+γ) sediment + cosmic	Internal dose rate (α+ß) enamel	ß dose rate (µGy/a)	Total dose rate (µGy/a)	DE (Gy)	U-uptake parameter p or <i>n</i>			ESR/U-series age (ka)
	(µGy/a)	(µGy/a)				Enamel	Dentine	Cementum	US or CAM
F level									
ARA0308	600 ± 60	93 ± 54	151 ± 52	844 ± 102	330 ± 17	-0.73 ± 0.06	1.04 ± 0.28	0.86 ± 0.26	392 ± 43
G level									
ARA0108	436 ± 52	15 ± 29	20 ± 20	471 ± 65	241 ± 11	0.34 ± 0.20	0.79 ± 0.27	0.79 ± 0.27	513 ± 67
ARA0304	436 ± 52	1 ± 6	120 ± 40	557 ± 67	241 ± 11	6.45 ± 0.89	-0.43 ± 0.09	NC	433 ± 48
ARA0305	436 ± 52	1 ± 10	90 ± 50	527 ± 75	216 ± 15	8.96 ± 1.33	8.66 ± 1.29	NC	410 ± 51
ARA0303	436 ± 52	17 ± 13	50 ± 28	503 ± 61	219 ± 6	3.67 ± 0.61	-0.47 ± 0.09	-0.24 ± 0.12	436 ± 52
ARA0107*	436 ± 52	117 ± 14	332 ± 87	885 ± 61	286 ± 15	-0.004 ± 0.001	-0.89 ± 0.05	-0.89 ± 0.05	396 ± 57
ARA0106*	436 ± 52	151 ± 49	81 ± 21	668 ± 75	298 ± 15	-0.003 ± 0.001	-0.89 ± 0.03	-0.08 ± 0.12	433 ± 43
ARA0302	436 ± 52	16 ± 18	192 ± 33 186 ±	644± 65	291 ± 10	-0.675± 0.05	-0.57 ± 0.06	NC	452 ± 43
ARA0105	436 ± 52	6 ± 27	130	628 ± 149	272 ± 42	5.73 ± 1.33	-0.87 ± 0.06	NC	433 ± 78
J level									
ARA0104	880 ± 80	300 ± 108	286 ± 28	1465 ± 138	501 ± 17	0.26 ± 0.15	0.12 ± 0.14	NC	342 ± 30
ARA1001	880 ± 90	140 ± 58	221 ± 54	1241 ± 125	442 ± 10	-0.09 ± 0.12	-0.34 ± 0.10	-0.31 ± 0.10	357 ± 35
ARA1002	880 ± 90	93 ± 52	125 ± 35	1098 ± 115	449 ± 11	0.46 ± 0.18	0.25 ± 0.16	0.01 ± 0.13	410 ± 42
L level									
ARA1003	1178 ± 120	65 ± 71	320 ± 51	1563 ± 150	436 ± 15	-0.91 ± 0.05	-0.85 ± 0.05	NC	279 ± 25
P level									
ARA0101*	630 ± 60	284 ± 61	198 ± 25	1112 ± 91	475 ± 10	-0.33 ± 0.07	-0.003 ± 0.001	-0.003 ± 0.001	428± 34
ARA0102*	630 ± 60	242 ± 62	256 ± 23 124 ±	1128 ± 90	442 ± 10	-0.47 ± 0.06	-0.004 ± 0.001	NC	392 ± 30
ARA0301*	630 ± 60	59 ± 113	124 ± 120	813 ± 212	432 ± 73	0.56 ± 0.35	-0.50 ± 0.11	-0.002 ± 0.001	532 ± 106

Table 2: Components of dose-rates for US model of teeth and sediment, D_E values and ESR-US age estimates with correspondent p-values for fossil teeth from Arago.

External dose-rates correspond to both sediment dose and cosmic dose (β + g). Two types of measurements have been performed.

About 100g of sediment including rock fragments when they are present, were measured at least one month after it has been inserted in a box.

Sediments have been sampled in the same square and at the same height as the teeth.

TL dosimeters have been placed in different levels at the exact location of the analyzed sediments.

For samples lacking cementum tissue, the enamel was directly in contact with the sediment. The case dentineenamel-cementum was used for some equid teeth, the cementum layer was thick enough to prevent the enamel from sediment beta particles.

NC means No Cementum.

^{*} ages are calculated using a CAM program.

References

Adamiec, G., Aitken, M.J., 1998. Dose-rate conversion factors: update. Ancient TL 16, 37-50.

Arsuaga J.L., Martinez I., Arnold L.J., Aranburu A., Gracia-Téllez A., Sharp W.D., Quam R.M., Falguères C., Pantonja-Pérez A., Bischoff J.L., E. Poza-Rey, Parés J.M., Carretero J.M., Demuro M., Lorenzo C., Sala N., Martinon-Torres M., N. Garcia, Alcazar de Velasco A., Cuenca-Bescos G., Gomez-Olivencia A., Moreno D., Pablos A., Shen C.C., Rodriguez L., Ortega A.I., Garcia R., Bonmati A., Bermudez de Castro J.M., Carbonell E., 2014. Neandertal roots: cranial and chronological evidence from Sima de los Huesos. Science, 344, 1358-1363.

Ashton, N., Lewis, S.G., De Groote, I., Duffy, S.M., Bates, M., Bates, R., Hoare, P., Lewis, M., Parfitt, S.A., Peglar, S., Williams, C., Stringer, C., 2014. Hominin footprints from Early Pleistocene deposits at Happisburgh, UK. Plos One, 9, 2, e88329.

Bahain, J.J., Falguères, C., Laurent, M., Voinchet, P., Dolo, J.M., Antoine, P., Tuffreau, A., 2007. ESR chronology of the Somme River Terrace system and first human settlements in Northern France. Quaternary Geochronology 2, 356-362.

Carbonell, E., Bermudez de Castro, J.M., Arsuaga, J.L., Allue, E., Bastir, M., Benito, A., Càceres, I., Canals, T., Diez, J.C., van der Made, J., Mosquera, M., Ollé, A., Pérez-Gonzalez, A., Rodriguez, J., Rodriguez, X.P., Rosas, A., Rosell, J., Sala, R., Vallverdu, J., Verges, J.M., 2005. An Early Pleistocene hominin mandible from Atapuerca-TD6, Spain. Proc Natl Acad Sci USA 102:5674–5678.

Carbonell, E., Bermudez De Castro, J.M., Arsuaga, J.L., Diez, J.C., Rosas, A., Cuenca-Bescos, G., Sala, R., Mosquera, M., Rodriguez, X.P., 1995. « Lower Pleistocene hominids and artifacts from Atapuerca-TD6 (Spain). *Science* 269, pp. 826-830.

Carbonell, E., Bermúdez de Castro, J.M., Parés, J.M., Pérez-González, A., Cuenca-Bescós, G., Ollé, A., Mosquera, M., Huguet, R., van der Made, J., Rosas, A., Sala, R., Vallverdú, J., García, N., Granger, D.E., Martinón-Torres, M., Rodríguez, X.P., Stock, G.M., Vergès, J.M., Allué, E., Burjachs, F., Cáceres, I., Canals, A., Benito, A., Díez, C., Lozano, M., Mateos, M., Navazo, M., Rodríguez, J., Rosell, J., Arsuaga, J.L., 2008. The first hominin of Europe. Nature 452, 465–470.

Chaline, J., 1981. Les rongeurs de la Caune de l'Arago à Tautavel et leur place dans la biostratigraphie européenne, in: H. de Lumley, J. Labeyrie (Eds), Absolute dating and Isotope Analysis in Prehistory - Methods and limits, Prétirage, CNRS, Paris, 1981, pp.193-203.

Duval, M., Grün, R., Falguères, C., Bahain, J.-J., Dolo, J.-M., 2009. ESR dating of Lower Pleistocene fossil teeth: Limits of the single saturating exponential (SSE) function for the equivalent dose determination. Radiation Measurements 44, 477-482.

Falguères, C., Yokoyama, Y., Shen, G.J., Bischoff, J.L., Ku, T.-L., Lumley, H. de, 2004. New U-series dates at the Caune de l'Arago, France. Journal of Archaeological Science, 31, 941-952.

Falguères C., Bahain J.J., Dolo J.M., Mercier N. & Valladas H. (2007). On the interest and the limits of using combined ESR/U-series model in the case of very late uranium uptake. Quaternary Geochronology, 2, 403-408.

Falguères, C., Bahain, J.J., Duval, M., Shao, Q., Han, F., Lebon, M., Mercier, N., Pérez-González, A., Dolo, J.M., Garcia, T., 2010. A 300-600 ka ESR/U-series chronology of Acheulian sites in Western Europe. Quaternary International 223-224, 293-298.

Galotti R., Peretto C., 2014. The Lower/early Middle Pleistocene small debitage productions in Western Europe: New data from Isernia La Pineta t.3c (Upper Volturno Basin, Italy), Quaternary International, XX, 1-18, in press.

Grün, R., Schwarcz, H.P., Chadam, J.M., 1988. ESR dating of tooth enamel: coupled correction for U-uptake and U-series disequilibrium. Nucl. Tracks Radiat Measur. 14, 237-241.

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

Grün, R., 2009a. The relevance of parametric U-uptake models in ESR age calculations, Radiation Measurements 44, 472-476.

Grün, R., 2009b. The DATA program for the calculation of ESR age estimates on tooth enamel. Quaternary Geochronology 4, 231-232.

Guérin, C., 1981. Les rhinoceros (Mammalia, Perissodactyla) du gisement pléistocène moyen de la caune de l'Arago à Tautavel. Signification stratigraphique, in: H. de Lumley, J. Labeyrie (Eds), Absolute dating and Isotope Analysis in Prehistory - Methods and limits, Prétirage, CNRS, Paris, pp. 163-191.

Han F., Falguères C., Bahain J.-J., Shao Q., Duval M., Lebon M., Garcia, T., Dolo J.-M, Perrenoud C., Shen G.J., Lumley H. de, 2010. Effect of deposit alterations on the dating of herbivorous teeth from Arago cave by the ESR-U series method, Quaternary Geochronology, 5, 376-380.

Hanquet, C., Desclaux, E., 2011. Analyse paléoécologique des communautés de micromammifères de la Caune de l'Arago (Tautavel, France) dans le contexte des migrations de faunes en Europe méridionale au cours du Pléistocène moyen. Quaternaire, 22(1), 35-45.

Lefèvre, J.C., Raynal, J.P., Vernet, G., Kieffer, G., Piperno, M., 2010. Tephro-stratigraphy and the age of ancient Southern Italian Acheulean settlements: The sites of Loreto and Notarchirico (venosa, Basilicate, Italy). Quaternary International, 223-224, 360-368.

Lisiecki, L.E., Raymo, M.E., 2005. A Pliocene-Pleistocene stack of 57 globally distributed benthic δ^{18} O records. Paleoceanography, 20, PA1003.

Lumley H. de, Labeyrie, J., 1981. Absolute dating and Isotope Analysis in Prehistory - Methods and limits, Prétirage, 720 pp, CNRS, Paris.

Lumley, H. de, Grégoire, S., Barsky, D., Batalla, G., Bailon, S., Belda, V., Briki, D., Byrne, L., Desclaux, E., El Guenouni, K., Fournier, A., Kacimi, S., Lacombat, F., Lumley, M.-A. de, Moigne, A.-M., Moutoussamy, J., Paunescu, C., Perrenoud, C., Pois, V., Quilès J., Rivals, F., Roger, T., Testu, A., 2004. Habitat et mode de vie des chasseurs paléolithiques de la Caune de l'Arago (600 000–400 000 ans). L'Anthropologie, 108, 159-184.

Lumley, H. de, Lumley, M.A. de, Merle des Iles, M.R., Moigne, A.M., Perrenoud, C., 2014. Historique des recherches sur lma Caune de l'Arago, Tautavel-en-Roussillon, Pyrénées orientales. In : Caune de l'Arago, Tautavel-en-Roussillon, Pyrénées orientales, France, Tome 1, directed by H. de Lumley, chapter 2, 35-171.

Marsh, R., 1999. Beta-gradient isochrons using electron paramagnetic resonance: towards a new dating method in archaeology. Ph.D. Dissertation, McMaster University, Hamilton.

Moigne, A.M., Palombo, M.R., Belda, V., Heriech-Briki, D., Kacimi, S., Lacombat, F., Lumley de, M.A., Moutoussamy, J., Rivals, F., Quilès, J., Testu, A., 2006. Les faunes de grands mammifères de la Caune de l'Arago (Tautavel) dans le cadre biochronologique des faunes du Pléistocène moyen italien. L'Anthropologie, 110, 788-831.

Moncel, M.H., Despriée, J., Voinchet, P., Tissoux, H., Moreno, D., Bahain, J.J., Courcimault, G., Falguères, C., 2013. Early evidence of Acheulean settlement in Northwestern Europe – La Noira Site, a 700 000 year-old occupation in the Center of France. PLOSone, 8, e75529.

Palombo, M.R., 2007. What is the boundary for the QUternary period and Pleistocene epoch? The contribution of turover patterns in large mammalian complexes from north-western Mediterranean to the debate. Quaternaire, 18, 1, 35-55.

Rivals, F., Deniaux, B., 2003. Dental microwear analysis for investigating the diret of an argali population (*Ovis ammon antiqua*) of mid-Pleistocene age, Caune de l'Arago, eastern Pyrenees, France. Palaeogeography, Palaeoclimatology, Palaeoecology, 193, 443-455.

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. Quaternary Geochronology, 10, 406-411.

Toro-Moyano, I., Martínez-Navarro, B., Agustí, J., Souday, C., Bermúdez de Castro, J.M., Martinón-Torres, M., Fajardo, B., Duval, M., Falguères, C., Oms, O., Parés, J.M., Anadón, P., Julià, R., García-Aguilar, J.M., Moigne, A.M., Patrocinio Espigares, M., Ros-Montoya, S., Palmqvist, P. (2013). The oldest human fossil in Europe dated to ca. 1.4 Ma at Orce (Spain). Journal of Human Evolution, 65, 1-9.

Yokoyama, Y., Nguyen, H.V., 1981. Comptes Rendus Académie des Sciences de Paris, 292, 741-744.

Yokoyama, Y., Falguères, C., Quaegebeur, J.P., 1985. ESR dating of quartz from quaternary sediments: first attempt. Nuclear Tracks 10, 921-928.

Yokoyama, Y., Quaegebeur, J.P., Bibron, R., Léger, C., Nguyen, H.V., Poupeau, G.,1982. Datation du site de l'Homme de Tautavel par la résonance de spin électronique (ESR), Comptes Rendus Académie des Sciences de Paris, 294, 759-764.

Wagner, G.A., Krbetschek, M., Degering, M., Bahain, J.J., Shao, Q., Falguères, C., Voinchet, P., Dolo, J.M., Garcia, T., Rightmire, G.P., 2010. Radiometric dating of the type-site for *Homo heidelbergensis* at Mauer, Germany. PNAS, 107, 19726-19730.