

## Dating Palaeolithic cave art: why U-Th is the way to go.

Alistair. W. G. Pike<sup>a</sup>, Dirk. L. Hoffmann<sup>b,c</sup>, Paul B. Pettitt<sup>d\*</sup>, Marcos García-Diez<sup>e</sup>, João Zilhão<sup>f</sup>

<sup>a</sup>Department of Archaeology, University of Southampton, Avenue Campus, Highfield Road, Southampton, SO17 1BF, UK. E-mail: [A.W.Pike@soton.ac.uk](mailto:A.W.Pike@soton.ac.uk)

<sup>b</sup>Max Planck Institute for Evolutionary Anthropology, Department of Human Evolution, Deutscher Platz 6, 04103 Leipzig, Germany, E-mail: [dirk.hoffmann@eva.mpg.de](mailto:dirk.hoffmann@eva.mpg.de)

<sup>c</sup>Bristol Isotope Group, School of Geographical Sciences, University of Bristol, University Road, Bristol, BS8 1SS, UK

<sup>d</sup>Department of Archaeology, Durham University, South Road, Durham, DH1 3LE, UK. E-mail: [paul.pettitt@durham.ac.uk](mailto:paul.pettitt@durham.ac.uk) (corresponding author)

<sup>e</sup>Department of Geography, Prehistory and Archaeology, University of the Basque Country (UPV/EHU), c/ Tomás y Valiente s/n, 01006 Vitoria, Spain. E-mail: [marcos.garcia@ehu.eus](mailto:marcos.garcia@ehu.eus)

<sup>f</sup>University of Barcelona/ICREA, Departament de Prehistòria, Història Antiga i Arqueologia (SERP), c/ Montalegre 6, 08001 Barcelona, Spain. E-mail: [joao.zilhao@ub.edu](mailto:joao.zilhao@ub.edu)

\*Corresponding author

### *Abstract*

The chronology of European Upper Palaeolithic cave art is poorly known. Three chronometric techniques are commonly applicable: AMS <sup>14</sup>C, TL and U-Th, and in recent years the efficacy of each has been the subject of considerable debate. We review here the use of the U-Th technique to date the formation of calcites which

can be shown to have stratigraphic relationships to cave art. We focus particularly on two recent critiques of the method. By using specific examples from our own work using this method in Spain we demonstrate how these critiques are highly flawed and hence misleading, and we argue that the U-Th dating of calcites is currently the most reliable of available chronometric techniques for dating cave art.

### *Keywords*

Uranium-Thorium; Chronology; Cave art; Paleolithic; Calcite.

### *Introduction*

Despite more than a century of discovery and research, the chronology of cave art is still poorly understood. Only three chronometric techniques have come to supplement relative schemes based on thematic and stylistic analysis: Radiocarbon ( $^{14}\text{C}$ ) using Accelerator Mass Spectrometry (AMS), TL (Thermoluminescence) and Uranium-Thorium (U-Th). For the former, the rationale relies upon the dating of small amounts of charcoal used to create art; assuming measurements are accurate and contamination is not an issue, this produces an age for the creation of the charcoal, which may or may not relate directly to its subsequent use as an art pigment. Because of this 'old charcoal' issue, many dates for cave art produced with  $^{14}\text{C}$  have been intensely debated, and some that were initially published even withdrawn (Pettitt and Bahn, 2003 *contra* Valladas and Clottes, 2003). The U-Th method, by contrast, produces ages for the formation of calcite speleothems; if it can be demonstrated that these have a clear stratigraphic relationship with the art of concern it can produce maximum ages (if the art is created upon it) or minimum ages (if it overlies – i.e. has formed on top of – the art). In theory, the TL method can produce similar information, but its usefulness is hindered by the size of the associated uncertainty interval (i.e. its error), as typical standard deviations are about 10% of the mean age.

In 2012 we published U-Th dates on calcites associated with cave art in a number of caves in Northern Spain, including Altamira, El Castillo and Tito Bustillo (Pike et al., 2012). Among our conclusions we were able to demonstrate that some examples of non-figurative art – a red disk and a red hand stencil in El Castillo – were older than 37.3 ka and 40.8 ka respectively, showing that some cave art is at least Early Upper Palaeolithic in age, and sufficiently close to the time of arrival of the first modern humans and the disappearance of Neanderthals to justify the construction of a testable hypothesis regarding authorship. Since our publication, a few archaeologists and one dating specialist have published critiques of the U-Th method (Clottes, 2012; Pons-Branchu et al., 2014; Sauvet et al., 2015), arguing that U-Th dates on calcite associated with cave art – specifically our own – are unreliable:

1. Because of the open system behaviour of calcite, and because we did not obtain corroborating dates from alternative dating methods, especially  $^{14}\text{C}$ .
2. Because of a potentially incorrect initial (detrital)  $^{230}\text{Th}$  correction that could seriously affect the accuracy of the U-Th results.

We also came under criticism from these authors because of our reliance on minimum ages, our sampling methodology, and the chronological hypotheses we are testing. Although they survived the refereeing processes common to respectable international journals, as scientific debate we thought these authors' criticisms were unimpressive, and often highly misleading. We present here a robust response to what we perceive as a number of basic mistakes promulgated in these papers. Given that proponents of the  $^{14}\text{C}$  method for dating cave art have hardly ever responded scientifically to the numerous critiques of this method's applications (the many references are summarised in Pettitt and Bahn, 2015), we argue that the method we employ is the most reliable that we have at present for establishing the chronological development of cave art in Europe. We do so by dissecting each point made by the critics.

*U-Th dating: open system issues and corroborative dating*

All chronometric dating methods are limited by their accuracy (how close their age estimates come to the real age of a target sample, reflecting numerous issues that may affect the final result) and precision (the resulting uncertainty or age range of the measurement, i.e. its error). The main assertion of Pons-Branchu et al. (2014) and Sauvet et al. (2015) is that, in the absence of independent 'verification' or 'confirmation' (in their terminology) of U-Th dates by other methods, or a detailed consideration of the U concentrations and  $^{234}\text{U}/^{238}\text{U}$  ratios, one cannot rule out the possibility that our samples are affected by open system behaviour. Open system behaviour entails the loss or gain of U or Th from the calcite subsequent to its formation, thereby affecting the  $^{230}\text{Th}/^{234}\text{U}$  to produce erroneously younger or – more usually older – dates. It is obvious how such inaccuracies – if true – would seriously affect our understanding of the chronology of cave art if they were perpetuated.

All geochemists acknowledge that open system behaviour can exist in calcite; it is obvious to us, and in fact the scientific understanding of calcite behaviour is a specific research expertise of one of us (Hoffmann et al., 2009; Hoffmann et al., 2010; Fensterer et al., 2010; Gutjahr et al., 2013; Scholz et al., 2014). But every geochemist, however, would acknowledge that open-system behaviour is the *exception* rather than the rule. At the outset, then, the few examples highlighted by Pons-Branchu et al. (2014) and Sauvet et al. (2015) should therefore be judged against the many *thousands* of U-Th dates that have been published and which are not considered to be in any way problematic by the world's geochemistry community. To present only the very few exceptional cases introduces an unnecessarily misleading bias into the debate.

But let us focus on the theoretical issue of open-system inaccuracy. Pons-Branchu et al. (2014) suggest that leaching of U from calcites would be detectable from our samples if we had published our U concentrations (we publish them here). They describe how the alpha-recoil of  $^{234}\text{U}$  (i.e. the energy imparted to the calcite lattice when  $^{238}\text{U}$  decays) can lead to damage of the calcite crystal lattice and thus to the preferential leaching of  $^{234}\text{U}$  over  $^{238}\text{U}$ , and suggest that open system behaviour can be identified from anomalous  $^{234}\text{U}/^{238}\text{U}$  ratios. This is certainly an observable effect

for samples of *geological age* (i.e. many millions of years old), but it is geochemically naive to believe that such an effect – if it exists – will be at all significant over the Upper Pleistocene timescales we are dealing with. Such preferential leaching can only occur after the calcite is formed, and only at lattice sites where  $^{238}\text{U}$  has decayed.  $^{238}\text{U}$  has a half-life of 4.5 billion years; thus only a tiny percentage of  $^{234}\text{U}$  within calcite that is a few tens of thousands of years old will derive from the decay of  $^{238}\text{U}$ . The rest of the  $^{234}\text{U}$  will have been incorporated from the drip-water along with  $^{238}\text{U}$ . As an example, in a sample in which the initial  $^{234}\text{U}/^{238}\text{U}$  is 1.119 (i.e. sample O-83 of Pike et al. 2012), only 0.0006% of the  $^{238}\text{U}$  will decay over 41.4 ka. If that percentage of the  $^{234}\text{U}$  were leached from the sample (it is a maximum, because some of the  $^{234}\text{U}$  generated from  $^{238}\text{U}$  will decay to  $^{230}\text{Th}$  and not all alpha recoil sites will be vulnerable to leaching) it would be too small to be detected from differences in  $^{234}\text{U}/^{238}\text{U}$  to unleached samples. Furthermore, and more importantly, removing this amount of U from the system would actually have a *negligible* effect on the resulting U-Th date (i.e. less than one year!). By arguing that we have not used the  $^{234}\text{U}/^{238}\text{U}$  to rule out open system behaviour, Pons-Branchu et al. (2014) conjure mountains out of non-existent molehills in an apparent attempt to discredit a widely used and accepted dating technique.

With the exception of the examples given by Sauvet et al. (2015) where  $^{230}\text{Th}/^{234}\text{U}$  is larger than the theoretical equilibrium value (i.e. 1 when  $^{234}\text{U}/^{238}\text{U}=1$ ) – which is the case for *none* of our samples – open system behaviour cannot be identified *a priori*. It is usually identified when dates fall out of perceived stratigraphic order, at which point a *post hoc* explanation of open-system behaviour is often cited. For example, U concentration is used to explain the observed open system behaviour, but it *cannot* be used to predict it. U concentrations can vary greater than 100% within a few millimetres in coeval calcite layers (e.g. Hoffmann et al., 2009), and in El Castillo cave the U concentrations of our samples vary from 84 to 2000 ng/g (Table 1), with no correlation between U concentrations and the age of each sample. Thus there is no *a priori* evidence for open-system behaviour available from uranium concentrations. The assumption by Pons-Branchu et al. (2014) and Sauvet et al. (2015) that it is likely that our dates are affected by open-system behaviour appears to be based not on

inconsistencies in our data (given that they present no evidence that our data are problematic), but on a *post hoc* dislike of the dates we have produced, and they use an unrepresentative selection of the published literature to attempt to discredit U-Th dating in order to gain credibility for their stance. By being highly selective and citing rare examples of open-system behaviour in cave calcite, Pons-Branchu et al. (2014) could unfairly undermine a dating method that has a long and important history in understanding earth systems science.

It is, of course, the dating of calcites pertinent to cave art that is of concern here. Despite this, however, many of the examples cited by Pons-Branchu et al. (2014) and Sauvet et al. (2015) do not actually derive from caves; instead they derive from shallow rockshelters, which are very different systems to the deep caves we have sampled, or in the case of the Borneo cave, the problematic date comes from a sample noted by the authors as being macroscopically porous calcite (Plagnes et al., 2003) – which in all cases we ourselves would avoid. In any case, the test for open system behaviour used in these examples, i.e. a comparison of U-Th to  $^{14}\text{C}$  dates, is problematic, as we discuss below.

The standard test for closed system behavior in cave sciences is the demonstration that stratigraphically related samples result in stratigraphically ordered U-Th dates, i.e. trending from older to younger along a stratified sequence, or yield indistinguishable ages within uncertainties. In order to examine this we have, wherever possible, taken multiple samples along (through) the growth axis of the calcite. At the time of publication of Pike et al. (2012, Fig. S1), available opportunities to do so were somewhat limited, although those we had obtained showed no anomalies. Subsequently, we have, however, amassed a corpus of stratigraphically ordered samples which show that open system behavior is *very rare*. These will be published shortly, when our sampling programme is complete. Nevertheless, this appears to be insufficient for Sauvet et al. (2015); while they have confidence when the technique is applied to thick calcites (because the inner layers of these formations are unlikely to have been affected by leaching), they argue that thin, ‘unstratified’ calcites without a known growth axis – which they assume characterize those we have sampled – are more problematic. Given that this is the standard

method for checking for closed system behavior in earth science with either thick or thin calcites, these non-specialists appear, therefore, to cast doubt on *all* U-Th dates on cave calcites produced for whatever purpose.

Sauvet et al. (2015) and Pons-Branchu et al. (2014) advocate the use of  $^{14}\text{C}$  dating of speleothem calcite as an independent 'control' for U-Th dating of the same material. This is not a test that geochemists would use for a number of reasons. The  $^{14}\text{C}$  dating of calcite suffers from many more uncertainties than U-Th, and has been shown to be affected by a suite of problems including: considerable inaccuracies caused by many sources of contamination (Genty and Massault, 1999, Fohlmeister et al., 2011; Genty et al., 2011); the inclusion of a variable dead carbon fraction; and open-system behaviour (Holmgren et al., 1994; Pazdur et al., 1995). In stark contrast, contamination of U-Th samples can be *identified* using presence of common Th ( $^{232}\text{Th}$ ). Given the many more and often unidentifiable sources of inaccuracy in the  $^{14}\text{C}$  dating of calcite, why on earth would one choose a  $^{14}\text{C}$  date as a 'control' for a U-Th date? Remarkably, this is, however, the stance that Sauvet et al. (2015) and Pons-Branchu (2014) take, in stark contrast to the geochemistry community practice where the situation is reversed and U-Th dates are actually taken as controls for  $^{14}\text{C}$  dates in order to understand the many problems with  $^{14}\text{C}$  dating of calcite (Hoffmann et al., 2010; Genty et al., 2011; Griffith et al., 2012; Southon et al., 2012). Correcting  $^{14}\text{C}$  results via the U-Th dating of the same samples is in fact how radiocarbon calibration operates beyond the reach (~12,000 years) of tree-ring chronologies (Hoffmann et al., 2010). To follow the recommendations of Sauvet et al. and Pons Branchu et al. would be a reversal of established geochemical practice without grounds to do so.

The assumption that the maximum offset caused by the dead carbon fraction between a closed system  $^{14}\text{C}$  date and a closed-system U-Th date would be around 1900 years (i.e. around 20% dead carbon fraction) – used in the examples given by Pons-Branchu et al. (2014) and Sauvet et al. (2015) – simply does not match with scientific observations. The dead carbon fraction can be highly variable in karst systems both within and between speleothems. In calcite samples from France, Belgium and Scotland, Genty et al. (2001) demonstrated such variability in the dead

carbon fraction to lie between 5 and 37%, far greater than the 5-20% cited by Sauvet et al. (2015) and Pons-Branchu et al. (2014). In any case, where the dead carbon fraction is shown to be consistent between samples, it is U-Th dating that provides this evidence in the form of control dates. It is on this basis, therefore, that well-known attempts have been made, to calibrate radiocarbon using U-Th dating of calcite (Hoffmann et al., 2010; Reimer et al., 2013).

The problems with  $^{14}\text{C}$  do not end there. Even if we assume  $^{14}\text{C}$  dating to be unproblematic for calcite, comparing  $^{14}\text{C}$  dates with U-Th dates on the same sample does not compare like with like. The U-Th date of a calcite sample comprises an average of the dates of each layer of that sample weighted for U concentration, whereas a  $^{14}\text{C}$  date comprises the carbon-weighted average of the age of the layers. Since the U concentration can vary by several 100s of percent between layers of calcite in a single speleothem (Hoffmann et al., 2010; calcites used in this study had U concentrations between 64 und 350 ng/g) it would not be surprising to find discrepancies between  $^{14}\text{C}$  and U-Th dates even in the absence of any apparent problems with either technique (see Figure 1).

Figure 1 schematizes the most common scenario in the sampling and dating of calcite overlying art — one where the calcite accretion, even though thin, may well be multi-layered and cannot be assumed to have formed over a single, short period of time. In such a scenario, and assuming that no contamination or open system issues affect the ages obtained, the two distinct results returned by radiocarbon and uranium series would both be correct minimum ages for the underlying art. That radiocarbon returned a younger age cannot be used to make inferences on the reliability of the older uranium series age and is in fact entirely to be expected because of the way in which the average age of a mixed sample is impacted by the different radioactive decay processes at work in each method.

In the case of U-Th, we measure the accumulation in the sample of a daughter isotope ( $^{230}\text{Th}$ ) as a function of the decay of both the parent isotope ( $^{234}\text{U}$ ) and of that daughter isotope into other members of the series. In the case of  $^{14}\text{C}$  we measure the loss of a radioactive isotope over time. Therefore the oldest layer contributes the least  $^{14}\text{C}$  to the mixed sample, but the most  $^{230}\text{Th}$ , which results in a



difference in the dates calculated using the different dating methods. In Figure 1 (Scenario A) where the thickness of the two layers is equal, and the uranium concentration is homogenous, the  $^{14}\text{C}$  date of the mixed sample is 15.4 ka BP (18.7 cal ka BP) compared to a U-Th date of 24 ka BP. This difference will vary with the respective age, thickness and U concentration of the two layers, so in a second scenario (B) we calculate ages where the oldest layer has three times the U concentration of the youngest. While the  $^{14}\text{C}$  date remains the same (18.7 cal ka BP) the U-Th date is now 31.7 ka. These are both accurate minimum age estimates for the art yet they diverge by 13 ka. Therefore the discordance between  $^{14}\text{C}$  and U-Th dates in mixed samples is not a good indication of open system behaviour, yet this is the evidence that Sauvet et al. (2015) and Pons-Branchu et al. (2014) rely on to argue that the U-Th dating of calcite in association with art is not reliable.

From the above, it should be clear why radiocarbon dating of such samples cannot be used to 'control' the U-Th ages we obtained, and why the only possible control for their reliability is the geochemistry sciences standard: that the ages get older as one samples through the calcite towards the pigment. Cross-dating by  $^{14}\text{C}$  and U-Th of a short-lived coral or an individual calcite layer from a stalagmite makes sense for the calibration of  $^{14}\text{C}$  (but not the other way around). But such cross-dating would be methodologically absurd when the chronological homogeneity of the sample is unknown and cannot be assumed, as is the case with most, if not all of our samples and those examples used to argue against U-Th dating by Pons-Branchu et al. (2014) and Sauvet et al. (2015).

So much for  $^{14}\text{C}$ . Pons-Branchu et al. (2014) and Sauvet et al. (2015) suggest other methods such as TL or protactinium dating could also be used as 'controls' for U-Th. While protactinium ( $^{231}\text{Pa}$ ) dating has been successful on high U and large  $\text{CaCO}_3$  samples such as corals, and *in theory* can provide a robust test for closed system behaviour of speleothem carbonates, it is naïve to suggest that it could be used successfully in this context. While it has been shown that sub-fg (femtogram) quantities of  $^{231}\text{Pa}$  can be measured (Shen et al., 2003), the precision of such measurements at the level of  $^{231}\text{Pa}$  concentrations typical of our samples would limit their use as a check for closed system behaviour. For the four oldest samples in Pike

et al. (2012) the weights were between 21 and 80 mg, and U concentrations 270-1300 ng/g. This would yield between 4 and 8 fg of  $^{231}\text{Pa}$ .

Cheng et al.'s (1998) model predicts the effect on  $^{230}\text{Th}/^{238}\text{U}$  and  $^{231}\text{Pa}/^{235}\text{U}$  of a single U loss or gain event at a time,  $T_d$  years ago. Simplifying this model by assuming  $^{234}\text{U}/^{238}\text{U}=1$ , we calculated the % difference,  $\Delta R$ , in the observed  $^{231}\text{Pa}/^{235}\text{U}$ , between the ratio predicted if a 41 ka old sample had remained a closed system and a sample of true age 33 ka that has undergone U loss. Figure 2 shows that a sample dating to around 33 ka (i.e. at the Gravettian-Aurignacian boundary) that yields an apparent U-Th date of 41 ka due to a single U leaching event will give a maximum difference in  $^{231}\text{Pa}/^{235}\text{U}$  from a closed system 41 ka value of 3.8% for leaching in the very recent past, and less than this if leaching occurs earlier. Thus, we would require a precision on our  $^{231}\text{Pa}/^{235}\text{U}$  measurement very much better than 4% (almost certainly as low as 1%) to differentiate a closed system Aurignacian (41 ka) date from an open system date of 33 ka (i.e. the Gravettian-Aurignacian boundary – an age that would significantly alter the conclusions of Pike et al. 2012). We are very doubtful this could be achieved with the 3-5% uncertainty quoted for 2 fg  $^{231}\text{Pa}$  samples from seawater given by Shen et al. (2003). A more recent study on U-Pa analyses (on volcanic rocks which are probably a better proxy for calcite than seawater) by Koorneef et al. (2010) using MC-ICPMS shows a reproducibility of  $^{231}\text{Pa}/^{235}\text{U}$  on 200 fg  $^{231}\text{Pa}$  samples in the range of 3-4 %. Thus, the required high precision  $^{231}\text{Pa}/^{235}\text{U}$  analyses are simply not possible. Furthermore, our Figure 2 model predictions are also the best case scenario, as we have not accounted for uncertainties in  $^{230}\text{Th}/^{238}\text{U}$ , or  $^{234}\text{U}/^{238}\text{U}$ , and, perhaps more importantly, in contrast to the dating of corals and the determination of  $^{231}\text{Pa}$  concentrations in seawater, there will be the much greater additional uncertainty of an uncharacterized initial  $^{231}\text{Pa}$  in 'dirty' samples deriving from caves. Even if precision is improved, this latter issue is likely to prevent  $^{231}\text{Pa}$  dating ever being a useful tool for testing for open system behaviour in small calcite samples.

Other dating methods based on charge accumulation – e.g. ESR and TL – suffer from problems of reconstructing annual dose rates in the dynamic environment of a growing speleothem. Since much of the internal dose derives from the U in the

calcite, they *cannot* be a good control for U-Th, as disequilibrium between U and Th (i.e. relating to the U-Th date) must be accounted for in the calculation of dose rate. This is simply why the geochemistry community have almost exclusively used U-Th to date calcite, despite TL being available for nearly a half century. Furthermore, in the example given by Sauvet et al. (2015) – La Garma cave, Cantabria (González Sainz, 2003) – the TL sample and the U-Th samples derive from different sampling locations and are therefore not inter-comparable, and none of them are stratified above the cave art. Thus the ‘gap’ between the dates that Sauvet et al. (2015) discuss simply cannot be interpreted in the context of the reliability of the methods, nor the age of the art. Elsewhere in the same cave (Arias and Ontañón, 2008) TL samples obtained on a similar calcite formation (i.e. one that is near but not on top of art) gave a date of  $64.2 \pm 7.1$  ka (MAD-2075), which should surely indicate to Sauvet et al. (2015) that there is something wrong with the method (i.e. these TL samples are not stratigraphically related to the art).

#### *Correction for initial $^{230}\text{Th}$ (detritus)*

Another potential problem identified by both Pons-Branchu et al. (2014) and Sauvet et al. (2015) is that of correction for initial  $^{230}\text{Th}$ . Detritus (e.g. fine particulates) can be incorporated into precipitating (forming) calcite, bringing with them  $^{230}\text{Th}$  and thus rendering apparent U-Th ages too old. We correct for this using the measured  $^{232}\text{Th}$  and an assumed detrital fraction with an activity ratio  $^{230}\text{Th} / ^{232}\text{Th} = 0.8 \pm 0.4$  (note that Pons-Branchu et al., 2014 incorrectly state that the value we used was  $1.25 \pm 0.625$ ). Our correction method is the standard practice in U-Th dating when it is not possible to determine the exact  $^{230}\text{Th}/^{232}\text{Th}$  value (Richards and Dorale, 2003), and has the very conservative uncertainty fully propagated to the final date. For one cave site (Tito Bustillo), we were able to demonstrate that the isotopic signature of detrital contamination falls exactly into the range we use for correction (sample O-21), and the cave setting is representative of our work in Northern Spain. However, we accept that the true value of detrital  $^{230}\text{Th}/^{232}\text{Th}$  *may* lie outside this range, where it is not possible to do a direct measurement on insoluble residuals or apply

isochrons, but the example given by Pons-Branchu et al. (2014) seems simply biased towards their desire for our published dates to be younger. They recalculate our oldest date (O-83) using a detrital  $^{230}\text{Th}/^{232}\text{Th}$  of  $2.5\pm 0.5$  – larger than our range – which yields a corrected date of  $39.3\pm 0.7$  ka. But they fail to illustrate what would happen to the dates if a detrital correction smaller than ours were used. Let us show them: this would yield a corrected date older than our corrected date of  $41.4\pm 0.6$  ka and would tend towards our uncorrected date of  $42.4\pm 0.3$  ka as the detrital  $^{230}\text{Th}/^{232}\text{Th}$  tended towards zero.

Nevertheless, given that we are dealing with minimum ages, a date of  $>38.6$  ka (i.e. the minimum age calculated using their arbitrary detrital correction) would not alter our conclusions. A minimum age of  $>42.1$  ka might.

#### *U-Th dating and sampling issues.*

As with  $^{14}\text{C}$ , U-Th dating requires the removal of physical samples from the cave wall, and it goes without saying that the nature of such sampling will always be of concern. It is something that we take very seriously indeed. We are therefore particularly dismayed by the exaggerated assertion by Sauvet et al. (2015) that ‘the *significant damage* caused by sampling, conducted by scraping with a scalpel or drilling with a carbide drill bit (Pike et al., 2012) is a matter of grave concern’ (our emphasis). As with any act of archaeological destruction – be it excavation, radiocarbon, or indeed U-Th dating – sampling needs to be carefully considered, carefully undertaken, carefully witnessed, and carefully recorded and published for posterity. The concern to preserve and protect the legacy of antiquity is of course paramount, and, needless to say, justifies the considerable bureaucracy and consultation that accompanies *every* single act of sampling. In *every case* our own sampling – remember that it is removing naturally accumulated speleothems, not art – has been undertaken under the supervision of independent specialists (usually the representatives of the caves themselves, governmental institutions responsible for their protection, and/or the archaeologists responsible for the curation or study of the site). We have meticulously documented our sampling, and the final nature of

our 'destruction'. Since beginning our sampling programme we have successfully applied to revisit and re-sample several caves that we have previously sampled; surely our applications to do so would have been refused if the authorities felt we were causing in any way 'significant' damage to the cave or its art.

Sauvet et al. (2015) unfairly imply that our 'significant' damage is out of proportion to the usefulness of the dates we have obtained. They have included one of our sampling locations that was photographed without the inclusion of a scale. Without themselves noting the specific dimensions of our sampling (in their figure 5), they imply that our scalpels and drills are irresponsibly running rampant through the caves of Western Europe. This could not be further from the truth. Our typical sample sizes are less than 10 mm in maximum dimension, and we can work with sample masses of less than 10 mg. Figure 3A shows the example Sauvet et al. use, but in this case with the addition of a scale. The sample here is about 2cm across, but it is also the largest sample we have ever taken and therefore unrepresentative of the bulk of our previous work. Methodological developments in the seven years since this sample was removed mean we have reduced our sample mass requirement to eight times smaller than the example Sauvet et al. give. (e.g. Figure 3, B+C). We carefully remove layers of calcite stratified above the pigment of concern, and we stop sampling before the pigment layer is reached. In any case, samples containing pigment would be rendered useless as they would thereby be contaminated by the high detrital component of the pigments themselves. Thus we *never* remove from the caves *any* part of the art itself. This sampling strategy stands in stark contrast to attempts made by Sauvet's colleagues to apply radiocarbon dating to cave art charcoal, which removes samples from the art itself. In some cases this has comprised *numerous* and *repeated* sampling and re-sampling of charcoals (e.g. in the case of the art of Peña de Candamo – Corchón et al. 2014, 2015) and has even resulted in the publication of problematic dates which have subsequently been withdrawn pending 'future verification' (Valladas et al., 2005, pp. 111; note this was a decade ago and we are still to see this 'verification'). It surely begs the question of whether such truly destructive sampling is justified. By contrast, the effect of our sampling is to leave a small patch of calcite that is cleaner (brighter) than the

surrounding calcite, and through which the underlying pigment is visible, which will no-doubt naturally accumulate surface dirt and return to a more typical calcite colour in time. If our samples are magnified and distributed without a proper scale, however, they do indeed look disfiguring, but to do so is to grossly misrepresent our methodology and professionalism. We welcome informed and objective criticism, but far from being impartial and objectively critical Sauvet et al.'s poor reporting of our sampling is yet another example of an uninformed, often incorrect, partial and biased approach to what could be a profitable discussion of the merits and limitations of the various methods for dating cave art.

In this context, it is surprising that Pons-Branchu et al. (2014) support the sampling strategies of Aubert et al. (2007), and of which Sauvet et al. (2015) are only mildly critical; as Aubert et al.'s (2007) U-Th methodology is largely similar to ours, we can only imagine that this derives from a misunderstanding of Aubert et al.'s sampling methodology. Aubert et al. abrade small samples of calcite in the laboratory using a drill (*not*, as implied by Sauvet et al. (2015), by laser ablation; the laser merely measures U, Th and Fe concentrations to guide sample size and to identify the pigment layer). The sample sizes reported by Aubert et al. (2007) are 67, 301 and 447 mg, which are *considerably larger* than ours. The main difference between our sampling strategy and that of Aubert et al. (2007) is that the latter remove layers of calcite *as well as* pigment from the cave wall, by cutting or *coring through the art*. These samples may therefore include calcite stratified beneath the pigments of concern, which has the added advantage of providing maximum ages. But those concerned with the conservation of cave art (rather than of cave calcite) would surely not support such a strategy that removes portions of the art that it purports to date. We do not, never have, and would never, consider doing this.

Incidentally, we suspect that the sampling resolution of Aubert et al. (2014) has been overstated since a sample of 474 mg represents a volume of calcite 1 cm x 1 cm x 0.16 cm (assuming calcite density as 2.7 gcm<sup>-3</sup>), not 1 cm x 1 cm x 0.01 cm as Aubert et al. (2007), and subsequently Sauvet et al. and Pons-Branchu et al. report. If a layer 0.01 cm was sampled then the sample removed would have to be approximately 4

cm x 4 cm or approximately 16 times the surface area of our typical samples, or for a 1 cm x 1 cm sample, the thickness of the layer sampled would be 16 times the reported thickness.

*U-Th dating, hypotheses and interpretation: minimum ages, early cave art and its creators*

Sauvet et al. (2014) incorrectly suggest that a large number of U-Th dates we obtained for speleothems as part of a programme of parietal art dating in Cantabrian caves were 'much younger than expected and point to [sic] the Holocene.' Of course these results were not 'much younger than expected': we had no *a priori* expectations as to when speleothems formed, and we are of course well aware that speleothems did not stop forming after the Pleistocene, but commonly formed during the Holocene (e.g. Pons-Branchu et al., 2014, pp. 217 and also this volume); U-Th dates on calcites which form atop cave art simply provide *minimum ages* for the art on which they formed, a concept which Pons-Branchu et al. (2014) and Sauvet et al. (2015) appear to struggle with. Sauvet et al. (2015) note several studies in which the rate of growth of speleothems has been shown to be variable (from very rapid to intermittent to negligible) and come to the rather obvious conclusion that 'there is no general rule [of speleothem growth] and each case should be considered in relation to its own specific characteristics.' This is why U-Th measurements on speleothems overlying art provide minimum ages (and also why <sup>14</sup>C dates are unlikely to agree with U-Th dates). As Sauvet et al. (2015) note, if one were to assume that the age of the underlying art was for some reason close to the age of the overlying speleothem this would result in an underestimation of the art's age; yes, but of course no specialist would make this mistake. As to why Sauvet et al. (2015) regard the term 'minimum age' as 'euphemistic' we are unclear; as we have always made clear minimum ages are just that and we make no assumptions about how much older the underlying art is. We acknowledged this very issue in our interpretation of the minimum ages for a red disk and a hand stencil in El Castillo (García-Diez et al., 2015), where we stressed that with minimum ages of 37.3 ka and 40.8 ka respectively one *cannot rule out* the possibility that the art was created by

Neanderthals. We stand by this statement: if we did not, we would be guilty of a subjective interpretation which *ruled out* the possibility that Neanderthals produced these examples of art, even though there is no evidence for this. As undergraduates soon become aware, archaeology is often overturned by the appearance of evidence after long periods of its apparent absence. Surely we do not need to repeat the phrase *absence of evidence is not evidence of absence*. To Pons-Branchu et al. (2014), however, scientific openness and hypothesis testing represents a 'recurring quest for a "rudimentary" Neanderthal art...that would constitute the origins of the art of *Homo sapiens* [which] appears to us the reflection of our incapacity as researchers to conceive of another approach to the subject'. Furthermore they argue that we 'take advantage of the fact that this date is located within the confines of the transition between Neanderthals and the first Modern Humans to introduce the hypothesis that Neanderthals may have been the authors of these red marks. While there is no archaeological argument in favour of such an assertion, an ambiguous phrase such as ... "it cannot be ruled out that the earliest paintings were symbolic expressions of the Neanderthals" suffices to introduce doubt, even if the double negative appears to mitigate the comment.' (ibid., pp. 219).

We do not regard our statement as ambiguous. It is true – and we defy anyone to contradict – that it remains an *open question* as to whether or not Neanderthals produced examples of cave art. This is hardly ambiguous. Our critics may be surprised to hear that our null hypothesis can be expressed quite specifically, that *Neanderthals did not engage in painting in caves*. If they have any doubt about this they should contemplate how else we could construct a testable hypothesis on the basis of minimum ages for art. Had we constructed the alternative hypothesis, i.e. that Neanderthals did paint caves, then every single one of our minimum ages would support this; it would only be falsifiable if we had maximum ages for every piece of art, and the dates all fell after 42 ka (i.e. after the disappearance of Neanderthals in the region). If the journalists who subsequently covered our work reported that it was based on the hypothesis that Neanderthals painted caves, they might be forgiven for not understanding how scientific hypotheses are constructed and falsified, but we would expect members of the scientific community to. We currently



have no dating evidence that supports the painting of caves by Neanderthals, and this will remain so unless we find dates that falsify our hypothesis, but for some reason our critics would rather that we did not look.

These mistakes cut to the heart of scientific methodology. Clottes' (2012) account (implying that our 'apparent caution' in terminology was deliberately used to hand the 'gift' to journalists that suggested we were implying Neanderthals did create some of the art) does not help. How should one go about constructing and testing hypotheses? We know that Neanderthals were curating and using pigments and that they were frequenting deep caves; given this, and the lack of dating for the overwhelming majority of cave art currently known to us, it is surely a possibility that Neanderthals created some art. In terms of hypothesis forming, that there is no evidence of this as yet is immaterial: only a few years ago there was no convincing or accepted evidence that Neanderthals used pigments or created bone tools. While we must of course operate critically within the disciplinary consensus established on the basis of existing evidence, surely these authors do not recommend that we close ourselves off to hypothesis testing on the strength of 'consensus' established on the basis of flimsy evidence? Does anyone advocate the importance to science of 'thinking *inside* the box'? Clottes, perhaps, who appears to have misunderstood our rather simple point, however, noting that 'it is indeed rather foolhardy to put forth such a provocative *interpretation* on the basis of a single date. We must remain cautious and refrain from any excessive exploitation of these results until independent chronological and chronometric data are available to confirm them' (ibid., 6 our emphasis). To Clottes, our simple question – can we rule out or in that some of the earliest art in El Castillo was made by Neanderthals – has become *interpretation*. When even senior specialists make such rudimentary errors, this creates a field in which engaging in constructive debate becomes extremely difficult.

### *Discussion and debate*

Our final concerns reflect the misleading way in which the detractors of U-Th dating of cave art have distorted its efficacy both to the general public and to the scholarly

and heritage communities. We hope that we have dispensed with their methodological objections above. The issue of conservation of caves and cave art remains to be addressed. On this issue, Sauvet et al.'s (2015) argument contains two key passages:

'If the preservation of this invaluable heritage is taken into consideration, as it should always be, the damage caused to prehistoric artworks by sampling appears too high a cost with respect to the information gained'...

and...

'In recognizing this destruction, the 'Decorated Caves' section of the French Commission of Historical Monuments has recently prohibited 'the sampling of calcite for purposes of U-Th dating in the perimeter of decorated areas' (decision taken on 2013/10/24).'

With the second point Sauvet et al. (2015) magnify their objections to U-Th by giving them the weight of the backing of a governmental commission. There are several problems with their stance, however:

(a) As we have discussed above, Sauvet et al. (2015) misrepresent our methods, as we cause *no damage to the artwork* of concern, which remains untouched by the small-scale sampling of only the overlying calcite.

(b) If Sauvet et al. (2015) were objectively interested in the conservation issues raised by sampling for chronometric dates surely they would have called for a moratorium on the direct dating of cave art by AMS  $^{14}\text{C}$ , which, contrary to our method, *does entail destructive sampling of cave art*; despite this, and despite numerous objections that have been raised, but never addressed, about this method's application to cave art, they remain silent on  $^{14}\text{C}$ . It is obvious to us that the 'conservation argument' as presented, far from an objective and sensible archaeological concern, is merely a smokescreen used to confuse the real scientific issues.

(c) Sauvet et al. (2015) cite the decision of the 'Decorated Caves' section of the French Commission of Historical Monuments (CNMH) but fail to mention that the opinion of the physicist, presented to the Commission, surmised that the scientific method *was reliable* even though the archeological interpretation of the results obtained was disputable. In short, the 2013 CNMH decision is based entirely on the 'conservation' issue, rather than the scientific methodology that Clottes and Sauvet et al. in particular attack so vehemently.

(e) Finally, Sauvet et al. misrepresent the specific contents of the CNMH decision. What the CNMH actually decided was as follows:

*'La section « Grottes ornées » de la CNMH demande aux CIRA de ne pas autoriser la réalisation de prélèvements de calcite à des fins de datation dans le périmètre des champs ornés. Elle demande par ailleurs que toutes les demandes de prélèvement liées à cette méthode de datation soient systématiquement renvoyées devant elle pour un examen sous l'angle de la conservation.'*

To put this in plain English:

the CNMH does *not* prohibit the sampling of calcites underlying or overlying cave art; it simply demands that any such requests be forward to it by the regional authorities that are normally responsible for the issue of permits (the CIRAs), so that such requests can be assessed on the basis of potential conservation problems. To put this another way, it takes over from the CIRAs the power to issue the permits, but *per se* prohibits nothing.

### *Conclusions*

Scientific methodologies applied to major archaeological questions will always attract debate; that is the natural of science. Open and objective critiques should be encouraged, but these terms cannot be said to characterise the recent attacks on the

U-Th method that we have applied to the chronology of cave art. We hope that we have successfully dismissed unwarranted and misleading objections to the technique, and we hope that we have provided a justification as to why we believe that U-Th dating of calcites provides the best scientific method for dating cave art that currently exists.

No scientific dating method is without its negative aspects, but these should not be exaggerated. All processes of sampling, by their very nature, are destructive, although the discrete sizes of samples required for U-Th dating of calcites allow this method to be used without *any* damage to cave art. Its disadvantages, by contrast, are that the technique requires the measurement of a large number of samples, the costs of which in time and money are relatively large, and the necessity of working with minimum or maximum ages. Because of this, raising unjustified and unfounded doubts about the method does a disservice to the archaeological and Quaternary-science community. Why spread naive or incorrect rumours about a reliable and respected technique? This can only hinder the continued application of the technique and prevent the testing of archaeological hypotheses that are of interest to the field in its widest sense. Surely this serves only the maintenance of the very 'consensus' models that we should be questioning. What we would welcome is contributions to debate deriving from the desire to advance knowledge, promote understanding and improve methodologies.

### *References*

Arias P., Ontañón R., 2008. Zona arqueológica de La Garma (Omoño, Ribamontán al Monte). Campañas 2000-2003. In: Actuaciones Arqueológicas en Cantabria 2000-2003. Consejería de Cultura, Turismo y Deporte, Gobierno de Cantabria, Santander. pp. 43-60.

Aubert, M., O'Connor, S., McCulloch, M., Mortimer, G., Watchman, A., Richer-La Fleche, M., 2007. Uranium-series dating rock art in East Timor. *Journal of Archaeological Science* 34 (6), 991-996.

Aubert, M., Brumm, A., Ramli, R., Sutikna, T., Sapromo, E. W., Hakim, B., Morwood, M. J., Van Den Berg, G. D., Kinsley, L., Dosseto, A., 2014. Pleistocene cave art from Sulawesi, Indonesia. *Nature* 514: 223-227.

Cheng, H., Edwards, R. L., Murrell, M. T. and Benjamin, T. M., 1998. Uranium-thorium-protactinium dating systematics. *Geochimica et Cosmochimica Acta* 62 (21): 3437-3452.

Clottes, J., 2012. Datations U-Th, évolution de l'art et Néanderthal. *International Newsletter on Rock Art* 64: 1-6.

Corchón, M.S., Garate, D., Valladas, H., Rivero, O., Pons-Branchu, E., Hernando, C., 2014. Back to the point: new datings for La Peña de Candamo (Asturias). *Zephyrus* LXXIII: 67-81.

Corchón, M.S., Garate, D., Rivero, O., Valladas, H., Pons-Branchu, E., Murelaga, X., Ortega, P., Vicente, F.J., 2015. U-Series and  $^{14}\text{C}$  datings for a newly discovered decorated area in the Palaeolithic cave of La Peña de Candamo (Asturias, Northern Spain). *Journal of Archaeological Science Reports* 3: 371-80.

Fensterer, C., Scholz, D., Hoffmann, D., Mangini, A., Pajon, J.M., 2010.  $^{230}\text{Th}/\text{U}$ -dating of a late Holocene low uranium speleothem from Cuba. *IOP Conference Series: Earth and Environmental Science* 9, 012015.

Fohlmeister, J., Kromer, B., Mangini, A., 2011. The influence of soil organic matter age spectrum on the reconstruction of atmospheric  $^{14}\text{C}$  levels via stalagmites. *Radiocarbon* 53 : 99-115.

García-Diez, M. Garrido, D., Hoffmann, D.L., Pettitt, P. B., Pike, A.W.G., Zilhão, J., 2015. The chronology of hand stencils in European Palaeolithic rock art: implications

of new U-series results from El Castillo Cave (Cantabria, Spain). *Journal of Anthropological Sciences* 93: 1-18.

Genty, D., Massault, M., 1999. Carbon transfer dynamics from bomb-14C and d13C time series of a laminated stalagmite from SW France - Modelling and comparison with other stalagmite records. *Geochimica et Cosmochimica Acta* 63: 1537-1548.

Genty, D., Baker, A., Massault, M., Proctor, C., Gilmour, M., Pons-Branchu, E., Hamelin, B., 2001. Dead carbon in stalagmites: Carbonate bedrock paleodissolution vs. ageing of soil organic matter. Implications for C-13 variations in speleothems. *Geochimica et Cosmochimica Acta* 65: 3443-3457.

Genty, D., Konik, S., Valladas, H., Blamart, D., Hellstrom, J., Touma, M., Moreau, C., Dumoulin, J.-P., Nouet, J., Dauphin, Y., Weil, R., 2011. Dating the Lascaux cave gour formation. *Radiocarbon* 53: 479-500.

González Sainz, C., 2003. El conjunto parietal de la galería inferior de La Garma (Omoño, Cantabria). Avance a su organización interna. In: *El Arte Prehistórico desde los inicios del siglo XXI. Primer Symposium Internacional de Arte Prehistórico de Ribadesella, Ribadesella*, pp. 201-222.

Griffiths, M.L., Fohlmeister, J., Drysdale, R.N., Hua, Q., Johnson, K.R., Hellstrom, J.C., Gagan, M.K., Zhao, J.-X., 2012. Hydrological control of the dead carbon fraction in a Holocene tropical speleothem. *Quaternary Geochronology* 14: 81-93.

Gutjahr, M., Vance, D., Hoffmann, D.L., Hillenbrand, C.-D., Foster, G.L., Rae, J.W.B., Kuhn, G., 2013. Structural limitations in deriving accurate U-series ages from calcitic cold-water corals contrast with robust coral radiocarbon and Mg/Ca systematics. *Chemical Geology* 355: 69-87.

Hoffmann, D.L., Spötl, C. and Mangini, A., 2009. Micromill and in situ laser ablation sampling techniques for high spatial resolution MC-ICPMS U-Th dating of carbonates. *Chemical Geology* 259: 253-261.

Hoffmann, D.L., Beck, J.W., Richards, D.A., Smart, P.L., Singarayer, J.S., Ketchmark, T. and Hawkesworth, C.J., 2010. Towards radiocarbon calibration beyond 28 ka using speleothems from the Bahamas. *Earth and Planetary Science Letters* 289: 1-10.

Holmgren K, Lauritzen S.E, Possnert G., 1994.  $^{230}\text{Th}$ / $^{234}\text{U}$  and  $^{14}\text{C}$  dating of a Late Pleistocene stalagmite in Lobatse II cave, Botswana. *Quaternary Science Reviews* 13: 111-119.

Koornneef, J.M., Stracke, A., Aciego, S., Reubi, O., Bourdon, B., 2010. A new method for U–Th–Pa–Ra separation and accurate measurement of  $^{234}\text{U}$ – $^{230}\text{Th}$ – $^{231}\text{Pa}$ – $^{226}\text{Ra}$  disequilibria in volcanic rocks by MC-ICPMS. *Chemical Geology* 277 (1) : 30-41.

Pazdur, A., Pazdur, M.F., Pawlyta, J., 1995. Paleoclimatic implications of radiocarbon dating of speleothems from the Cracow-Wielun upland, southern Poland. *Radiocarbon* 37: 103-110.

Pettitt, P., Bahn, P.G., 2003. Current problems in dating Palaeolithic cave art: Candamo and Chauvet. *Antiquity* 77: 134-41.

Pettitt, P., Bahn, P., 2015. An alternative chronology for the art of Chauvet Cave. *Antiquity* 89: 1-12.

Pike, A.W.G., Hoffmann, D.L., García-Diez, M., Pettitt, P.B., Alcolea, J., González-Sainz, C., de las Heras, C., Lasheras, J.A., Montes, R., Zilhão, J., 2012. Uranium-series dating of Upper Palaeolithic art in Spanish caves. *Science* 336: 1409-13.

Plagnes, V., Causse, C., Fontugne, M., Valladas, H., Chazine, J. M., Fage, L.H., 2003. Cross dating (Th/U- $^{14}\text{C}$ ) of calcite covering prehistoric paintings in Borneo. *Quaternary Research* 60 (2): 172-179.

Pons-Branchu, E., Bourillon, R., Conkey, M.W., Fontugne, M., Fritz, C., Gárate, D., Quiles, A., Rivero, O., Sauvet, G., Tosello, G., Valladas, H., White, R., 2014. Uranium-series dating of carbonate formations overlying Palaeolithic art; interest and limitations. *Bulletin de la Société Préhistorique Française* 111 (2): 211-24.

Reimer, P.J., Bard, E., Bayliss, A., Beck, J.W., Blackwell, P.G., Bronk Ramsey, C., Buck, C.E., Cheng, H., Edwards, R.L., Friedrich, M., Grootes, P.M., Guilderson, T.P., Hafliðason, H., Hajdas, I., Hatté, C., Heaton, T.J., Hoffmann, D.L., Hogg, A.G., Hughen, K.A., Kaiser, K.F., Kromer, B., Manning, S.W., Niu, M., Reimer, R.W., Richards, D.A.,

Scott, E.M., Southon, J.R., Staff, R.A., Turney, C.S.M., van der Plicht, J., 2013. IntCal13 and Marine13 radiocarbon age calibration curves 0 - 50000 years cal BP.

Radiocarbon 55: 1869-1887.

Richards, D.A., Dorale, J.A., 2003. Uranium-series chronology and environmental applications of speleothems. In: Bourdon, B., Henderson, G.M., Lundstrom, C.C. and Turner, S.P. (Eds.), Uranium-series Geochemistry. Mineralogical Society of America, Washington DC, pp. 407-460.

Sauvet, G., Bourrillon, R., Conkey, M., Fritz, C., Gárate, D., Rivero, O., Tosello, G., White, R., 2015. Uranium-Thorium dating method and rock art. Quaternary International (doi:10.1016/j.quaint.2015.03.053).

Scholz, D., Tolzmann, J., Hoffmann, D.L., Jochum, K.P., Spötl, C., Riechelmann, D.F.C., 2014. Diagenesis of speleothems and its effect on the accuracy of  $^{230}\text{Th}/\text{U}$ -ages. Chemical Geology 387: 74-86.

Shen, C.C., Cheng, H., Edwards, R.L., Moran, S.B., Edmonds, H.N., Hoff, J.A., Thomas, R.B., 2003. Measurement of attogram quantities of  $^{231}\text{Pa}$  in dissolved and particulate fractions of seawater by isotope dilution thermal ionization mass spectroscopy. Analytical Chemistry 75 (5): 1075-1079.

Southon, J., Noronha, A., Cheng, H., Edwards, R.L., Wang, Y., 2012. A high-resolution record of atmospheric  $^{14}\text{C}$  based on Hulu Cave speleothem H82. Quaternary Science Reviews 33: 32-41.

Valladas, H., Clottes, J. 2003. Style, Chauvet and radiocarbon. Antiquity 77: 142-45.

Valladas, H., Tisnérat-Laborde, N., Cachier, H., Kaltnecker, E., Arnold, M., Oberlin, C., Évin, J., 2005. Bilan des datations carbone 14 effectué sur des charbons de bois de la grotte Chauvet. Bulletin de la Société Préhistorique Française 102 : 109-13.

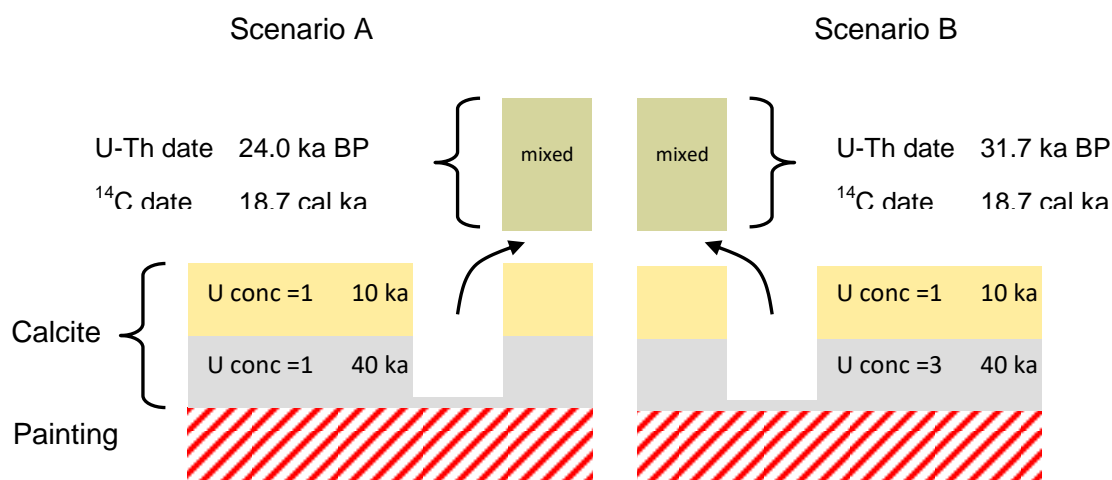


**Table 1.** U concentrations for U-Th samples in Pike et al. (2012).

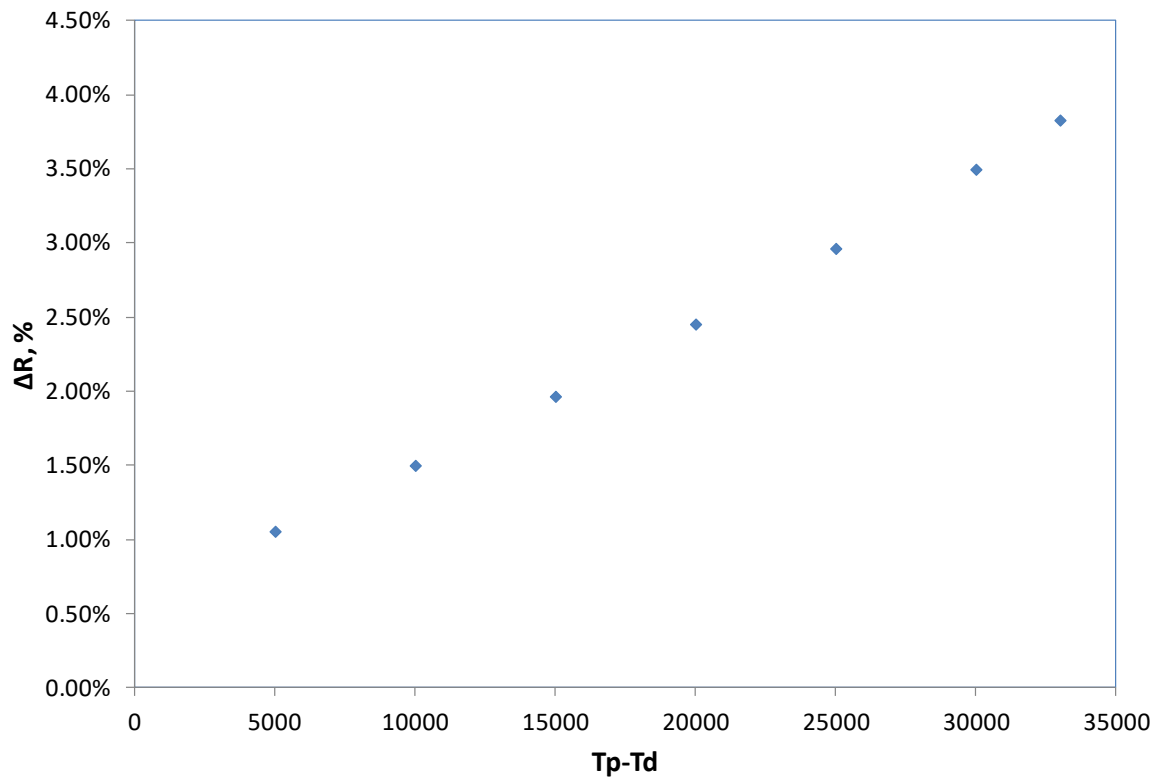
Sample BIG- UTh-	Site	Description	U (ng/g)
O-30	Tito Bustillo	Overlies red horse, <i>Ensemble X</i>	837.4±4.1
O-101	La Pasiega	Overlies red bovid, Pasiega C	724.8±6.5
O-103	La Pasiega	Overlies red megaloceros, Pasiega B	1530±10
O-109	La Pasiega	Overlies red undetermined figure, Pasiega B	1330±12
O-88	El Castillo	Overlies small red dot, <i>Gran Sala</i>	2000±12
O-106	La Pasiega	Overlies red undetermined figure, Pasiega B	9817±60
O-71	Altamira	Overlies black ibex, <i>La Hoya</i>	2290±92
O-107	La Pasiega	Overlies red bison, Pasiega B	599.0±3.9
O-108	La Pasiega	Overlies red bison, Pasiega B	673±4.4
O-105	La Pasiega	Overlies red horses, Pasiega B	1424.0±7.9
O-110	La Pasiega	Overlies red horse, Pasiega B	901.4±9.2
O-73	La Pasiega	Overlies red triangular symbol, Pasiega C	1318.8±7.2
O-102	La Pasiega	Overlies black ibex, Pasiega C	944.7±6.4
O-76	La Pasiega	Overlies red claviform, Pasiega B	54.9±1.2
O-46	Altamira	Overlies red tectiform, sector III	4026±38
O-84	El Castillo	Overlies red deer, <i>Galería del Bisonte'</i>	235.5±1.6
O-77	Covalanas	Overlies red bovid	358.5±2.9
O-78	Santián	Overlies red "hand-like" symbol	158.97±0.75
O-22	Tito Bustillo	Red pigment associated with anthropomorphic figure, <i>Galería de los Antropomorfos</i>	84.4±7.4
O-98	La Pasiega	Overlies small red dot, Pasiega C	789.0±5.0.
O-68	El Castillo	Overlies black horse, <i>El Paso</i>	133.22±0.87
O-56	Covalanas	Overlies red deer	628.9±4.8
O-60	Santián	Overlies red colour concentration on stalagmitic pillar, Main Corridor	72.84±0.72
O-91	El Castillo	Overlies black bovid, <i>Galería del Bisonte</i>	146.2±1.1
O-74	La Pasiega	Overlies yellow double arch motif, Pasiega C	911.1±5.6
O-100	La Pasiega	Overlies red deer, Pasiega C	1009±21
O-89	El Castillo	Overlies red 'bell', <i>Panel de los Campaniformes</i>	725.2±4.8
O-85	El Castillo	Overlies red rectangular motif, <i>Galería del Bisonte</i>	90.26±0.73
O-23	Tito Bustillo	Overlies red vulva, <i>Cámara de las vulvas</i>	139±12
O-97	La Pasiega	Overlies red deer, Pasiega C	150.00±0.83

O-17	Tito Bustillo	Overlies violet horse, <i>Ensemble IX</i>	1500.4±8.4
O-99	La Pasiega	Overlies red dot, Pasiega C	397.0±2.1
O-40	Las Aguas	Overlies red and engraved bison, Principal Panel	3580±350
O-14	Tito Bustillo	Overlies red horse, <i>Ensemble X</i>	219.1±1.1
O-86	El Castillo	Overlies black bison, <i>El Paso</i>	84.08±0.68
O-12	Tito Bustillo	Red horse head, <i>Ensemble X</i>	210.1±1.1
O-9	Tito Bustillo	Red horse, <i>Ensemble X</i>	154.47±0.80
O-67	El Castillo	New growth of broken scarf stalactite with red disk, <i>Galería del Bisonte</i>	90.02±0.51
O-81	El Castillo	Overlies red disk, <i>Corredor Techo de las Manos</i>	150.5±1.1
O-72	La Pasiega	Overlies red triangle, Pasiega C	509.0±2.9
O-43	Las Aguas	Overlies red quadrangular symbol, Chamber of Engravings	1470±150
O-53	Altamira	Overlies red 'spotted outline' horse, <i>Techo de los Polícromos</i>	9160±83
O-70	Las Aguas	Overlies brown 'T' sign, Principal Panel	1397±14
O-80	El Castillo	Overlies black indeterminate animal, <i>Corredor Techo de las Manos</i>	260.3±1.9
O-58	El Castillo	Overlies red negative hand stencil, <i>Techo de las Manos</i>	1646±11
O-21	Tito Bustillo	Red pigment associated with anthropomorphic figure, <i>Galería de los Antropomorfos</i>	112.00±0.61
O-69	El Castillo	Red disk, <i>Galería de los Discos</i>	373.9±2.0
O-50	Altamira	Overlies red claviform-like symbol, <i>Techo de los Polícromos</i>	1276±13
O-82	El Castillo	Overlies red negative hand stencil and underlies yellow outlined bison, <i>Panel de las Manos</i>	643.7±3.7
O-83	El Castillo	Overlies large red disk, <i>Panel de las Manos</i>	398.1±2.1

**Figure 1** Comparison of U-Th and  $^{14}\text{C}$  dates in mixed calcite layers of equal thickness but different ages. For both scenarios, the painting is covered in two layers of calcite, the youngest formed at 10ka BP and the oldest at 40ka BP. In Scenario A, both layers have the same U concentration (U conc=1). In scenario B, the oldest layer has 3 times the uranium (U conc = 3). The 'sample' is an equal mixture of the two layers. Because of the difference in the two radioactive systems ( $^{14}\text{C}$  decay vs  $^{230}\text{Th}$  accumulation) the date of the sample shows discordance between the  $^{14}\text{C}$  and the U-Th dating methods.



**Figure 2.** Predicted differences in  $^{231}\text{Pa}/^{235}\text{U}$  between a 41 ka closed system sample, and a sample of true age  $T_p$  of 33 ka that has undergone a single U loss event at age  $T_p - T_d$ . The parameter,  $F$ , defined by Cheng et al. (1998) as the change in U (i.e.  $U_{\text{old}}/U_{\text{new}}$ ) due to loss or gain is calculated to keep the  $^{230}\text{Th}/^{238}\text{U} = 0.313$  (i.e. to give an apparent U-Th age of 41 ka, to simulate the approximate age of our oldest sample).  $\Delta R$  is calculated as the % difference between the closed system, 41ka  $^{231}\text{Pa}/^{235}\text{U}$  activity ratio, and the open system  $^{231}\text{Pa}/^{235}\text{U}$  activity ratio.



**Figure 3.** Examples of our calcite sampling locations (after sampling) with scales. (A) Our example from Tito Bustillo illustrated by Sauvet et al., which is approximately 2cm in maximum dimension, is the largest and one of the earliest samples we have ever taken. The surface area of a sample will be defined by the thickness of the calcite, but methodological developments in the last 7 years mean that we can now work with samples as small as 10mg, approximately 8 times smaller than the sample removed from Tito Bustillo. (B) + (C) show examples more typical of our sampling, both taken in Maltravieso, which are around 1cm in maximum diameter. All images are of the cave wall after the total sample has been removed (i.e. at the end of sampling).

