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## Dating Palaeolithic cave art: Why U–Th is the way to go

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## 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 that 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.

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## 1. 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 (<sup>14</sup>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 <sup>14</sup>C have been intensely debated, and some that were initially published even withdrawn (Pettitt and Bahn, 2003 contra; Valladas and Clottes, 2003).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 (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 <sup>14</sup>C.

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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.

## 2. 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](#); [Fensterer et al., 2010](#); [Hoffmann et al., 2010](#); [Gutjahr et al., 2013](#); [Scholz et al., 2014](#)). But every geochemist, however, would acknowledge that open-system behaviour of speleothem calcite 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 and misleading bias into the debate.

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 also 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 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 through 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

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