## LETTERS

## Age of Zhoukoudian *Homo erectus* determined with <sup>26</sup>Al/<sup>10</sup>Be burial dating

Guanjun Shen<sup>1</sup>, Xing Gao<sup>2</sup>, Bin Gao<sup>1</sup> & Darryl E. Granger<sup>3</sup>

The age of Zhoukoudian Homo erectus, commonly known as 'Peking Man', has long been pursued, but has remained problematic owing to the lack of suitable dating methods<sup>1-7</sup>. Here we report cosmogenic <sup>26</sup>Al/<sup>10</sup>Be burial dating of quartz sediments and artefacts from the lower strata of Locality 1 in the southwestern suburb of Beijing, China, where early representatives of Zhoukoudian Homo erectus were discovered. This study marks the first radioisotopic dating of any early hominin site in China beyond the range of mass spectrometric U-series dating. The weighted mean of six meaningful age measurements,  $0.77 \pm 0.08$  million years (Myr, mean  $\pm$  s.e.m.), provides the best age estimate for lower cultural layers 7-10. Together with previously reported U-series dating of speleothem calcite<sup>3</sup> and palaeomagnetic stratigraphy<sup>4</sup>, as well as sedimentological considerations<sup>8,9</sup>, these layers may be further correlated to S6-S7 in Chinese loess stratigraphy or marine isotope stages (MIS) 17-19, in the range of ~0.68 to 0.78 Myr ago. These ages are substantially older than previously supposed and may imply early hominin's presence at the site in northern China through a relatively mild glacial period corresponding to MIS 18.

With an inventory of 6 fairly complete hominin crania and bones representing at least 40 individuals, 98 species of non-hominin mammalian fossils and tens of thousands of stone artefacts, the cave site of Zhoukoudian Locality 1 has remained the largest single source of *Homo erectus* and is one of the most important Palaeolithic sites in the world<sup>1</sup>. The site is a sedimentary infill within a vertical karstic fissure. Its ~40m-thick depositional sequence can be divided into 17 layers<sup>10</sup>. The lowermost layers 11–17 are fluvial, layers 6–10 are breakdown breccia from the cave walls and ceiling interbedded with silt and sand, layer 5 is travertine, and the uppermost layers 1–4 are silt and travertine with minor breakdown that accumulated after collapse of the ceiling<sup>11</sup>. Stone artefacts and hominin fossils have been recovered from layers 1–10, with most from a lower level in layers 8/9 and an upper level in layers 3–4 (ref. 1). Mammalian fossils are found in layers 1–13, with some primitive carnivores disappearing above layer 5 (ref. 12).

As part of a multidisciplinary study initiated in the late 1970s<sup>1</sup>, dating was carried out at several Chinese institutions using a variety of techniques. The following age sequence was proposed: ~700 thousand years (kyr) for the lowest fossiliferous layer 13, based mainly on palaeomagnetic stratigraphy<sup>4</sup>; ~500 kyr for the lowest hominin-fossil-bearing layer 10, based on fission-track dating of sphene grains; and ~230 kyr for the uppermost layers 1–3, based on  $^{230}$ Th/ $^{234}$ U dating of fossil materials<sup>2</sup>. These age assignments were generally supported by later  $^{231}$ Pa/ $^{235}$ U (ref. 5), fission-track<sup>6</sup> and electron spin resonance dating<sup>7</sup>. A time range of ~230 to 500 kyr ago for the hominin-fossil-bearing layers has been widely accepted by palaeoanthropologists, although with a few critical comments<sup>13</sup>.

In contrast, much older ages were determined using mass spectrometric U-series dating of intercalated pure and dense calcite samples, known to be a more reliable chronometer<sup>14,15</sup>. A date of  $400 \pm 8$  kyr ago was proposed for an upper horizon of layers 1/2,  $\sim 500$  kyr ago for the upper part of layer 5, and  $\geq 600$  kyr ago for the middle and lower parts of layer 5 (Fig. 1, ref. 3).

The suggestion that Zhoukoudian H. erectus is substantially older than previously estimated remains to be validated by independent checks. However, numerical dating beyond the upper limit of mass spectrometric U-series dating, ~600 kyr ago, is difficult in China because the lack of contemporaneous volcanic activity nearly precludes the application of <sup>40</sup>Ar/<sup>39</sup>Ar dating. Here we use burial dating with cosmogenic <sup>26</sup>Al and <sup>10</sup>Be in quartz<sup>16–19</sup>, which is often suitable for allochthonous cave sediments such as those at Locality 1. This method is based on radioactive decay of <sup>26</sup>Al  $(t_{1/2} = 717 \pm 17 \text{ kyr})^{17}$ and  ${}^{10}\text{Be} (t_{1/2} = 1.36 \pm 0.07 \text{ Myr})^{20}$ . These two nuclides are produced with a known  ${}^{26}\text{Al}/{}^{10}\text{Be}$  atomic ratio of 6.8:1 in quartz exposed to secondary cosmic radiation near the ground surface. Their initial concentrations depend on the mineral's exposure time, which in turn is controlled by the erosion rate of the host rock. If quartz grains from the surface are deeply buried, for example by deposition in a cave, then the production of cosmogenic nuclides nearly stops. Because <sup>26</sup>Al decays faster than <sup>10</sup>Be, the <sup>26</sup>Al/<sup>10</sup>Be ratio decreases exponentially with an apparent half-life of 1.52 Myr. This offers a means for dating quartz burial up to  $\sim$ 3–5 Myr ago<sup>16</sup>.

Burial dating was first applied to quartz gravels in caves for deriving river incision rates<sup>17</sup>. It was later applied to hominin sites at Sterkfontein in South Africa<sup>18</sup> and Sima del Elefante at Atapuerca in Spain<sup>19</sup>. The strengths of this method are its radiometric basis and its independence from other dating methods. However, it must be recognized that cave sediments can have complex stratigraphy, particularly in vadose fills. If fossils are mixed with quartz sediments with a prior burial history, the resulting age will be erroneously old.

Six quartz-bearing sand samples were collected, two of them (ZKD-12 and ZKD-13) from fluvial deposits in layers 12 and 13 and the other four (ZKD-6, ZKD-7, ZKD-8/9 and ZKD-10) from quartz-rich lenses or sublayers in layers 6, 7, 8/9 and 10, respectively. Moreover, four quartzite artefacts that directly indicate hominin presence were analysed from collections made in the 1930s from layers 8/9. The <sup>26</sup>Al and <sup>10</sup>Be concentrations and corresponding burial ages are presented in Table 1.

Three of the four quartzite artefacts yield results consistent within  $1\sigma$ , with an error-weighted mean age of  $0.72 \pm 0.13$  Myr. The fourth artefact (ST-3) gives an aberrant result of  $1.66 \pm 0.21$  Myr. This particular sample could have been taken from an older cave fill or terrace before manufacture. Among the sediment samples, those from layers 7, 8/9 and 10 yield consistent results, with an error-weighted mean age of  $0.81 \pm 0.11$  Myr. This is slightly older than, but within error of, the weighted mean of the results from the three artefacts, indicating that some sand might have entered the cave with a previous burial signal.

<sup>1</sup>College of Geographical Sciences, Nanjing Normal University, Nanjing, Jiangsu 210046, China. <sup>2</sup>Institute of Vertebrate Paleontology and Paleoanthropology, Academia Sinica, Beijing 100044, China. <sup>3</sup>Department of Earth and Atmospheric Sciences, Purdue University, West Lafayette, Indiana 47907-2051, USA.



Figure 1 Correlation of Zhoukoudian Locality 1 with loess stratigraphy and marine isotope record from Ocean Drilling Program (ODP) core 677 (ref. 21). The depositional sequence is constrained in time by the Brunhes/ Matuyama (B/M) palaeomagnetic boundary<sup>4</sup>, U-series ages of flowstones<sup>3</sup>, and cosmogenic burial ages reported here. Shading indicates primarily waterlain layers for Locality 1 stratigraphy<sup>11</sup> and palaeosols for the Luochuan loess profile<sup>22</sup>. Roman numerals denote the locations of hominin skulls<sup>1,11</sup>.

ZKD-6 gives an aberrant result,  $2.78 \pm 0.51$  Myr. This sample may possibly date to an earlier phase of cave formation, as it was collected from a thin sandy layer that is adhered to the north wall and is now out of stratigraphic contact with the main cross-section. The two samples from the basal fluvial sediments do not yield statistically meaningful results. Their inherited cosmogenic nuclide concentrations are quite low due to rapid erosion in their source area, leading to large uncertainty. Taken together, we consider the weighted mean of the six meaningful measurements,  $0.77 \pm 0.08$  Myr, to best represent the age for layers 7–10. This is consistent with both previous U-series<sup>3</sup> and palaeomagnetic<sup>4</sup> data.

Table 1	Cosmogenic nuclic	le concentrations	and burial ages
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Sample	[ $^{26}$ Al] (10 $^{6}$ at g $^{-1}$ )	[ $^{10}$ Be] (10 $^{6}$ at g $^{-1}$ )	$^{26}\text{Al/}^{10}\text{Be}$	Burial age*(Myr)	
Sediment					
ZKD-6	$0.073 \pm 0.018$	$0.040 \pm 0.004$	$1.82 \pm 0.49$	2.78 ± 0.51 (0.54)	
ZKD-7-2	$0.550 \pm 0.053$	$0.132 \pm 0.009$	$4.17 \pm 0.49$	1.00 ± 0.23 (0.24)†	
ZKD-8/9	$1.252 \pm 0.095$	$0.273 \pm 0.008$	$4.58 \pm 0.38$	0.75 ± 0.16 (0.17)†	
ZKD-10-2	0.568 ± 0.052	$0.120 \pm 0.006$	$4.72 \pm 0.50$	0.75 ± 0.21 (0.22)†	
ZKD-12	$0.105 \pm 0.030$	$0.021 \pm 0.006$	$5.10 \pm 2.01$	0.62 ± 0.74 (0.74)	
ZKD-13	$0.106 \pm 0.028$	$0.018 \pm 0.005$	5.89 ± 2.35	0.31 ± 0.74 (0.74)	
Artefacts (8/9)					
ST-1	0.199 ± 0.027	$0.040 \pm 0.002$	4.95 ± 0.72	0.67 ± 0.29 (0.29)†	
ST-2	0.476 ± 0.037	$0.100 \pm 0.003$	4.77 ± 0.39	0.73 ± 0.17 (0.17)*	
ST-3	0.371 ± 0.039	$0.122 \pm 0.003$	3.04 ± 0.33	1.66 ± 0.21 (0.24)	
ST-4	$0.568 \pm 0.083$	$0.120 \pm 0.005$	$4.72\pm0.71$	0.75 ± 0.29 (0.30)†	

\* Uncertainties  $(\pm 1\sigma)$  are expressed in two ways: the first includes analytical uncertainty only, and should be used when comparing burial ages; the second uncertainty listed in parentheses also includes systematic errors in half-lives, and should be used when comparing against ages from other radiometric dating methods.

† Ages included in the weighted mean for layers 7-10

Superscripts on ages indicate dating method (a, cosmogenic nuclides in artefacts; s, cosmogenic nuclides in sediment; u, U-series). Magnetostratigraphic data<sup>4</sup> are indicated by closed, open and half-closed circles for normal, reverse and uncertain polarity, respectively. Luochuan loess magnetic susceptibility data (unpublished) are provided courtesy of Dr Liping Zhou. Marine isotope stages are shown in italics.

The above age assignment may be refined in the context of the cave environment. Several previous studies have correlated sedimentary packages at Zhoukoudian Locality 1 with the Chinese loess stratigraphy and the marine isotope record<sup>8,9,21</sup>. Layers that predominantly consist of breakdown breccia (6, 8/9) or loess (4) may be correlated with colder, drier periods, whereas intervening lavers (3, 5, 7 and 10) consisting of waterlain sediments and/or flowstones may correspond with warmer, more humid periods (Fig. 1). Following the loess timescale<sup>22</sup>, the well-dated flowstone in layer 5 is probably associated with palaeosol 5 (S5) and MIS 13–15 from  $\sim$ 500–600 kyr ago, a period of prolonged warmth and humidity. Continuing downwards, the layer 6 breccia best corresponds to loess 6 (L6) and MIS 16. Layer 7 and the uppermost part of layers 8/9, containing waterlain sediments interbedded with flowstone, would correspond to S6 and MIS 17. The lower part of layers 8/9 is breakdown breccia, and would correspond to L7. The waterlain sediments in layer 10 and below, including the Brunhes/Matuyama boundary between layers 13 and 14, would then correspond to S7 and MIS 19 (refs 4 and 22).

The mammalian fauna support this interpretation. They indicate mixed steppe and forest environments with a trend towards increasing grasslands over time. Layer 5 contains unambiguous warm-climate fauna<sup>8,9</sup>. Other layers show a preponderance of cold-climate fauna associated with the breccia and more warm-climate fauna in the intervening sediments<sup>8,9</sup>. Limited oxygen isotope data also support climatic shifts. Teeth from *Equus sanmeniensis* in layers 8/9 and 4 (glacial) have  $\delta^{18}$ O values 3–4‰ lower than those in layers 10–11 (interglacial)<sup>23</sup>, consistent with enhanced monsoon strength that occurred during the colder, drier intervals.

The above climatic correlations indicate that layers 7–10, including the lower cultural level and the first hominin appearance at Locality 1, lie within the range of S6–S7 and MIS 17–19 from ~0.68 to 0.78 Myr ago. The lower cultural level in the breccia of layers 8/9 would thus correspond to L7 and MIS 18. Pending further confirmation, the assignment of these layers to a cooler and drier episode may imply hominin presence at the site through glacial–interglacial cycles. However, L7 and MIS 18 correspond to a relatively mild glacial period, as indicated by both marine  $\delta^{18}$ O and soil development within L7 (Fig. 1). Such mild glacial conditions may have been necessary for early *H. erectus pekinensis* to persist in northern China. Together with previous U-series dating of flowstone in layers 1/2, the hominin presence at the site is constrained to a total range of 0.40–0.78 Myr ago.

A reliable chronology is critical for resolving debate over the mode of Middle Pleistocene human evolution in East Asia<sup>24–26</sup>. Previously, the chronology of Chinese sites has been largely based on the U-series and electron spin resonance dating of fossil materials, which are known to be vulnerable to post-burial U migration<sup>14,27</sup>. <sup>230</sup>Th/<sup>234</sup>U dating of speleothem calcita<sup>3,28,29</sup> has repeatedly shown that the previous timescale for Middle–Late Pleistocene hominin sites in China may have been underestimated as a whole. The results of this paper show that such a tendency persists beyond the range of mass spectrometric U-series dating. It is foreseeable that <sup>26</sup>Al/<sup>10</sup>Be burial dating will be applied to other hominin sites in China and elsewhere, contributing substantially to a robust chronological framework and thereby to a better understanding of human evolution.

## **METHODS SUMMARY**

For quartzose sand samples, several kilograms of sediment were collected. Silt and clay were removed by a water rinse, and carbonates were dissolved in HCl. The remaining quartz-rich sand was sieved to >0.2 mm and leached several times in hot 5% HF/HNO<sub>3</sub> overnight (>12 h) with agitation. Following magnetic and gravimetric separation, the resulting quartz consisted of two populations: a darker-coloured quartz with a high native aluminium concentration, and a lighter-coloured quartz with a lower aluminium concentration. The darker-coloured grains were removed by handpicking. For quartzite artefacts, the samples were thoroughly cleaned in 1% HF/HNO<sub>3</sub> and then crushed to a grain size <0.5 mm. The quartz was further purified by repeated overnight leaching in 1% HF/HNO<sub>3</sub> in an ultrasonic tank.

Purified quartz was dissolved in 5:1 HF/HNO<sub>3</sub>, and spiked with ~0.3 mg <sup>9</sup>Be prepared from beryl. An aliquot was taken for aluminium determination by inductively-coupled plasma optical emission spectrometry using the method of standard additions. After evaporation and fuming of HF in H<sub>2</sub>SO<sub>4</sub>, aluminium and beryllium were separated on ion-exchange columns in 0.4 M oxalic acid, precipitated as hydroxides, and transformed to oxides in a furnace at 1,100 °C. BeO was mixed with niobium and Al<sub>2</sub>O<sub>3</sub> with silver for <sup>10</sup>Be/<sup>9</sup>Be and <sup>26</sup>Al/<sup>27</sup>Al measurement by accelerator mass spectrometer at PRIME Lab, Purdue University.

Burial ages were calculated following ref. 16. For samples in this paper, postburial production of cosmogenic nuclides by muons is safely ignored. Production rates are estimated for latitude 39° N and elevation 120 m (ref. 30), and adjusted for a revised <sup>10</sup>Be half-life<sup>20</sup>.

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**Author Contributions** G.S. and D.E.G. contributed equally to conceiving the project, organizing fieldwork, interpreting data and preparing the paper. X.G. provided access to the site and quartzite artefacts. Chemistry was performed by B.G. and D.E.G.

Author Information Reprints and permissions information is available at www.nature.com/reprints. Correspondence and requests for materials should be addressed to G.S. (gishen@njnu.edu.cn) or D.E.G. (dgranger@purdue.edu).