RECONCILING THE GREENLAND ICE-CORE AND RADIOCARBON TIMESCALES THROUGH THE LASCHAMP GEOMAGNETIC EXCURSION

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Abstract:

Cosmogenic radionuclides, such as ^{10}Be and ^{14}C, share a common production signal, with their formation in the Earth's upper atmosphere modulated by changes to the geomagnetic field, as well as variations in the intensity of the solar wind. Here, we use this common production signal to compare between the radiocarbon (IntCal) and Greenland ice-core (GICC05) timescales, utilising the most pronounced cosmogenic production peak of the last 100,000 years – that associated with the Laschamp geomagnetic excursion circa 41,000 years ago. We present 54 new ^{14}C measurements from a peat core ('TP-2005') from Tenaghi Philippon, NE Greece, contiguously spanning between circa 47,300 and 39,600 cal. BP, demonstrating a distinctive tripartite structure in the build up to the principal Laschamp production maximum that is not present in the consensus IntCal13 calibration curve. This is the first time that a continuous, non-reservoir corrected ^{14}C dataset has been generated over such a long time span for this, the oldest portion of the radiocarbon timescale. This period is critical for both palaeoenvironmental and archaeological applications, with the replacement of Neanderthals by anatomically modern humans in Europe around this time. By placing our Tenaghi Philippon ^{14}C dataset on to the Hulu Cave U-series timescale of Cheng et al. (2018) via Bayesian statistical modelling, the comparison of TP-2005 ^{14}C with Greenland ^{10}Be fluxes also implicitly relates the underlying U-series and GICC05 timescales themselves. This comparison suggests that whilst these two timescales are broadly coherent, the IntCal13 timescale is likely some ~1000 years too old circa 40,000 cal. BP.

1. Introduction

Among the most pressing questions in palaeoenvironmental research today is the reliable identification of synchronies or asynchronies of past climatic and environmental changes across the globe. A fundamental problem in identifying such temporal relationships in palaeorecords, however, is an inability to reliably compare inter-regional records beyond the limits of chronological uncertainty.

Arguably, the best and most-widely cited record of palaeoclimatic change – the key global reference ‘type site’ – is that provided by the Greenland ice-cores, due to their highly resolved suite of multi-proxy palaeoenvironmental data (NGRIP members, 2004; Steffensen et al., 2008), and their annual resolution, layer-counted chronology (Andersen et al., 2006; Rasmussen et al., 2006; Svensson et al., 2008). Conversely, the most utilised geochronological technique applied to late Quaternary palaeoenvironmental (and archaeological) sites elsewhere in the world is provided by radiocarbon (^{14}C) dating (Brauer et al., 2014). However, in order to compare data between the two timescales, one must assume that the respective ^{14}C and icecore layer-counted chronologies are consistent – an assumption that must undoubtedly incorporate uncertainties (Adolphi and Muscheler, 2016).

Here, we utilise the common production signal of the cosmogenic radionuclides ^{10}Be (beryllium-10) and ^{14}C (radiocarbon) to link together the Greenland ice-core and radiocarbon timescales for the oldest ~10,000 years of the radiocarbon timescale (i.e. the last ~50,000 years), taking advantage of the most pronounced cosmogenic production peak of the last 100,000 years – that associated with the Laschamp geomagnetic excursion circa 41,000 years ago.

1.1 Cosmogenic radionuclides and the Laschamp geomagnetic excursion

Cosmogenic radionuclides, such as ^{10}Be and ^{14}C, are formed in the Earth's upper atmosphere through the interaction of incoming high-energy cosmic rays with target nuclides (Lal and Peters, 1967). The cosmic ray flux is modulated by both the shielding effect of the Earth's magnetic field and the solar-induced interplanetary magnetic field (the 'solar wind'). The lower the strength of either the geomagnetic field or solar wind, the deflection of incoming cosmic rays is reduced, and the production of cosmogenic radionuclides is therefore greater (Elsasser et al., 1956).

The geomagnetic field exhibits long-term secular variation, including major reversals of the Earth's magnetic (dipole) field between normal and reversed configurations, which occur during periods of progressive decay in the Earth's dipole moment (Cox, 1969; Valet and Meynadier, 1993). Additionally, shorter-term ($<10^4$ years) 'stability crises' occur whereby the intensity of the geomagnetic field decreases more or less dramatically, but the field does not undergo a long-term reversal. These may coincide with geomagnetic excursions – periods of distorted dipole geometry when the virtual geomagnetic poles (VGPs) move away from the area of normal high-latitude secular variation – or even short-term (10^2-10^3 years) complete reversals (where VGPs temporarily migrate to higher latitudes of the opposite hemisphere) (Nowaczyk et al., 2012). The most prominent of these geomagnetic excursions over the past 100,000 years is known as the 'Laschamp event', dated to circa 41,000 years ago (Bonhommet and Babkine, 1967; Guillou et al., 2004; Singer et al., 2009). This event is characterised by a short-term full reversal of the geomagnetic field (Nowaczyk et al., 2012) and the lowest geomagnetic field intensities of the past 100,000 years, falling to approximately 10% of today's value (Laj et al., 2000; Nowaczyk et al., 2013).

Such geomagnetic events can provide global, temporally synchronous signals in palaeoenvironmental archives, observable directly in records of relative palaeointensity, as well as in records of cosmogenic nuclides (including ^{10}Be and ^{14}C). Thus, it is theoretically possible to link palaeoenvironmental archives using these isochronous signals (Brauer et al., 2014). ^{10}Be has a short (1-2 year) atmospheric residence time (McHargue and Damon, 1991), providing an excellent record of past cosmogenic nuclide production variation, and has been measured directly in the Greenland ice-cores (Yiou et al., 1997; Muscheler et al. 2004) (unlike ^{14}C, which is too low in abundance to detect within the ice). ^{14}C provides a less direct production marker, however, because of its incorporation into the global carbon cycle system and consequent exchanges between the global carbon reservoirs, thus complicating the intercomparison of such records.

1.2 The Greenland ice-core chronology

The Greenland ice-core chronology 'GICC05' is the most recent timescale applied to the Greenland ice-cores, tying together the GRIP (Johnsen et al. 1992), GISP2 (Grootes et al., 1993), NGRIP (NGRIP members, 2004) and NEEM (NEEM Community Members, 2013) records (Seierstad et al., 2014). For the entire time period covered by the ^{14}C dating technique, i.e., the last circa 50,000 years, GICC05 is based on direct counting of the annual layers within the ice (Andersen et al., 2006; Rasmussen et al., 2006; Svensson et al., 2008; Brauer et al., 2014). The uncertainty on the timescale is based upon the 'maximum counting error' (MCE) concept, whereby each uncertain layer is counted as ½ ± ½ year and added linearly. Thus, throughout the Last Glacial period, the MCE on GICC05 amounts to approximately 5%. It should be noted that GICC05 uses the notation 'b2k' – i.e., 'calendar years before its datum, AD 2000' – whereas herein we convert this to years 'BP' (before present, AD 1950), enabling more direct comparison with the Hulu Cave uranium (U-)series and IntCal13 timescales (below).

Since its introduction in 2005, GICC05 has now been utilised for over a decade, demonstrating the robustness of the chronology, though there have been recent suggestions of small scale errors. For example, Sigl et al. (2015) presented evidence, making use of the distinctive 'AD 775 and 994 events' recorded as both ^{10}Be and ^{14}C production spikes, as well as using tephra marker horizons, that the ice-core chronology is 7 years too old by the late first millennium AD. Over a longer time range, Buizert et al. (2015) presented evidence that GICC05, on average, misses 6.3 out of every 1,000 annual layers. This conclusion is based upon comparison of the respective oxygen isotope (δ^{18}O) records of the NGRIP ice-core and Hulu Cave (China) speleothem, which is independently U-series dated. However, this comparison of δ^{18}O records assumes synchroneity of the respective palaeoclimatic signals – an assumption that may not necessarily hold true (Lane et al., 2013; Brauer et al., 2014).

1.3 Radiocarbon dating and the IntCal timescale

In order to generate meaningful ages from the ^{14}C dating method, a calibration stage is required since the concentration of ^{14}C (relative to stable ^{12}C and ^{13}C) in the environment changes through time. This is the result of both the variations in production rate (Lal and Peters, 1967) outlined above and carbon cycle effects, which alter the global distribution of relatively older or younger carbon sources between the respective reservoirs of Earth's carbon cycle system through time (Broecker et al., 1960; Siegenthaler et al., 1980).

Calibration involves the comparison of samples' raw isotopic measurements with the internationally ratified, consensus radiocarbon calibration curve 'IntCal13' (Reimer et al., 2013), which itself is comprised of 'known age' material from a variety of palaeo-archives. For the last ~12,500 years, the IntCal curve is composed of independently dendro-chronologically dated wood. Previous research (Muscheler et al., 2004, 2008, 2014a; Adolphi and Muscheler, 2016) has utilised this high-resolution, continuous record of past variation in atmospheric ^{14}C concentrations ($\Delta^{14}C$) to tie this most recent period of the IntCal timescale to the ^{10}Be signal in Greenland. These authors found an offset of approximately 65 years between GICC05 and IntCal during the Preboreal (i.e. circa 12,500 to 10,000 years ago), with GICC05 seemingly including a small over-count – an offset consistent in scale with that proposed by Sigl et al. (2015). Since this latest portion of the ^{14}C calibration curve is composed of robustly dendrochronologically dated records, Muscheler et al. (2008) attributed this 65 year offset to uncertainties in the ice-core layer counting.

Further back in time, through to the methodological limit of radiocarbon dating (circa 50,000 years ago), however, the ^{14}C calibration curve is less certain. The central archive for this earlier period is that provided by plant macrofossils picked from the annually laminated sediments of Lake Suigetsu, Japan (Staff et al., 2011; Bronk Ramsey et al., 2012). Additional data are provided by speleothems (Hoffmann et al., 2010; Southon et al., 2012), marine corals (e.g. Fairbanks et al., 2005), and foraminifera from marine sediment cores (e.g. Hughen et al., 2006), all of which incorporate (marine- or dead carbon) ‘reservoir effects’ that require correction and thereby introduce additional uncertainties. These reservoir effects would also be expected to ‘smooth’ the atmospheric $\Delta^{14}C$ signal, making comparison to ^{10}Be records more complicated. Unlike these latter records, the Lake Suigetsu data provide a direct record of atmospheric $\Delta^{14}C$, and have previously been used to compare to both records of palaeomagnetic intensity (e.g. Nowaczyk et al., 2013) and to ^{10}Be in the Greenland ice-cores (e.g. Bronk Ramsey et al., 2012; Muscheler et al., 2014b). However, the Lake Suigetsu data are necessarily discontinuous – limited by the stochastic finds of plant macrofossil remains in the sediment profile – as well as being potentially less reliable due to the methodological problems associated with dating such small samples close to the radiocarbon detection limit (Muscheler et al., 2014b). As with the Greenland ice-cores (above), the Lake Suigetsu dataset also has relatively large cumulative counting uncertainties by ~40,000 years BP.

The promise of more reliable, continuous data for this older time period comes from floating tree-ring sequences, most notably long-lived New Zealand kauri (*Agathis australis*) (Turney et al., 2010, 2016; Hogg et al., 2013). Such records are limited in duration, however, by the up to ~2,000 year life-spans of individual trees, limiting their utility for comparison to the Greenland ^{10}Be record to relatively short periods of time (Muscheler et al., 2014b; Turney et al., 2016). Recently, Muscheler et al. (2014b) presented such a comparison, arguing that the Greenland ice-core and ^{14}C (IntCal) timescales were discordant circa 40,000 years ago, with the calibrated ^{14}C timescale apparently 1,200 years too old. This would be a highly significant finding, if true, since it compromises the inter-comparison of ^{14}C-dated palaeoenvironmental records with those dated by other methods. It also directly affects the interpretation of ^{14}C data through this time period across other disciplines, such as archaeological applications, with the replacement of Neanderthals by anatomically modern humans in Europe around this time (Higham et al., 2014). However, the study of Muscheler et al. (2014b) was necessarily limited to a short record (1,350 years), minimising the Δ^{14}C structure that could be compared with the equivalent ^{10}Be-inferred signal, and thereby reducing the reliability of the correlation drawn. Recently, Cheng et al. (2018) have provided an extended record from Hulu Cave (China) based upon radiocarbon data from two new speleothems (‘MSD’ and ‘MSL’), adding to the previously published dataset of Southon et al. (2012) from speleothem ‘H82’ which covered the period ~10.7 to 26.9 ka BP. As with the Lake Suigetsu dataset (above), this new Hulu Cave record now extends across the entirety of the radiocarbon dating method. The latter has the advantage of a highly precise U-series derived calendar age scale, and will provide the central archive of the next iteration of the consensus calibration curve, IntCal (Reimer et al., in prep., Radiocarbon).

As noted above, speleothems incorporate a reservoir effect, which requires correction, and therefore introduces further uncertainty into the ^{14}C values. Southon et al. (2012) and Cheng et al. (2018) both describe the “unusually small and stable” (450 ± 70 ^{14}C years) ‘dead carbon fraction’ (DCF) registered in these Hulu Cave speleothems, which makes them particularly attractive for radiocarbon calibration purposes. However, it would seem that this small and stable DCF is a result of the unique geological setting of the site, such that the ‘inbuilt age’ recorded by the speleothem dripwater is more of a ‘soil reservoir effect’, rather than a DCF, *sensu stricto*. The consequence of this is the favourable low and stable ‘DCF’; however, the pay-off is that the atmospheric ^{14}C signal is effectively smoothed at this resolution (~450 years), meaning that higher frequency signal is consequently lost.

Thus, there are both strengths and weaknesses in all of the aforementioned calibration records. To add to this current state of knowledge, therefore, we herein exploit new ^{14}C data from a continuous peat sequence from Greece, extending over a significantly longer time period (circa 47,300 to 39,600 cal. BP) than the kauri dataset utilised by Muscheler et al. (2014b). This enables us to use the entirety of the Δ^{14}C signal associated with the build-up to- and peak of the Laschamp excursion to enable more robust comparison of the calibrated ^{14}C and Greenland ice-core time scales. Our dataset also provides a direct record of atmospheric ^{14}C concentration, unlike the Hulu Cave speleothems, and provides continuous material for ^{14}C dating, unlike the stochastic Lake Suigetsu dataset, without the issues of small sample sizes associated with the latter record. The drawback of our new dataset, however, is the lack of independent chronology, which we necessarily need to obtain through Bayesian statistical modelling (section 3.2, below).

2. Study site

Tenaghi Philippon is situated in the Philippi peatland within the Drama Basin of NE Greece (Fig. 1). Since its discovery and initial exploitation in the 1960s, the site has become widely recognised as harbouring one of the best terrestrial archives of Quaternary climatic and environmental change in Europe (Wijmstra, 1969; Tzedakis et al., 2006; Pross et al., 2015 and refs. therein). Scientific drilling campaigns at the site have yielded a peat-dominated sequence that extends to a depth of nearly 200 m and covers the last ~1.35 Ma continuously. This sequence represents an extremely sensitive recorder of rapid climatic change both during glacial and interglacial boundary conditions, which is ascribed to the site’s intermediate position between higher-latitude (i.e., North Atlantic Oscillation- and Siberian Highinfluenced) and lower-latitude (monsoonally influenced) climatic regimes, its intramontane setting, and its proximity to the glacial refugia of thermophilous plant taxa (Pross et al., 2009, 2015).

In 2005, a new, 60 m long core (‘TP-2005’; 40°58’24” N, 24°13’26” E, 40 m asl) was recovered from Tenaghi Philippon (Pross et al., 2007). The core consists primarily of fen peat and is believed to represent continuous accumulation throughout the last circa 310 kyrs (Fletcher et al., 2013). A previous study (Müller et al., 2011) presented 20 accelerator mass spectrometry (AMS) ^{14}C dates, spanning the majority of the approximately 50,000 year ^{14}C dating time period, from the uppermost 15.28 m of the TP-2005 core (Table S1). Additionally, three tephra layers have been identified at 7.61 m, 9.70 m and 12.64-12.87 m core depths, and respectively geochemically correlated to the Y-2 tephra (resulting from the Cape Riva eruption of Santorini), Y-3 tephra (resulting from an eruption from the Campi Flegrei), and Y-5 tephra (from the regionally widespread Campanian Ignimbrite eruption, also from the Campi Flegrei) (Müller et al., 2011; Albert et al., 2015 Pross et al., 2015; Wulf et al., 2018). Accompanying palaeoenvironmental data are provided by a centennial resolution pollen record spanning Marine Isotope Stages (MIS) 4 to 2 (Müller et al., 2011).

3. Methods

3.1 Radiocarbon dating

Contiguous peat sub-samples (maximum 5 cm thick) from Tenaghi Philippon core 'TP2005' were taken from 12.87 to 14.80 cm depth – i.e. spanning the time period immediately preceding the deposition of the visible Campanian Ignimbrite (C.I.) tephra, back to the methodological limit of ^{14}C dating (circa 50,000 cal. BP). Each sub-sample was physically homogenised prior to a standard acid-base-acid (ABA) chemical pre-treatment for radiocarbon dating, following the method of Brock et al. (2010). The three main stages of this process (successive acid-, base-, and acid washes) are similar across most radiocarbon laboratories and are respectively intended to remove: (i) sedimentary- and other carbonate contaminants; (ii) organic (principally humic- and fulvic-) acid contaminants; and (iii) any dissolved atmospheric CO_2 that might have been absorbed during the preceding base wash. In this way, any potential secondary carbon contamination is removed, leaving the samples pure for subsequent combustion, graphitisation and accelerator mass spectrometry (AMS) ^{14}C dating. At the Oxford Radiocarbon Accelerator Unit (ORAU) ABA chemical pre-treatment of peat samples (laboratory pre-treatment code 'VV') involves successive 1 M HCl (20 mins, 80 °C), 0.2 M NaOH (20 mins, 80 °C) and 1 M HCl (1 hr, 80 °C) washes, with each stage followed by rinsing (≥3 times) with ultrapure MilliQ™ deionised water. From five samples, the base-soluble humic acid component extracted from the peat was additionally dated to provide supporting information on the likely contribution of mobile- (presumably, downward-percolating young-) contaminant to the primary base-insoluble ('humin') component of the peat samples. Specifically, this involved the collection of the base-soluble fraction of these samples and reacidification through the addition of 1 M HCl, followed by centrifugation and rinsing (twice) with ultrapure MilliQ™ deionised water (ORAU laboratory pre-treatment code 'HW'). AMS ^{14}C dating was subsequently performed on the 2.5 MV HVEE tandem AMS system at ORAU (Bronk Ramsey et al., 2004; Staff et al., 2014).

3.2 Chronological modelling

The TP-2005 ^{14}C data were analysed with the Bayesian statistical software OxCal ver. 4.3 (Bronk Ramsey, 2019), implementing a Poisson-process ('`P_Sequence`') deposition model (Bronk Ramsey, 2008). The `P_Sequence` model takes into account the complexity (randomness) of the underlying peat accumulation process, and thus provides the most realistic age-depth model for the TP-2005 peat profile on the calibrated radiocarbon timescale. For comparison purposes, we herein modelled our TP-2005 ^{14}C data on to both the recently published Hulu Cave dataset of Cheng et al. (2018), as well as the current consensus (IntCal) calibration curve (Reimer et al., 2013). The rigidity of each `P_Sequence` (i.e., the regularity of the peat accumulation rate) is determined iteratively within OxCal through a model averaging approach, based upon the likelihood (i.e., calibrated ^{14}C) data included within the model (Bronk Ramsey and Lee, 2013). '`Boundary`' functions were applied at the top and bottom of the '`P_Sequence`' (at 12.87 m and 14.80 m core depth, respectively) – the former providing a modelled, ^{14}C-derived age for the C.I. tephra. Objective outlier analysis was applied to down-weight any statistically anomalous data points (Bronk Ramsey, 2009; Bronk

Ramsey et al., 2010). An 'r-type' `Outlier_Model` was selected, allowing for short-term fluctuations in the ^{14}C concentrations between the respective radiocarbon reservoirs of the Tenaghi Philippon, Hulu Cave and IntCal13 datasets. (N.b., a premise of this paper is that the IntCal and Hulu Cave curves currently smooth out real, higher frequency 'wiggles' in atmospheric radiocarbon concentration, □^{14}C – i.e., that the datasets have short-term offsets in their apparent ^{14}C concentrations compared to the TP-2005 record – which is allowed for by the r-type `Outlier_Model`.) A prior '`Outlier`' probability of 5% was applied to all of the TP-2005 ^{14}C determinations, since there was no reason, *a priori*, to believe that any samples were more likely to be statistical outliers than others. As noted, both the Hulu Cave (Cheng et al., 2018) and IntCal13 ^{14}C calibration curve (Reimer et al., 2013) were used, with alternative comparison datasets from Lake Suigetsu (Bronk Ramsey et al, 2012), Bahamas speleothem (Hoffmann et al., 2010), and Cariaco Basin foraminifera (Hughen et al., 2006) plotted for comparison purposes only. The coding of these primary deposition models and the model output are given in the Supplementary Material (S1 and Tables S3 and S4).

Similar Poisson-process modelling was applied to the original TP-2005 ^{14}C determinations of Müller et al. (2011), using two successive `P_Sequences` for the lower and upper core sections, cross-referencing the upper `Boundary` of the lower `P_Sequence` (12.87m core depth; the lower contact of the C.I. tephra) to equal the lower `Boundary` of the upper `P_Sequence` (12.64m core depth; the upper contact of the C.I. tephra). Again, the model coding is given in the Supplementary Material (S2).

One consideration with the `P_Sequence` deposition model is that it produces an inevitable attenuation of the authentic □[1]C maxima and minima by 'pulling' the data to more

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[1] C production rates using the production rate model of Herbst et al. (2017) and the Local Interstellar Spectrum of Potgieter et al. (2014), assuming a constant solar modulation potential

closely fit the Hulu Cave or IntCal calibration datasets. Therefore, supporting age-depth models were subsequently generated in OxCal, simply applying a uniform ('U_Sequence') deposition model (Bronk Ramsey, 2008), rather than the P_Sequence. The coding of these supporting deposition models is also given in the Supplementary Material (S3), as is the model output (Tables S3 and S4). In reality, the two differing model assumptions (P_Sequence or U_Sequence) produce similar output (Figs. S1 and S2), reflecting the insensitivity of our conclusions presented herein to the choice of chronological model construction.

3.3. $\Delta^{14}C$ modelling from GRIP ^{10}Be fluxes and Black Sea and GLOPIS-75 VADM

GRIP ^{10}Be fluxes (Yiou et al., 1997; Muscheler et al., 2004) and estimates of the Earth's virtual axial dipole moment (VADM) from both the individual Black Sea record (Nowaczyk et al., 2012, 2013) and the GLOPIS-75 stack (Laj et al., 2004, 2014) were converted into $\Delta^{14}C$ using previously applied methods (Muscheler et al., 2004, 2005). First, VADM was converted into (~300‰ between 48,000 and 40,000 cal.BP), we ran the carbon cycle model with slightly reduced ocean diffusivity (70% of the preindustrial value, resembling reduced ocean ventilation

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of 800 MeV that resembles the modern average solar activity (Muscheler et al., 2016). In a second step, $\Delta^{14}C$ was modelled from GRIP ^{10}Be fluxes and VADM-based ^{14}C production rates using a box-diffusion carbon cycle model (Siegenthaler et al., 1980; Muscheler et al., 2004). We assume a $^{10}Be/^{14}C$ production rate ratio of 1:1 which is in agreement with $^{10}Be/^{14}C$ comparisons from the Holocene (Adolphi and Muscheler, 2016) as well as production rate models (Herbst et al., 2017). To match the amplitude of the overall $\Delta^{14}C$ increase in IntCal in the Glacial) and reduced air/sea exchange rates (75% of the preindustrial value, resembling increased sea ice extent). Note, that this only affects the overall amplitude of the modelled Δ^{14}C change, but not the shape of the curve, since these parameters were kept constant over the entire timeframe.

4. Results

Our 54 new ^{14}C determinations from TP-2005 are presented in Table S2 and, having been modelled against both the Hulu Cave dataset and IntCal13 (see section 3.2, above), are plotted against depth in Fig. 2. These new data suggest that the previous ^{14}C-based chronology of Müller et al. (2011) underestimated the true age of the peat sequence for the time period before circa 39,000 cal. BP; this may be due to insufficient chemical pre-treatment to remove (young/modern) contaminant carbon, which has an increasing influence on ^{14}C measurements with increasing age.

The inferred Δ^{14}C values from our new TP-2005 data show three periods of increasing Δ^{14}C values (Fig. 3). On the Hulu Cave U-series timescale these successive increases occur from circa 47,300 cal. BP to 45,600 cal. BP, reaching a maximum of approximately 450‰; from circa 44,900 cal. BP to 43,700 cal. BP, reaching a maximum of approximately 400‰; and from circa 43,200 cal. BP to 42,000 cal. BP, reaching a maximum of approximately 650‰. This final elevation represents the peak of the Laschamp geomagnetic excursion in TP-2005, and continues until at least the timing of the Campanian Ignimbrite (C.I.) tephra, dated to circa 39,600 cal. BP (Fig. 4), interrupted by a(t least one) depression in Δ^{14}C values between circa 41,000 and 40,400 cal. BP.

5. Discussion

An initial observation is that our new TP-2005 data provide no evidence for the extremely high □^{14}C values associated with the Laschamp geomagnetic excursion that have been suggested by some previous studies (e.g. Voelker et al., 2000; Hughen et al., 2006; Hajdas et al., 2011). There are also no data identified as being statistical outliers (Bronk Ramsey, 2009; Bronk Ramsey et al., 2010), demonstrating the integrity of the peat sequence both for reconstructing past variation in □^{14}C as well as for palaeoenvironmental research. We note that the age-depth profile for TP-2005 is more linear (especially at the younger end) when modelled on to the Hulu Cave dataset rather than the IntCal13 curve (Fig. 2), which implies greater congruence of the TP-2005 ^{14}C data with the Hulu Cave record (Cheng et al., 2018) rather than IntCal13 (Reimer et al., 2013).

Our new data (Fig. 3) show higher frequency □^{14}C variability than the ‘smoothed’ IntCal13 curve, which inevitably loses authentic signal when the contributing ^{14}C datasets are averaged into the consensus curve (Reimer et al., 2013; Fig. S4). The ^{14}C data from the two individual, non-reservoir corrected atmospheric ^{14}C datasets (TP-2005 and Lake Suigetsu) match each other within the bounds of statistical uncertainty. The Lake Suigetsu dataset shows higher frequency variability, however. One reason for this is the ~150 year smoothing of the TP-2005 data (due to the contiguous sub-sampling methodology applied), as compared to the annual signal contained within the individual Japanese terrestrial plant macrofossil samples. The other reason is the statistical ‘noise’ in the Lake Suigetsu data, which is the result of the methodological problems of dating very small individual plant macrofossil samples so close to the limit of ^{14}C detection (Muscheler et al., 2014b). For this latter reason, we prefer the TP2005 dataset (as compared to the Lake Suigetsu record) as more reliably representing the authentic signal in past variability of atmospheric radiocarbon concentration for this earliest portion of the ^{14}C time frame. We also reiterate that our TP-2005 data demonstrate a direct atmospheric signal, therefore avoiding the additional uncertainties associated with the reservoir effects of either the marine or speleothem datasets. The tripartite structure seen in the Tenaghi Philippon data also demonstrates higher amplitude shifts in the build-up to the principal Laschamp peak than the Hulu Cave dataset. As noted above (section 1.3), we suggest that this attenuation in the Hulu Cave record is the result of the longer ~450 year smoothing effect of the soil reservoir effect at the site.

However, since the TP-2005 data have necessarily been modelled on to the Hulu Cave and IntCal13 timescales (see section 3.2), such errors currently contained within these calibration datasets will propagate through into the placement of our TP-2005 data in calendar time and hence on the amplitude of the reconstructed Δ^{14}C. That said, the general shape of the Δ^{14}C data will be largely unaffected by this process and, consequently, we can compare TP2005 Δ^{14}C to the equivalent signal inferred from Greenland ^{10}Be to assess the concordance (or lack thereof) between the underlying Hulu Cave (U-series), IntCal, and Greenland ice-core (GICC05) timescales themselves.

Significantly, the general shape of the TP-2005 Δ^{14}C data, consisting of three successive rises in atmospheric ^{14}C concentration in the ~6,000 years leading up to the peak values associated with the Laschamp geomagnetic excursion (from circa 42,000 cal. BP in the TP2005 record), broadly tracks equivalent increases calculated from ^{10}Be flux measured in the GRIP ice-core (Yiou et al., 1997; Muscheler et al., 2004, 2014b) (Fig. 3). This is the first time that this clear, tripartite structure in Δ^{14}C has been directly observed in the build-up to the Laschamp excursion.

We can additionally compare our record with estimates of the Earth's dipole moment (virtual axial dipole moment, VADM) obtained from relative palaeointensity studies, to provide assessment of the role of the geomagnetic field in contributing to cosmogenic radionuclide production. To this end, we utilise both the Black Sea sediment record of Nowaczyk et al. (2013), drilled ~1000 km East of Tenaghi Philippon, as well as the GLOPIS75 globally-averaged curve (Laj et al., 2004, 2014). The Black Sea dataset is not truly independent, in that it has been tuned to the GICC05 timescale using palaeoenvironmental proxy data from the two archives (Nowaczyk et al., 2012). Likewise, the GLOPIS-75 dataset is composed of records aligned on to a single timescale (Laj et al., 2004, 2014). However, the inferred $\Delta^{14}C$ from both of these records closely mimics the variations evident in the Greenland ^{10}Be-inferred $\Delta^{14}C$ in both structure and amplitude, and also shares similar characteristics with the TP-2005 $\Delta^{14}C$ data from Tenaghi Philippon (Fig. 3).

Despite this general coherence in the $\Delta^{14}C$, ^{10}Be, and palaeomagnetic intensity records, there are also distinct differences evident. Firstly, the amplitude of the successive $\Delta^{14}C$ increases is vastly different in the ^{10}Be and VADM-inferred data, as compared to the TP-2005 dataset. And, whilst ^{10}Be and VADM indicate that the first two $\Delta^{14}C$ increases are about a factor of three smaller than the final rise circa 42,500 to 40,000 cal. BP, the initial two $\Delta^{14}C$ maxima in TP-2005 (as modelled on to the Hulu Cave dataset) are approximately ⅔ the amplitude of the final Laschamp peak (Fig. 3d). When instead modelled on to IntCal13 (Fig. 3e), the TP-2005 data show a comparable magnitude $\Delta^{14}C$ increase for all three steps, which clearly accords less well with the ^{10}Be and VADM-inferred signals. This is another line of argument in support of the Hulu Cave dataset as providing the more accurate $\Delta^{14}C$ record through this time interval compared to the current consensus calibration curve (IntCal13).

In terms of timing, the earliest □^{14}C maximum (circa 45,600 cal. BP) in TP-2005, as modelled on to the Hulu Cave U-series timescale, is represented by concomitant increases in both the Greenland ^{10}Be and Black Sea palaeointensity-inferred □^{14}C records. However, the second □^{14}C maximum (circa ~~46,000~~ 43,700 cal. BP) does not demonstrate such a correlation to the ^{10}Be or VADM-inferred records. Conversely, the third and final increase in TP-2005 □^{14}C to the principal Laschamp peak does appear similar in structure to the ^{10}Be and VADMinferred records, with an interruption to the rising □^{14}C trend circa 42,800 cal. BP evident in all of the records, before a resumption of increasing values up to the principal Laschamp production maximum. Again, we see a better fit of our TP-2005 data against these alternative ^{10}Be and palaeointensity-inferred □^{14}C records when modelled on to the Hulu Cave dataset rather than IntCal13 (Fig. 3). This is likely due to the IntCal13 curve containing incorrect structure, particularly around the timing of the principal Laschamp peak itself. This is unsurprising since the constituent datasets of IntCal13 are themselves in disagreement at this time (Fig. S4). It would appear that the DCF of the independently U-series dated Bahamas speleothem record (Hoffmann et al., 2010) is being over-corrected at this time. Conversely, the □^{14}C of the Cariaco Basin dataset (Hughen et al., 2006) appears too high, and it is likely that errors in either the marine reservoir correction or, more likely, the climatically wiggle-matched timescale of this latter record is responsible for the erroneous structure in IntCal at this time.

As noted above, the second maximum in the TP-2005 □^{14}C data circa 43,700 cal. BP is not represented by equivalent signal in the ^{10}Be or VADM-inferred datasets. We therefore hypothesise that the signal evident in the direct (TP-2005) □^{14}C record at this time is the result of processes internal to the global carbon cycle. We note that, as with all such radiocarbon calibration datasets, firm conclusions should not be drawn until corroboration is provided from further archives. Such support is provided for the subsequent □^{14}C minimum, however, with an equivalent minimum seen in the New Zealand kauri record of Turney et al. (2010; which was also utilised by Muscheler et al. 2014b) when that record is also modelled on to the Hulu Cave dataset. Interestingly, a similar interruption to the longer-term □^{14}C increase to the principal Laschamp □^{14}C maximum is also seen in both the TP-2005 and kauri records circa 42,800 cal BP, providing further corroboration for the authenticity of this signal.

One further difference between the structure of the TP-2005 and Greenland ^{10}Be-inferred □^{14}C occurs in the aftermath of the principal Laschamp peak. Whereas the ^{10}Be data show a steady decline from circa 41,000 to 39,000 cal. BP, the TP-2005 □^{14}C data exhibit an initial, equivalent decline (which is not seen in the Hulu Cave or IntCal13 datasets; Fig. S4), but then return to higher □^{14}C values again at around 40,200 cal. BP. Similar structure is hinted at in the Lake Suigetsu record; however, it remains unclear as to how much of the higher frequency signal in the Suigetsu record is genuine and how much is noise. The lack of a comparable signal in the ^{10}Be flux suggests that, if genuine, this □^{14}C signal would also be related to processes internal to Earth's carbon cycle. Even more speculatively, we note the approximate coincidence of this return to higher □^{14}C with Heinrich Stadial 4, during which the Atlantic Meridional Overturning Circulation (AMOC) is believed to have been significantly reduced in strength (Böhm et al., 2015; Eggleston et al., 2016). The AMOC reduction would have less efficiently removed relatively ^{14}C-enriched CO_2 from the atmosphere and less efficiently returned relatively ^{14}C-depleted CO_2 from the deep ocean. Therefore, there is a theoretical expectation that □^{14}C would increase at about this time, which would not be seen in the ^{10}Be and VADMinferred records. The afore-mentioned period of divergence in □^{14}C and ^{10}Be circa 43,800 cal.

BP does not coincide with a Heinrich Stadial, but it does coincide with a 'non-Heinrich' Stadial (Greenland Stadial 12), which we again speculate as being related to the signal seen in TP2005 $\Delta^{14}C$.

With regard to the alignment of the palaeointensity and TP-2005 $\Delta^{14}C$ signals, the Black Sea and Tenaghi Philippon datasets can be unambiguously synchronised at the younger end of the TP-2005 data via the presence of the C.I. isochron in both records. Our TP-2005 ^{14}C-derived age of 39,556 ± 310 cal. BP for the C.I. (as modelled on to the Hulu Cave timescale; 39,87739,165 cal. BP, 95.4% highest probability density range; Fig. S5) is within statistical agreement (at 95.4% confidence) with the GICC05-implied age in the Black Sea record of 39,350 years BP (Nowaczyk et al. 2012, 2013), providing additional support for the alignment of our TP2005 dataset with the Black Sea record at this point in time. We further note the statistical agreement between our TP-2005 inferred age for the C.I. (at 95.4% confidence) with both the widely quoted $^{40}Ar/^{39}Ar$ age of 39,230 ± 110 years BP (2σ) presented by De Vivo et al. (2001) and the more recently published $^{40}Ar/^{39}Ar$ age of 39,850 ± 140 years BP (2σ) given by Giaccio et al. (2017), noting that our TP-2005 inferred age falls centrally between these two $^{40}Ar/^{39}Ar$ age estimates. Significantly, our TP-2005 inferred age for the C.I. on the IntCal13 timescale (38,725 ± 239 cal. BP; Fig. S5) is too young compared to these alternative age estimates (by ~1,100 years as compared to the Giaccio et al. 2017 $^{40}Ar/^{39}Ar$ age). This provides further support for the key finding above that IntCal13 is not accurate circa 40,000 years ago, and that the Hulu Cave speleothem provides a better representation of the authentic radiocarbon calibration curve at this point in time.

6. Conclusions

We have presented a record of atmospheric radiocarbon concentration (Δ^{14}C) from Tenaghi Philippon core TP-2005 that provides a unique, continuous and direct (non-reservoir corrected) record of Δ^{14}C for the earliest ~10,000 years of the ^{14}C dating method. Our data demonstrate higher frequency variability than the smoothed IntCal13 consensus calibration curve (Reimer et al., 2013) or the recently published Hulu Cave speleothem dataset (Cheng et al. 2018), yet lack the noise of the Lake Suigetsu dataset (Bronk Ramsey et al., 2012) or the additional reservoir uncertainties of the marine (Fairbanks et al., 2005; Hughen et al., 2006) and speleothem (Hoffmann et al., 2010; Cheng et al. 2018) datasets. Thus, we have been able to compare Δ^{14}C with the shared cosmogenic production signal of ^{10}Be in the Greenland ice cores and direct palaeo-magnetic intensity records from the Black Sea (Nowaczyk et al., 2013) and the GLOPIS-75 stack (Laj et al., 2004, 2014). These datasets demonstrate a similar pattern in the build up to and through the principal peak of the Laschamp geomagnetic excursion. By placing our ^{14}C dataset on to both the Hulu Cave U-series and IntCal13 timescales via Bayesian statistical modelling, the comparison of our TP-2005 Δ^{14}C dataset with these alternative records also implicitly relates the underlying U-series, IntCal13 and GICC05 timescales themselves. We suggest that, whilst the timescales are in broad agreement, the TP-2005 Δ^{14}C data match the Greenland ^{10}Be-inferred data more closely when modelled on to the Hulu Cave dataset rather than the IntCal13 curve. This suggests that there is erroneous structure currently included within the IntCal curve, which will be significantly improved upon with the addition of the Hulu Cave dataset to the upcoming iteration of the IntCal calibration curve. It is unsurprising that we would find erroneous structure within IntCal13 given that the underlying, contributing ^{14}C datasets to IntCal are themselves in significant disagreement with each other at this time, and we deem it most likely that the main error is incorporated from the climatically wigglematched timescale of the Cariaco Basin dataset. Our TP-2005 data also suggest that there is missing structure from the smoothed IntCal and Hulu Cave curves between circa 47,000 cal. BP and 43,000 cal. BP. Thus, we provide a revised approximation of the authentic structure of the radiocarbon calibration curve for the earliest ~10,000 years of the ^{14}C dating method, which will have implications for all users of the technique over this time period.

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Figure Captions:

Fig. 1. Location of the Tenaghi Philippon site, Eastern Macedonia, NE Greece. Inset shows the location of sediment core TP-2005 within the Drama Basin.

Fig. 2. Revised age-depth profile (green) for core TP-2005 from Tenaghi Philippon, as compared to the previously published dataset of Müller et al. (2011; red), generated by independent `P_Sequence` deposition modelling in OxCal ver.4.3 (Bronk Ramsey, 2008, 2019; Bronk Ramsey and Lee, 2013) on to the Hulu Cave ^{14}C calibration dataset of Cheng et al. (2018). Equivalent age-depth profiles are additionally plotted for the same TP-2005 datasets (this study, blue; and Müller et al., 2011, grey) as modelled on to the IntCal13 calibration curve (Reimer et al., 2013). Modelled probability density functions are plotted with the 68.2% highest probability density range interpolations overlain. For the unmodelled data, see Supplementary Figure S3.

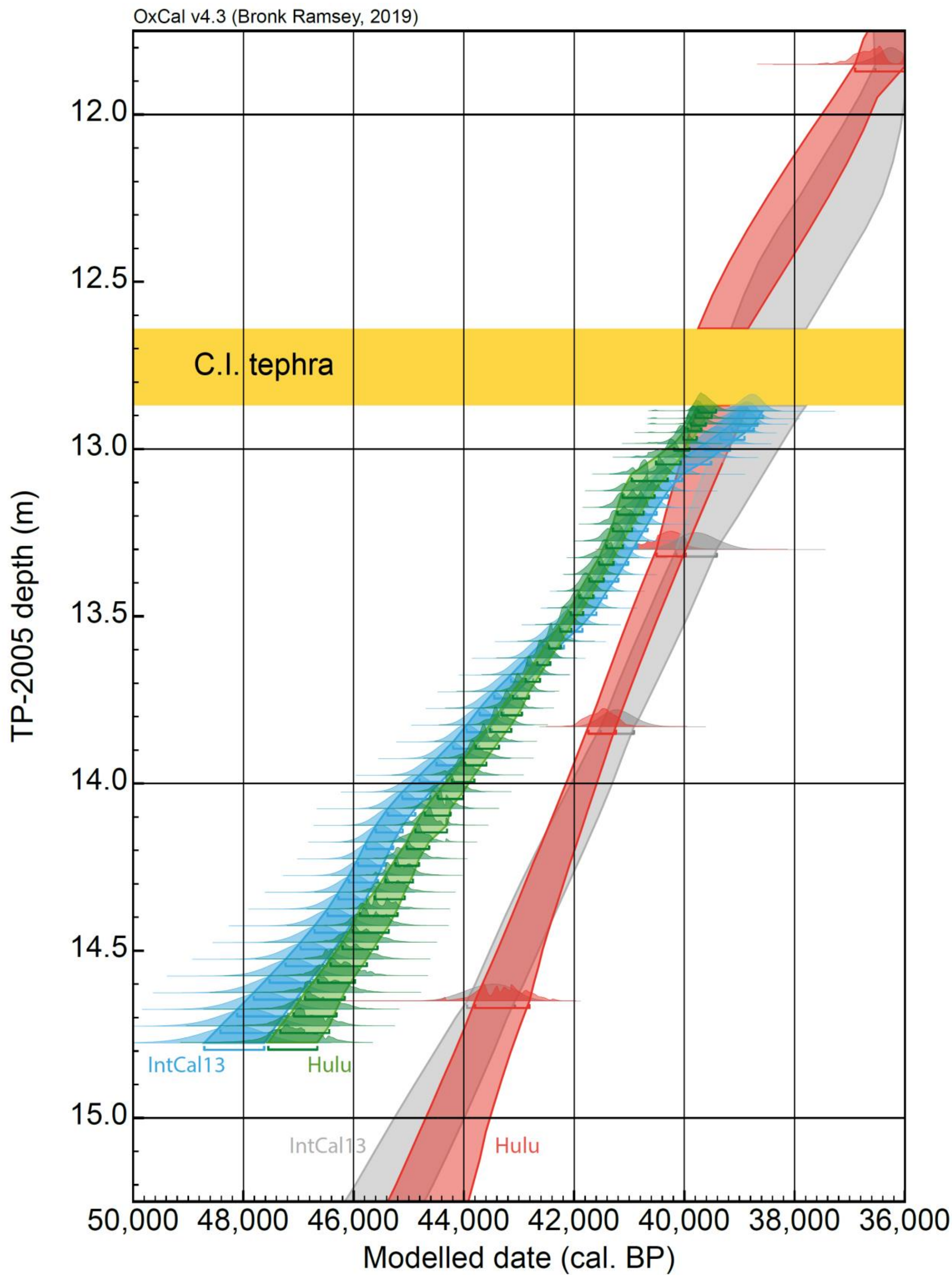
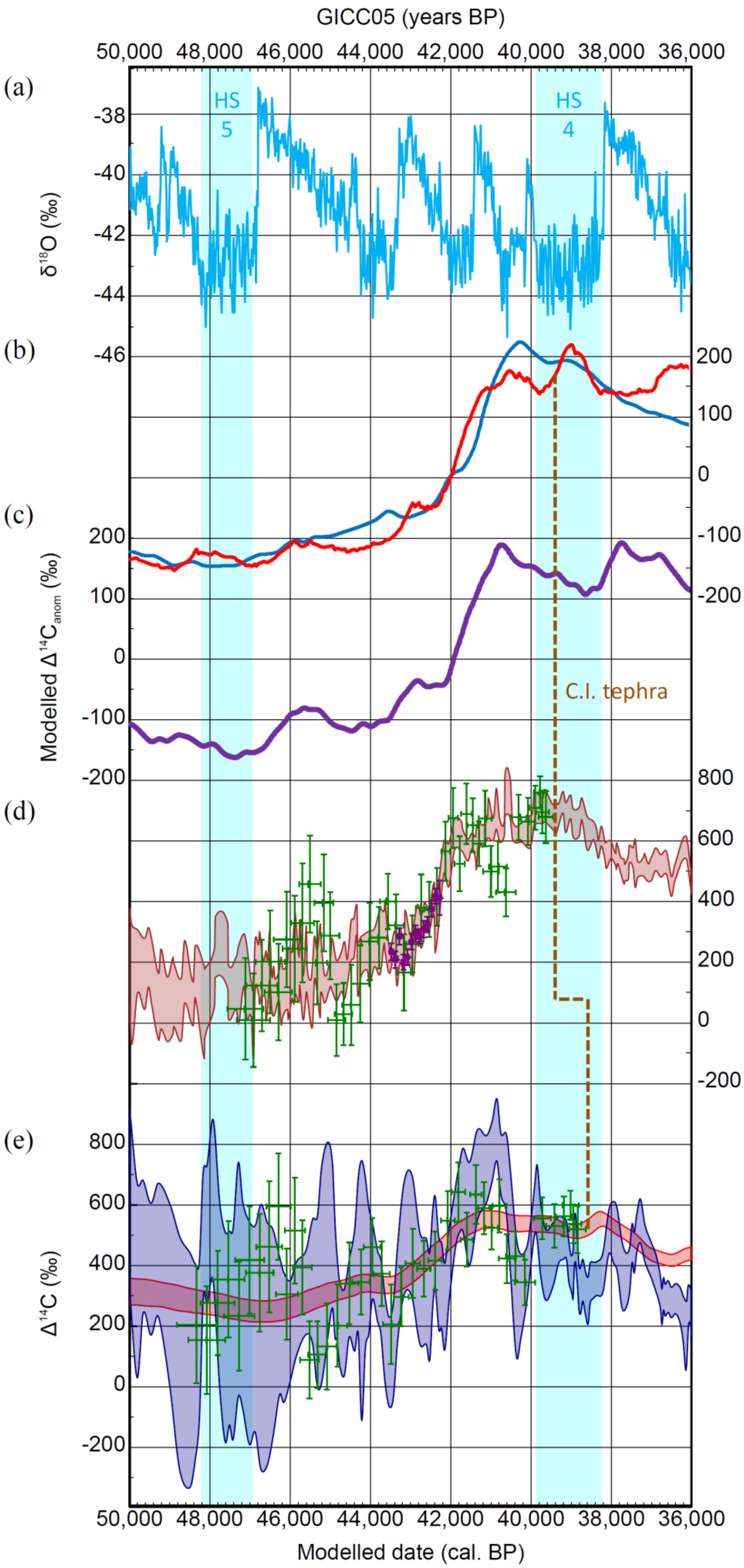


Fig. 3. Comparison of the shared production signals of the cosmogenic nuclides ^{14}C and ^{10}Be with relative palaeointensity. **(a)** NGRIP δ^{18}O (NGRIP members, 2004; light blue data series);

(b) Inferred $\Delta^{14}C$ from the GLOPIS-75 stack (Laj et al., 2004, 2014; blue data series) and Black Sea (Nowaczyk et al., 2013; as 'tuned' to GICC05, red data series) relative palaeointensity datasets; **(c)** Inferred $\Delta^{14}C$ from Greenland ^{10}Be flux (Yiou et al., 1997; Muscheler et al., 2004, 2014b); **(d)** Reconstructed atmospheric ^{14}C concentrations ($\Delta^{14}C$) based on Tenaghi Philippon core TP-2005 (dark green data points; this paper), as well as the kauri dataset of Turney et al. (2010; purple data series), as modelled against the Hulu Cave ^{14}C calibration dataset (Cheng et al., 2018; pink curve); **(e)** Reconstructed atmospheric ^{14}C concentrations ($\Delta^{14}C$) based on Tenaghi Philippon core TP-2005 (dark green data points; this paper) as modelled against IntCal13 (red curve). For comparison, the Lake Suigetsu (Bronk Ramsey et al., 2012) (blue dataset) is additionally plotted. For clarity, all data are plotted at 68.2%/1σ probability ranges. (a-c) are all plotted on the GICC05 timescale BP (Andersen et al., 2006; Rasmussen et al., 2006; Svensson et al., 2008); (d) is plotted on the Hulu Cave U-series timescale; and (e) is plotted on the IntCal13 cal. BP timescale. Additionally, the shaded light blue boxes mark the approximate timings of Heinrich stadials HS4 and HS5 (Sanchez Goñi and Harrison, 2010); the hashed brown line marks the position of the Campanian Ignimbrite (C.I.) tephra in the Tenaghi Philippon and Black Sea records.


GICC05 (years BP)
50,000 48,000 46,000 44,000 42,000 40,000 38,000 36,000
(a)
(b)
(c)
(d)
(e)
HS 5
HS 4
δ18O (‰)
-38
-40
-42
-44
-46
Modelled Δ14Canom (‰)
200
100
0
-100
-200
C.I. tephra
Δ14C (‰)
800
600
400
200
0
-200
Modelled date (cal. BP)

40.