ENVIRONMENTAL STUDIES

Annual radiocarbon record indicates 16th century BCE date for the Thera eruption

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The mid-second millennium BCE eruption of Thera (Santorini) offers a critically important marker horizon to synchronize archaeological chronologies of the Aegean, Egypt, and the Near East and to anchor paleoenvironmental records from ice cores, speleothems, and lake sediments. Precise and accurate dating for the event has been the subject of many decades of research. Using calendar-dated tree rings, we created an annual resolution radiocarbon time series 1700–1500 BCE to validate, improve, or more clearly define the limitations for radiocarbon calibration of materials from key eruption contexts. Results show an offset from the international radiocarbon calibration curve, which indicates a shift in the calibrated age range for Thera toward the 16th century BCE. This finding sheds new light on the long-running debate focused on a discrepancy between radiocarbon (late 17th–early 16th century BCE) and archaeological (mid 16th–early 15th century BCE) dating evidence for Thera.

INTRODUCTION

The Minoan eruption of Thera (Santorini) in the second millennium BCE was one of the largest volcanic eruptions in the past 4000 years (*1*). The event has been intensively studied from archaeological and paleoenvironmental perspectives (*2*) because it provides a geological marker that, if precisely dated, could synchronize Bronze Age histories of the Aegean, Egypt, and the Near East and anchor a wide range of contemporary environmental data. Dating has, however, proved problematic because of observed discrepancies between timelines derived from archaeological evidence and those based on radiocarbon dating (*2*–*7*).

Debate over a date for the Thera eruption

Measurements of radiocarbon (^{14}C) from legumes and grains (*3*, *8*, *9*) buried directly beneath the eruption deposits, and an olive branch (*10*) buried within them, cluster in the range c.1650–1600 cal BCE relative to the internationally agreed radiocarbon calibration curve, IntCal13 (*11*), which converts radiocarbon determinations from samples of unknown age into calendar age estimates (*12*). These estimates place the eruption in the second half of the 17th century BCE (*9*, *13*). The reliability of this radiocarbon-based date range has been debated (*4*, *5*, *14*) because it places the event earlier in time than certain archaeological synchronizations between sites in the Aegean, Egypt, and Levant would suggest possible. Evidence indicates that the eruption occurred after the start of the New Kingdom in Egypt, which, according to proponents of conventional, archaeologically based chronology, is considered to be sometime after c.1550 to 1500 BCE (*4*, *14*), although arguments have been presented to move the start of this range as early as c.1570 BCE (*7*, *15*). An extensive integrated radiocarbon and archaeological study of dynastic Egypt (*16*), covering 1700 years and incorporating 211 radiocarbon dates, can be used to support both the conventional and earlier proposed date ranges by indicating a start date for the New Kingdom of between 1570 and 1544 BCE. The problem remains, however, that direct radiocarbon evidence for the Thera eruption currently places this event multiple decades earlier than the earliest possible start of the New Kingdom.

Arguments in support of the most recently proposed late 17th century calibrated calendar range for Thera (*9*, *13*, *17*) have focused on the consistency with which a large number of radiocarbon determinations from different laboratories, on different sample types, from secure archaeological contexts immediately predating the eruption, calibrate to the same point in time. While this logic confirms the synchronicity of the network of archaeological contexts, and the interlaboratory agreement on an approximate temporal window during which the eruption occurred, the derived calibrated calendar date ranges are highly dependent on how accurately IntCal13 represents radiocarbon levels for the time period. Many of the existing radiocarbon determinations have been recalibrated multiple times, with calibrations to older iterations of IntCal [for example, (*3*, *10*)] returning probability distributions ranging into the 16th century because of a slight shift in the position of a well-described (*3*) radiocarbon plateau in the calibration curve. Given the sensitivity of these critical data to the shape of the calibration curve, any improvement or insight regarding calibration in this period would clearly be widely beneficial.

Exploring annually resolved ^{14}C

Understanding of past timelines for human and environmental interaction has been transformed by the development of an internationally agreed radiocarbon calibration curve [IntCal13 (*11*)] based on the best available ^{14}C measurements produced from samples with independently estimated or known calendar ages. For the past 12,000 or so years, measurements are largely derived from consecutive, sometimes overlapping, decadal and semidecadal blocks of known-age tree rings. These determinations are then drawn together to form a single calibration curve against which new measurements on a range of carbon-based materials can be compared. The accuracy and precision to which one can date a new determination depend critically upon three factors: the volume and faithfulness of the calibration database in representing the level of atmospheric ^{14}C; the statistical methodology; and, crucially for precision, the shape of the curve itself, where many plateaus and inversions limit the potential precision achievable. Previous investigations

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(*18*) as to whether calibration accuracy and precision could be improved by the use of radiocarbon determinations for single years (versus multiyear blocks) focused on the question of improving calibrated ranges for materials representing a single, short season of annual growth. With typical year-to-year variations of c.8 to 16 ^{14}C years (1 or 2‰) reported, a sampling resolution of less than c.10 or so years appeared to offer little advantage, given that errors on individual determinations are also typically within that range or greater. However, the discovery (*19*) that variations of up to 96 ^{14}C years (12‰) can simultaneously occur between single years in tree ring series in both hemispheres (*20*), and the use (*21*, *22*) and potential (*23*) of these marker events to synchronize multiple chronologies, has initiated renewed investigation of annual ^{14}C.

As increasing numbers of annual resolution radiocarbon determinations become available for inclusion in IntCal, the utility of these data and the consequences of other subdecadal variability that may be hidden in the current multiyear resolution record must be explored. We selected the period 1700–1500 BCE for a systematic investigation of the potential contribution of annual ^{14}C to improve or better define the limitations for radiocarbon dating during the period most closely associated with the eruption of Thera.

RESULTS

Testing the IntCal13 calibration curve with multispecies annual data

We measured ^{14}C in single tree rings of known age from high-altitude bristlecone pine (*Pinus longaeva* D.K. Bailey) from the White Mountains of California and low-altitude oak (*Quercus* sp.—the dominant species used in IntCal13) from County Kildare, Ireland. We compared these measurements with the IntCal13 raw data and with the IntCal13 curve. Our 285 annual data points are all internally consistent and fall within the 2σ range of most of the raw data underlying IntCal13 (Fig. 1A); however, when compared to IntCal13, both species show a clear and sustained offset in the annual measurements (Fig. 1B) between c.1660 and 1540 BCE. The degree of offset observed is less against earlier iterations of IntCal, which do not include the full IntCal13 data set and feature less statistical smoothing (Fig. 2). Outside this range, our annual measurements merge back toward IntCal, indicating that this offset is not a result of a specific Arizona Accelerator Mass Spectrometry (AMS) laboratory bias (from 1700–1660 BCE, our data agree with IntCal13 at 1σ). Arizona has also shown no evidence of a bias in recent laboratory intercomparison work.

The IntCal13 raw data for 1700–1500 BCE consist of 119 ^{14}C measurements, derived from overlapping blocks of between 4 and 20 tree rings, run at five different laboratories over several decades (Fig. 2). These measurements are all derived from north European oak. The offset that we observe is of comparable size to those previously suggested to result from regional differences in ^{14}C uptake (*16*), yet our annual Irish oak data agree more closely with our annual bristlecone data than with the European oaks represented by IntCal13. To see such close correlation between separate species from strongly contrasting growth regions, thousands of kilometers apart, strengthens the credibility of a hemisphere-wide calibration curve. At the same time, it indicates that for the period c.1660–c.1540 BCE, where our data correlate more closely with one another than with IntCal13, these combined annual data are more likely to correctly represent ^{14}C. Similar points of correlation and offset (*24*) are being explored in other sequences of annual data versus IntCal13 in a number of time periods.

Testing the implications of the offset for calibration

To investigate the implications of the changed calibration curve described by the ICCP17 data set, we replicated the statistical procedures underlying IntCal13 (*25*) to create a combined curve based on our annual oak and pine data (ICCP17). We then recalibrated a number of radiocarbon determinations, from secure pre- or immediately post-eruption contexts, to both IntCal13 and ICCP17 and compared the difference.

The change of c.30–40 ^{14}C years (4 to 5‰) only makes a substantial difference to calibrated date ranges, within possible error per determination, where samples previously calibrated to the slope in the curve move onto the plateau area. This magnifies the implications for calibrating many samples from secure pre- or immediately post-Thera contexts because a large number of these, produced at different radiocarbon laboratories, on different types of material from different contexts, now move firmly onto the radiocarbon plateau. Examples of this effect for the two primary data sets from Santorini are provided in Table 1 and Fig. 3. In each case, the posterior mean based on ICCP17 moves into the 16th century BCE. Furthermore, the younger end of posterior age distribution is considerably extended in all scenarios, magnified by the plateau in the ICCP17 data. This plateau and the resultant wide spread in the posterior age range for the different models underline the difficulty in presenting a definitive precise radiocarbon-based age for Thera.

The issue caused by the plateau is particularly apparent in the case of the Akrotiri seed assemblage (*9*), which varies over c.360 radiocarbon years (including error) for samples from the same context. This can now be contrasted with a measured range of variability of c.260 radiocarbon

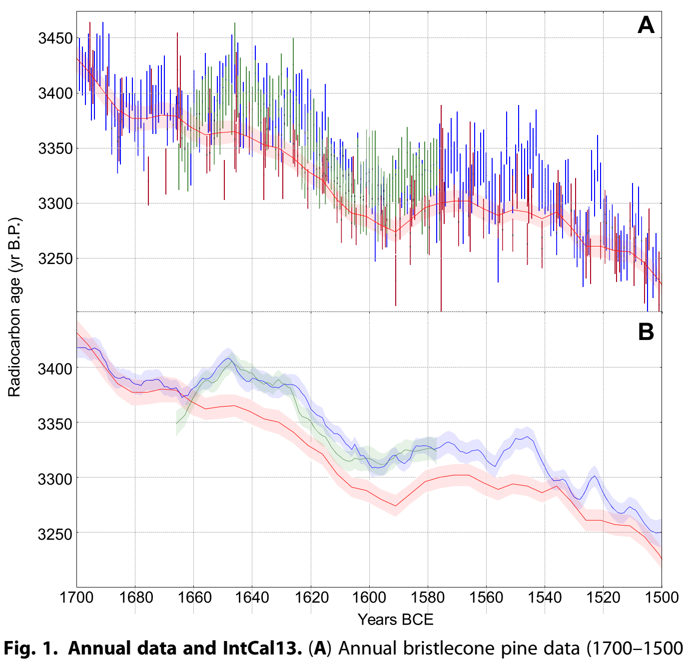

Fig. 1. Annual data and IntCal13. (**A**) Annual bristlecone pine data (1700–1500 BCE; blue) and Irish oak data (1661–1576 BCE; green) are shown relative to the IntCal13 raw data (red) and all show 2σ error. The majority of our annual data fall within the 2σ error of the IntCal raw data. (**B**) Calibration curves for pine (blue) and oak (green) constructed in the same way as IntCal13 (red) are shown with 1σ error. From 1700 BCE to c.1660 BCE, and after 1540 BCE, good agreement is shown between our data and the IntCal13 raw data. Between c.1660 BCE and c.1540 BCE, both species, from growth environments c.7900 km apart, indicate a clear and sustained offset from IntCal13.

years (including error) across the entire period of 1700–1500 BCE (Fig. 1A). While mixed-age issues with dates from the same archaeological context can be addressed via a range of Bayesian modeling techniques, in view of the very wide spread of dates in this case, and the sensitivity of any derived average to the position of the radiocarbon plateau, a different approach is worth considering. It should be noted that a number of uncalibrated radiocarbon determinations for the most recently living materials in a range of pre-eruption deposits cluster around c.3310 ± 35 B.P. (before present). These include the younger of two animal bones buried by the Thera tsunami deposit on Crete (*17*), the outer part of the Santorini olive branch (*10*), the younger of more recently measured seeds from Akrotiri (*9*), and materials from Tell el-Ajjul (Gaza) (*26*) (from immediately below an influx of geochemically provenanced pumice from Thera contemporary with the start of the New Kingdom in Egypt). Differences observed between dates for these contexts have mainly focused on the

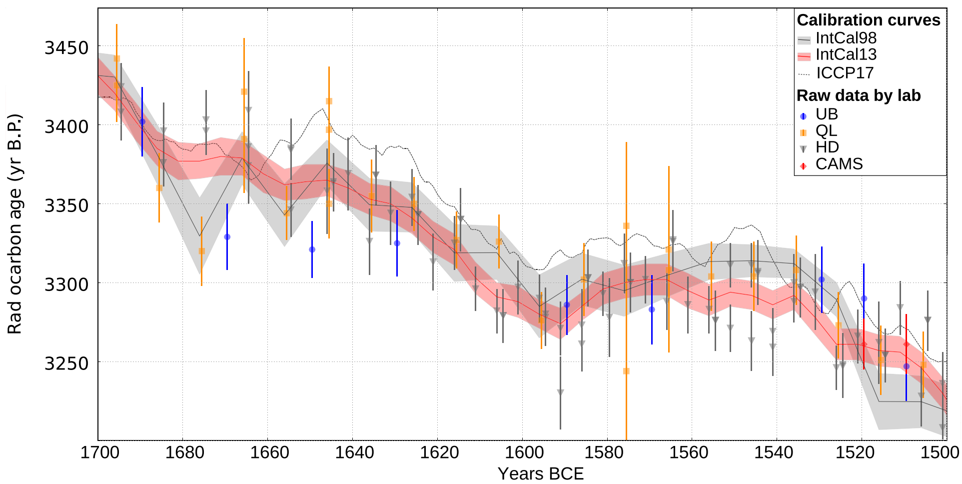

Fig. 2. IntCal raw data with IntCal13 and IntCal98 curves. Raw determinations of multiyear blocks of north European oak making up IntCal13, 1700–1500 BCE with the derived IntCal13 curve. Comparison with the IntCal98 curve provides an example of how the addition of extra data and changes in the statistical methodology used to produce the curve has changed the shape over the years. The dashed line shows the midpoint of the ICCP17 curve, which is less offset from IntCal98.

Table 1. Volcanic destruction layer radiocarbon determinations calibrated to IntCal13 and ICCP17. Using OxCal 4.3, we compare results for a number of previously published modeling scenarios. Models 1 to 3 show different modeled approaches to the assemblage of annual materials from pre-eruption contexts on Akrotiri (*8*, *9*, *15*, *41*). Models 4 and 5 show different modeled approaches for the Santorini olive branch, with assumptions of annual growth (*10*); annual growth with counting uncertainties of 25 and 50% (*10*); or an ordered sequence of unknown years, which moves from older to younger (*13*). All values for the olive branch represent posterior estimates of the outermost ring except model 7 (a simple ordered sequence), where the value relates to the estimated age of the outermost measurement.

Sample	Published model justification	Posterior mean cal BCE (2σ/95% credible interval)	
		IntCal13	ICCP17
Akrotiri seed assemblage	1: Archaeological assemblage represents a phase (number of different years) before the eruption (Tau boundary function) (*9*, *15*)	1625 (1646–1606)	1581 (1619–1536)
Akrotiri seed assemblage	2: Selected archaeological assemblage represents the same year before the eruption (adjusted for volcanic context, 3326 ± 11 ^{14}C B.P.) (*41*)	1608 (1658–1535)	1576 (1614–1539)
Akrotiri seed assemblage	3: Selected archaeological assemblage represents the same year before the eruption (unadjusted, 3345 ± 9 ^{14}C B.P.) (*41*)	1637 (1664–1614)	1587 (1664–1541)
Olive branch segments	4: Accepts olive increments as measured by Friedrich *et al.* (*10*) are annual	1615 (1626–1605)	1589 (1610–1567)
Olive branch segments	5: Ring count is increased by 25%, and gap uncertainty set to 25% of section count (*10*)	1611 (1627–1596)	1574 (1608–1511)
Olive branch segments	6: Gap uncertainty set to 50% of the ring count (*10*)	1622 (1721–1597)	1577 (1612–1528)
Olive branch segments	7: Treats sequence of olive segments as ordered but with no information on ring counts/gaps (*13*)	1614 (1642–1559)	1570 (1614–1538)

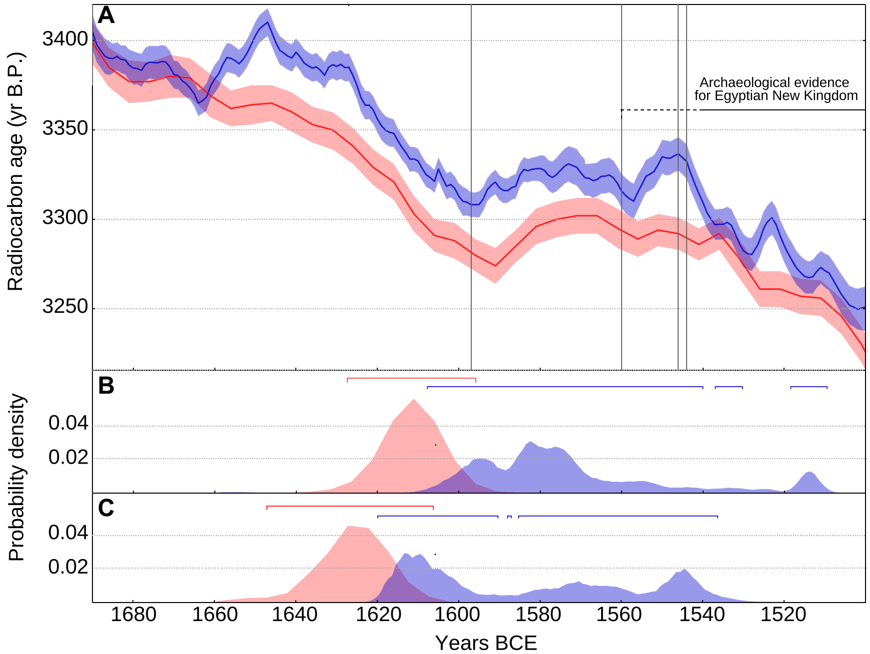

Fig. 3. Examples of the change in date range for materials from key Thera contexts. (**A**) Combined oak and pine ICCP17 calibration curve (blue) compared with IntCal13 (red), with 1σ error quoted as standard. The shift in the OxCal probability distribution for the olive branch (**B**) (based on model 5, Table 1) and the seeds (**C**) (based on model 1, Table 1) are color-coded relative to each curve. The black dashed line (upper right) shows the earliest possible start date for the Egyptian New Kingdom within the archaeological framework, with the solid line indicating the more conventional range. Single vertical lines mark bristlecone pine "marker years" at 1597, 1560, 1546, and 1544 BCE referred to in the text. Note the presence of pronounced solar cycles in the plateau region, especially between 1560 and 1520 BCE. These features will be the focus of future work to improve calibration and chronological synchronization for this time period.

calibrated versions of these measurements, which can vary greatly in a plateau region depending on the calibration curve, average or model used, and differences in error on each measurement.

The sequence of four radiocarbon measurements from the Santorini olive branch (*10*) has the advantage of representing a period of time rather than the single season (or range of single seasons) represented by other Thera relevant samples. This offers the possibility to define a wiggle-matched fit to a given calibration curve to help constrain the date across the plateau. Unfortunately, however, olive trees have indistinct growth boundaries that may represent multiple growth phases in a single year or may miss periods of growth (*27*). As a result of this, and other potential problems such as the branch being dead before the eruption or missing a number of years of outermost growth, the sample has been greatly discussed (*4*, *9*, *13*, *15*, *27*, *28*). In response, a number of modeled scenarios have been published for the four consecutive ^{14}C measurements produced from the sample. These reflect different views on the likelihood that the 72 counted increments in the branch represent annual growth. The most recently published (*13*) and widely agreed modeling scenario, which does not attempt to argue yearly growth, but rather presents a simple ordered sequence of older wood to younger wood, results in a 2σ range of 1614–1538 cal BCE for the outermost wood sample relative to ICCP17 (Table 1). The change in probability distribution if 25% certainty is placed on the counted growth bands is shown in Fig. 3B, with Fig. 3C showing the same for an average of the Akrotiri annual material modeled using a Tau boundary function.

DISCUSSION

Assuming that the ICCP17 data more faithfully reflect radiocarbon levels for the time period than IntCal13, the resultant shift in the posterior distributions for calibrated ages of pre-Thera eruption contexts illustrates how radiocarbon evidence for the dating of Thera could now be argued to be compatible with multiple lines of evidence for dating of the New Kingdom (18th dynasty) in Egypt. The revised calibrated ranges shown in Table 1 provide the flexibility for the long debated dating discrepancy to be resolved where the earlier proposed start of c.1570/1560 BCE is accepted, but also, extended credible intervals beyond 1550 BCE (in all but scenario 4) offer a period of overlap with conventional, archaeologically based chronology. We do note, however, that our data indicate that a date for the Thera eruption more recent than c.1510 BCE is highly unlikely, which remains at odds with certain archaeological arguments (*14*), and credible intervals do not exclude an eruption in the late 17th century BCE.

The recalibrated results shown in Table 1 provide a new focus to search the proxy records for markers that can be directly linked to Thera, in particular, the ice core records, where volcanic acidity and tephra can be used to more closely define specific eruptions (*22*). There is some indication that dating for the ice core record during this period may need adjustment (*29*). New radiocarbon determinations for the Aniakchak II eruption (*30*) (3270 ± 40 ^{14}C years B.P. for a peat layer just below the tephra) suggest that the geochemically anchored ice layer for this event [c.1641 BCE (*31*)] should become more recent, in which case new acidity levels in the ice could become viable candidates for Thera in the 16th century BCE. In particular, this should be explored using the well-established (*22*, *32*) correlation between volcanic acidity layers in the ice and anomalous growth in upper treeline bristlecone pine. While our data place previous tree ring marker dates suggested for Thera [1626–1628 BCE (*33*, *34*)] outside the 95% probability range, the bristlecone pine chronology has a ring width minimum at 1597 BCE and a frost-damaged ring at 1560 BCE (*32*), indicating major volcanic events around these years. The 1597 BCE date is earlier than indicated by the posterior mean of our revised dating of the olive branch and Akrotiri assemblage but within the 95% credible interval and could be argued to correlate with a chemical response recorded in a speleothem at Sofular Cave in Turkey (*35*), which is independently dated using ^{230}Th dating and layer counting. However, given the complexity of the cave environment and errors associated with the dating method used, it also seems quite possible that this chemical signature could also extend to a connection with an eruption at 1560 BCE (and beyond), with sulfur elevated between c.1620 and 1530 ± 25 BCE. The 1560 BCE date, which represents the first proxy marker for a volcanic event in the bristlecone pine record beyond the posterior mean values derived for this study, would fit with the earlier proposed start for the New Kingdom in Egypt, the revised reign of Ahmose, and the description of an unusual and catastrophic storm from the Ahmose "Tempest Stela" (*36*). Equally, other bristlecone growth anomalies at 1546 and 1544 BCE also warrant further investigation, given the 95% credible intervals for the new radiocarbon ranges and compatibility with conventional, archaeologically derived dates for the start of the New Kingdom.

This study demonstrates the advantages of annual ^{14}C time series to beneficially augment the coarser-resolution measurements of IntCal13. We show that this can be particularly important in transition periods around ^{14}C plateaus, where small changes in the curve can have a large effect on calibrated ages. Here, annual measurements are able to provide finer detail than measurements on multiyear blocks. The results indicate that the Thera eruption occurred during a plateau in ^{14}C production, making current radiocarbon ranges less precise and limiting the potential of radiocarbon dating to provide an exact date for the event. Results do, however, provide a strong basis for new investigation of the utility of solar cycles, and patterns of more rapid ^{14}C production identified c.1580–1520 BCE in improving this situation. They also provide a basis for a renewed focus on proxy records for the 16th century BCE, starting with the precise bristlecone calendar dates of 1597, 1560, 1546, and 1544 BCE. Future work will address any potential impact of the suggested alteration to the calibration curve for other established models overlapping with the time period [for example, (*16*)]. The annual time series for this period should also be extended and replicated at other laboratories using different regional tree ring chronologies to more fully explore the implications for accuracy and precision made possible via an annually based calibration resource. Work is also needed to explore the degree of interlaboratory variation in annual ^{14}C data and to fully understand the combination of factors leading to the difference between IntCal13 and the annual time series. No definitive calibrated radiocarbon range for the Thera eruption is currently possible, but the altered position of the ^{14}C plateau indicates that improved calibration has much to offer chronological synchronization of human and environmental timelines in this period.

MATERIALS AND METHODS

Experimental design

The main objectives of the study were to sample long time series of tree rings from North American bristlecone pine and Irish oak for an annual or growth season–specific representation of ^{14}C for the calendar year in which each tree ring formed. Samples were selected from well-replicated master tree ring chronologies with absolute calendar dates assigned to the tree rings according to standard dendrochronological procedures (*37*). Individual annual growth increments from the mid-second millennium BCE were then dissected using a binocular microscope and steel blade. For the pine, the whole tree ring was used for each year represented. For the oak, which has a distinct ring porous structure with large "earlywood" vessels known to include ^{14}C from the previous growth year (*38*), only the "latewood" was sampled. The bristlecone pine samples represent a c.45 day growth season from mid to late June until late July or early August, with limited potential for photosynthesis outside the growing season. The oak latewood samples represent late May/June. Together, they represent the main growth season in the Mediterranean.

Wood samples were converted to holocellulose at the University of Arizona AMS Laboratory one sample at a time. This involved standard 1 N HCl/NaOH/HCl extractions at 70°C, followed by a holocellulose extraction at 70°C using a bleaching solution made from sodium chlorite, HCl, and water. The dissection method generated submillimeter-thick slabs with large surface areas and short diffusion distances that facilitated the uptake of reagents and the extraction of contaminants. Treated samples appeared bright white in color. Samples were combusted to CO_2 and converted to graphite using standard procedures (*39*) and then measured using a National Electrostatics Corporation AMS system, operated at a terminal voltage of 2.5 MV. The ^{14}C/^{13}C ratio of each sample was compared to National Institute of Standards and Technology standards SRM4990B and 4990C, and the resulting fractionation was corrected to a δ^{13}C value measured offline on a stable isotope mass spectrometer.

The interlaboratory comparability for measurements of bristlecone pine was published in 2017 (*40*) with sample collection for the published data ending 1 month before our period of data collection began. Also, a 2017 interlaboratory comparison organized as part of the IntCal Dendrochronological focus group by L. Wacker (ETH Zurich) tested performance in measuring annual radiocarbon content in European oak tree ring sequences. In both, the scatter of the Arizona measurements away from the mean of the clustered measurements fell within the quoted error.

Samples for this study were run in a nonconsecutive sequence over a 2-year period with standardized protocols throughout. The nonsequential nature of data collection is important to stress as it confirms that the offset is not caused by an intermittent laboratory factor.

Statistics

To explore the implications of calibrating Thera contexts against our annual data as opposed to IntCal, an ICCP17 calibration curve was constructed from our annual oak and pine using the same random walk approach used in IntCal13 (*25*). The prior variance on the random walk for this ICCP17 data set was chosen to match that used for IntCal13. Calibrations against both ICCP17 and IntCal13 were then performed using OxCal v.4.3 (*12*).

We attempted to follow previously published models as closely as possible, but some adjustments were necessary. Several options for the wiggle matching of sequences were available by adjusting the prior on the calendar ages as appropriate to the specific model that we wished to consider. To incorporate uncertainty on the accuracy of the olive tree ring counts (*10*), our prior was built sequentially as follows: $\pi(t_1) \propto 1$—an improper prior expressing no prior knowledge about start of sequence; $\pi(t_2 - t_1|t_1) \propto N(c_1, \zeta_1^2)$ s.t. $t_1 < t_2$—modeling the uncertain ring count between t_1 and t_2 and known order; …; $\pi(t_n - t_{n-1}|t_1,\ldots,t_{n-1}) \propto N(c_2, \zeta_n^2)$ s.t. $t_{n-1} < t_n$—the final uncertain count between t_{n-1} and t_n and the known order. Here, c_i is the expected number of rings/years between t_i and t_{i+1}, and ζ_i is the corresponding uncertainty.

To impose only ordering constraints on the olive tree ring segments, the prior was constructed by assuming $t_1,...,t_n \in [0, 60000]$ cal B.P. with our n individual tree rings being drawn independently and uniformly at random from this interval. The prior for $t_1,...,t_n$ was then the joint distribution of their ordered values, a constant, and equivalent to the limiting case of the uncertain ring counting prior

$$\pi(t_1,...,t_n) \propto \begin{cases} 1 & \text{if } t_1 < \ldots < t_n; \\ 0 & \text{otherwise.} \end{cases}$$

These priors required minor adjustments from the OxCal sequence defaults, which propose boundaries to introduce a uniform phase model prior. Our priors are, however, the correct choices if one's stated intention is to remove any constraint on the ring counting altogether and because the selection (number and location) of samples within the olive tree was not random but designed (*10*). Conversely, the phase model generates a prior that is dependent on the number of determinations of the tree taken. Were the olive branch to be sampled at more points, a phase model would shrink the estimated number of years elapsed between the outermost and innermost rings. This approach is not appropriate because the olive's age estimates should be invariant to the number of determinations.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/8/eaar8241/DC1

Table S1. Annual determinations of Irish oak and North American bristlecone pine used in this study.

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