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U/Th-dating living and young fossil corals from the central tropical Pacific

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Abstract

This study evaluates the accuracy of U/Th dates for young ($<$ a few thousand years old) reef corals, both living and fossil, and explores strategies for refining those dates. The high precision of the U/Th method ($\pm$ 1–2%) for dating young corals is well-established. Earlier studies have demonstrated the method's accuracy for select samples of known age. However, these studies have focused on typical samples that have extremely low ^{232}Th concentrations (tens of pg/g). Here we study the dating systematics of young corals that have low but significant amounts of ^{232}Th (up to 1000 pg/g), indicating the presence of small fractions of non-radiogenic ^{230}Th (i.e. ^{230}Th not generated by in situ U decay). We report U/Th ages for living and subaerially exposed fossil corals from Palmyra Island, located in the central tropical Pacific, that range from 50 to 700 yr old. The Palmyra corals contain varying amounts of ^{232}Th and small fractions of associated non-radiogenic ^{230}Th. Uncertainty associated with the correction for non-radiogenic ^{230}Th can lead to significant errors in U/Th dates. We have characterized non-radiogenic ^{230}Th/^{232}Th values, $(^{230}Th/^{232}Th)_{nr}$, as a means of minimizing this source of error. We calculate $(^{230}Th/^{232}Th)_{nr}$ values ranging from 0 to 2×10^{-5} for the Palmyra living corals by comparing measured U/Th dates to absolute dates for the living coral, whose chronology is well-established. For the fossil corals, we employ three different approaches to arrive at $(^{230}Th/^{232}Th)_{nr}$ estimates. First, we compare measured U/Th dates to absolute dates in samples from a young fossil coral that overlaps the living coral. Next, we use the firm relative dating constraints imposed by five overlapping fossil corals from the 14th–15th centuries to calculate $(^{230}Th/^{232}Th)_{nr}$ values. Finally, we attempt to anchor the 14th–15th century floating coral chronology to an absolute chronology by correlating the climate signals in the coral records to those in absolutely dated climate proxy records. All lines of evidence point to a range of $(^{230}Th/^{232}Th)_{nr}$ for fossil corals that overlaps the range determined for the living coral, suggesting that most of the thorium is primary or is added while the coral is still alive. Our work also demonstrates the utility of multiple $(^{230}Th/^{232}Th)_{nr}$ estimates. Most importantly, we demonstrate a method by which accurate ($\pm$ 5 yr) U/Th-based chronologies can be obtained for young fossil corals with significant ^{232}Th concentrations.

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1. Introduction

Accurate U/Th dating requires distinguishing between a sample's radiogenic ^{230}Th (that produced by in situ ^{238}U decay) and its non-radiogenic ^{230}Th (that derived from the surrounding environment). The power of U/Th dating stems from the fact that levels of non-radiogenic ^{230}Th, $(^{230}\mathrm{Th})_{nr}$, are small or negligible in most uranium-rich precipitates, such as carbonates, owing to the difference in solubility between uranium and thorium in the natural environment (a difference of five orders of magnitude). As a result, most precipitates from surface waters have low thorium/uranium ratios, and therefore low to negligible levels of non-radiogenic ^{230}Th and ^{232}Th. In fact, thorium concentrations in coralline aragonite are so low that coral ^{232}Th levels were not known prior to the advent of high-precision mass spectrometry techniques [1]. Because of these low thorium levels, corrections for non-radiogenic ^{230}Th are generally negligible for corals that are older than a few thousand years or that contain less than $\sim$100 pg/g ^{232}Th [1–3]. However, recent work has shown that some young corals can have ^{232}Th concentrations significantly higher than tens of pg/g [4], requiring corrections for the contribution of non-radiogenic ^{230}Th to the U/Th age. For the purposes of dating, corals with negligible $^{230}\mathrm{Th}_{nr}$ and low ^{232}Th are clearly preferable. Yet these characteristics are not always available in any set of samples, making the dating of high-^{232}Th corals necessary.

The $^{230}\mathrm{Th}_{nr}$ correction involves combining sample ^{232}Th concentrations with estimates of the ^{230}Th/^{232}Th ratio of the non-radiogenic thorium, $(^{230}\mathrm{Th}/^{232}\mathrm{Th})_{nr}$. When combined with the wide range of ^{230}Th/^{232}Th values observed in the natural environment [5–10], the $^{230}\mathrm{Th}_{nr}$ correction can introduce substantial uncertainty to U/Th ages if ^{232}Th is high and knowledge of ^{230}Th/^{232}Th in the environment of interest is poorly known. Here we investigate the U/Th isotope systematics of a collection of young corals with high ^{232}Th concentrations as a test of the accuracy of U/Th dates under some of the most demanding of circumstances for the method.

Independent time constraints are needed for accurate determination of $(^{230}\mathrm{Th}/^{232}\mathrm{Th})_{nr}$ values. Massive corals are well-suited for characterizing the isotopic composition of non-radiogenic thorium, because their climate proxy records and annual banding provide independent, firm dating constraints. In principle, $(^{230}\mathrm{Th}/^{232}\mathrm{Th})_{nr}$ can be calculated from the U/Th age equation, given a sample of known age, and thorium and uranium isotopic analyses. In practice, we obtain the best constraints on $(^{230}\mathrm{Th}/^{232}\mathrm{Th})_{nr}$ from those samples with high ^{232}Th. Previous studies on low-^{232}Th corals [1,2] have found insignificant levels of $^{230}\mathrm{Th}_{nr}$. On the other hand, a recent study of living, or modern, corals and young fossil corals from Sumatra documents cases where very young corals contain relatively high ^{232}Th and significant non-radiogenic ^{230}Th [11]. The latter result may be related to Sumatra's proximity to continent-derived sources of thorium. Therefore, it is important to assess the role of $^{230}\mathrm{Th}_{nr}$ corrections in coral U/Th dates at a site far removed from continental influences.

In this study we draw from a large collection of young modern and fossil coral sequences from Palmyra Island, located in the central tropical Pacific Ocean (Fig. 1, inset). In general, our strategy is to match the climate signals from contemporaneous corals, comparing the corals' U/Th dates with independent chronological constraints. These comparisons include samples from a living coral collected while growing underwater and samples from subaerially exposed fossil corals, characterized by different diagenetic histories. Multiple U/Th dates from the same coral head (for which we can count annual growth bands) provide another test of the range of both $(^{230}\mathrm{Th}/^{232}\mathrm{Th})_{nr}$ ratios and ^{232}Th concentrations. We constrain the mechanisms and rates of possible early coral diagenesis by comparing the isotopic characteristics of a living coral to those of a contemporaneous fossil coral. Therefore, our results outline an approach by which accurate U/Th dates can be obtained for

very young, high-[^{232}Th] corals. Our principal motivation in pursuing this work is to generate accurate U/Th-based chronologies for young fossil corals that could be used to reconstruct tropical climate for the last millennium [12].

2. U/Th systematics

The U/Th-dating method in corals relies on the fact that coral skeletons form with very low Th/U ratios (^{232}Th/^{238}U values of $\sim 10^{-5}$), which they inherit from seawater without a large degree of fractionation. The ensuing ^{238}U-decay chain includes ^{230}Th, whose activity generally increases with the age of the coral. If the coral remains a closed system with respect to U and Th and ^{230}Th$_{nr}$ is negligible, the following equations can be used to calculate ages from ^{230}Th$_{rad}$, ^{234}U, and ^{238}U activities:

$$1-\left(\frac{^{230}\mathrm{Th}_{rad}}{^{238}\mathrm{U}}\right)_{act} = e^{-\lambda_{230}T} - \left(\frac{\delta^{234}\mathrm{U}(0)}{1000}\right)\left(\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\right)(1-e^{(\lambda_{234}-\lambda_{230})T}) \quad (1)$$

$$\delta^{234}\mathrm{U}(T) = \delta^{234}\mathrm{U}(0)e^{\lambda_{234}T} \quad (2)$$

(after Edwards et al. [1]; modified from Bateman [13] and Broecker [14]) where λs are decay constants as reported in Cheng et al. [15] and T is the age.

The fact that young corals contain both ^{232}Th and ^{230}Th means that the measured ^{230}Th abundance includes a radiogenic ^{230}Th as well as a non-radiogenic ^{230}Th component. If the non-radiogenic ^{230}Th component is small and the coral has remained a closed system, Eqs. 1 and 2 suffice for calculating accurate U/Th ages. However, if ^{230}Th$_{nr}$ is significant, a correction for ^{230}Th$_{nr}$ must be made. Using ^{232}Th concentrations as a proxy for the amount of ^{230}Th$_{nr}$, the correction is applied to the measured (^{230}Th$_{meas}$/^{238}U)$_{act}$ ratio to obtain the (^{230}Th$_{rad}$/^{238}U)$_{act}$ term in Eq. 1:

$$\left(\frac{^{230}\mathrm{Th}_{rad}}{^{238}\mathrm{U}}\right)_{act} = \left(\frac{^{230}\mathrm{Th}_{meas}}{^{238}\mathrm{U}}\right)_{act} - \left(\frac{^{232}\mathrm{Th}}{^{238}\mathrm{U}}\right)_{act}\left(\frac{^{230}\mathrm{Th}_{nr}}{^{232}\mathrm{Th}}\right)_{act} e^{-\lambda_{230}T} \quad (3)$$

For samples that are younger than $\sim$1000 yr, such as the Palmyra corals, $\exp(-\lambda_{230}T)$ is very close to 1, and can be ignored. Taken together, Eqs. 1–3 illustrate that accurate, precise U/Th dates require precise measurements of ^{230}Th, ^{232}Th, ^{234}U, and ^{238}U abundances. If the ^{232}Th/^{238}U value is low, as is the case for virtually all U/Th age determinations in surface corals, the second term on the right-hand side of Eq. 3 becomes negligible. However, if the ^{232}Th/^{238}U value is high, then knowledge of (^{230}Th/^{232}Th)$_{nr}$ becomes important; it is this latter case that we address in the present study.

3. Sources of Th in the central tropical Pacific

In those cases where ^{232}Th/^{238}U is high, the isotopic characterization of the non-radiogenic Th may not be a simple problem, as a sample's (^{230}Th/^{232}Th)$_{nr}$ value likely reflects a combination of multiple Th sources. For example, we identify several potential sources of non-radiogenic Th at Palmyra that may affect the Palmyra corals' (^{230}Th/^{232}Th)$_{nr}$ values. First, wind-blown dust likely reaches Palmyra, although in small quantities, given its distance from the continents [16]. Dust likely contains a ^{230}Th/^{232}Th ratio close to 4×10^{-6}, assuming that it reflects the average chemical composition of bulk Earth at secular equilibrium [17,18]. Another source of non-radiogenic Th to the corals is the seawater, in the form of dissolved and particulate Th. The closest available seawater Th data were collected nearly 1000 km from Palmyra [8], with surface ^{230}Th/^{232}Th values of $5–10\times10^{-6}$ and deep-water values of up to 2×10^{-4}. As Palmyra lies in a region that is affected by upwelling variability associated with the El Niño-Southern Oscillation, deeper waters with elevated ^{230}Th/^{232}Th ratios could affect Palmyra's seawater Th chemistry. Lastly, carbonate sands that are produced by ongoing erosion of the coral reef may contribute Th with ^{230}Th/^{232}Th ratios as high as 1×10^{-2}, in the case of 5000 yr old

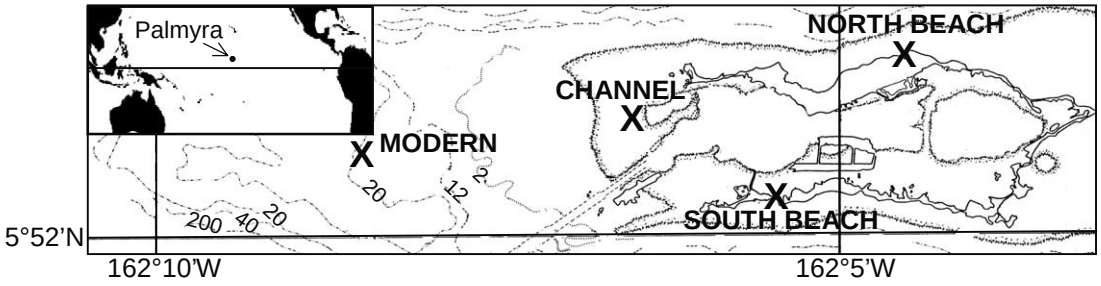

Fig. 1. Location and map of Palmyra Island. The sites where we collected the modern and fossil corals used in this study are marked with an 'X'. Sample names correspond to the sites shown here: NB, North Beach; SB, South Beach; and CH, Channel Site. Depth contours are given in meters.

carbonates that lie exposed on some areas of the reef (Richard Fairbanks, unpublished data).

4. Sampling and analytical methods

The modern and fossil corals analyzed here were collected on Palmyra Island during field expeditions in June, 1998 and May, 2000 (Fig. 1). During the first trip, we recovered a 3 m long core from a living coral with conventional underwater hydraulic drilling equipment in approximately 10 m of water on a reef extending to the west of the island. We will refer to this coral as the 'modern' coral throughout the paper. During the first and second trips we drilled over 100 1–2 m long fossil coral cores from beaches on the north, west, and east sides of the island. The fossil coral heads are piled in what appear to be storm deposits, inferred from the presence of large tree trunks and other debris found scattered in and among the coral heads. Further evidence for the storm-derived nature of these deposits comes from the corals themselves: they lack the heavy bioerosion that typically occurs in colonies that remain in seawater for extended periods after death.

The U/Th dating was performed at the Minnesota Isotope Lab (MIL) at the University of Minnesota, following procedures outlined in Edwards et al. [1] and subsequently updated in Cheng et al. [15] (thermal ionization mass spectrometry (TIMS) analyses) and Shen et al. [19] (inductively coupled plasma mass spectrometry (ICP-MS) analyses). Generally, we U/Th-dated two to five samples of different ages from each of the coral heads presented here, and performed replicate analyses wherever practical. In the case of the fossil corals, we sub-sampled for U/Th chemistry at least 5 cm away from any clear irregularities such as burrows and weathering rinds, which are visibly distinct from the original coral skeleton. We prepared the samples by breaking several grams of coral (representing $\sim$1 yr of coral growth) into small pieces for visual inspection, removing any discoloration with an exacto knife. The samples were ultrasonicated and rinsed three times, and placed in a drying oven overnight at 30°C. We then dissolved 0.5–1 g of coral in concentrated HNO_3 acid and added a MIL mixed spike containing ^{229}Th, ^{233}U, and ^{236}U. The purified U and Th fractions were either loaded onto zone-refined rhenium filaments, in the case of analysis by thermal ionization mass spectrometry, or dissolved in 1% HNO_3 for analysis by ICP-MS. While the majority of the samples were analyzed by TIMS using two Finnigan MAT 262 RPQ mass spectrometers, several samples were analyzed by high-resolution ICP-MS using a tech-

Table 1
Comparison of ICP-MS- and TIMS-based Th measurements made on splits from the same Th sample solutions (the U data for all samples were collected by ICP-MS only), all errors are reported as 2σ

Sample name	Date AD		[^{232}Th] (pg/g)	
	TIMS	ICP-MS	TIMS	ICP-MS
CH23	1269 ± 8	1262 ± 8	210 ± 7	217 ± 7
SB11	1203 ± 8	1199 ± 10	30 ± 8	19 ± 8
NB12	977 ± 10	960 ± 10	81 ± 8	78 ± 8

Table 2
U/Th results for modern and fossil corals

Sample[a]	$[^{238}U]$ [b] (ppm)	$(^{230}Th/^{238}U)_{act}\times10^3$ [c]	$(^{234}U/^{235}U)_{atomic}\times10^3$ [d]	$\delta^{234}U$ (i) [e] (‰)	$[^{232}Th]$ [f] (pg/g)	Assigned date[g]	Uncorrected date[h]	Date min. correction[i]	Date max. correction[j]	Method
Modern-1A	2.481 ± 5	1.257 ± 22	8.880 ± 13	143.0 ± 1.9	910 ± 9	1888	1880 ± 2	1889	1922	TIMS
Modern-1B	2.487 ± 3	1.189 ± 32	8.8652 ± 65	146.0 ± 1.3	534 ± 15	1888	1887 ± 3	1892	1911	TIMS
Modern-2	2.506 ± 4	0.690 ± 25	8.9220 ± 83	145.1 ± 1.8	148 ± 13	1944	1935 ± 2	1937	1942	TIMS
NB9-1A	2.534 ± 3	0.862 ± 22	8.8590 ± 76	146.1 ± 1.1	767 ± 8	1936	1917 ± 2	1924	1952	TIMS
NB9-1B	2.583 ± 3	0.827 ± 14	8.8458 ± 79	145.4 ± 1.2	628 ± 6	1936	1920 ± 2	1926	1948	TIMS
NB9-1C	2.447 ± 4	0.800 ± 22	8.7814 ± 67	144.1 ± 1.8	385 ± 12	1936	1925 ± 2	1929	1943	TIMS
NB9-2A	2.385 ± 7	1.162 ± 19	8.885 ± 10	148.3 ± 3	354 ± 10	1910	1890 ± 2	1894	1908	TIMS
NB9-2B	2.567 ± 22	1.080 ± 25	8.782 ± 57	144.2 ± 9	278 ± 12	1910	1898 ± 3	1901	1910	TIMS
CH5-1A	2.697 ± 4	6.419 ± 46	8.8190 ± 72	146.3 ± 1.3	1115 ± 9	1405	1386 ± 5	1396	1433	TIMS
CH5-1B	2.653 ± 4	6.369 ± 38	8.8459 ± 74	145.8 ± 1.3	925 ± 5	1405	1390 ± 4	1399	1430	TIMS
CH5-2	2.587 ± 2	5.995 ± 77	8.826 ± 12	146.4 ± 1.6	209 ± 8	1433	1428 ± 8	1432	1438	ICP-MS
CH5-3A	2.745 ± 4	5.563 ± 69	8.825 ± 12	148.2 ± 1.5	166 ± 8	1461	1471 ± 7	1472	1478	ICP-MS
CH5-3B	2.460 ± 3	5.632 ± 86	8.793 ± 16	145.2 ± 2.1	997 ± 21	1461	1463 ± 8	1473	1509	ICP-MS
SB6-1	2.611 ± 3	5.947 ± 46	8.825 ± 10	145.6 ± 1.4	461 ± 4	1442	1431 ± 5	1435	1451	TIMS
SB6-2A	2.509 ± 3	6.093 ± 122	8.830 ± 21	143.8 ± 2.7	45 ± 9	1412	1418 ± 12	1418	1420	ICP-MS
SB6-2B	2.224 ± 5	6.083 ± 109	8.865 ± 18	145.5 ± 2.4	0 ± 28	1412	1420 ± 10	1420	1420	ICP-MS
SB5-1A	2.849 ± 4	6.026 ± 54	8.8475 ± 65	146.7 ± 1.1	228 ± 5	1447	1424 ± 5	1426	1433	TIMS
SB5-1B	2.698 ± 9	6.078 ± 54	8.760 ± 13	145.0 ± 3	96 ± 9	1447	1420 ± 5	1421	1424	TIMS
SB5-2A	2.500 ± 1	6.070 ± 72	8.9206 ± 84	148.5 ± 1.2	97 ± 12	1427	1422 ± 7	1423	1427	TIMS
SB5-2B	2.629 ± 6	6.066 ± 51	8.796 ± 13	143.5 ± 2.3	123 ± 15	1427	1420 ± 5	1421	1426	TIMS
SB5-3A	2.537 ± 3	6.389 ± 46	8.8660 ± 55	146.1 ± 1.1	172 ± 11	1407	1391 ± 5	1392	1398	TIMS
SB5-3B	2.657 ± 5	6.304 ± 46	8.7811 ± 85	145.9 ± 1.7	89 ± 11	1407	1399 ± 5	1399	1402	TIMS
SB5-4	2.754 ± 4	5.962 ± 54	8.832 ± 12	143.4 ± 2.7	109 ± 9	1440	1432 ± 7	1433	1437	ICP-MS
SB7-1	2.698 ± 4	6.536 ± 52	8.8653 ± 68	146.7 ± 1.3	298 ± 8	1352	1375 ± 5	1378	1388	TIMS
SB7-2A	2.726 ± 5	6.852 ± 89	8.833 ± 17	142.5 ± 2.2	36 ± 11	1327	1344 ± 9	1344	1346	ICP-MS
SB7-2B	2.740 ± 6	6.967 ± 109	8.868 ± 21	143.4 ± 2.7	37 ± 13	1327	1334 ± 11	1334	1335	ICP-MS
CH9-1	2.451 ± 1	7.198 ± 50	8.8140 ± 61	145.2 ± 1.2	442 ± 8	1324	1311 ± 5	1316	1332	TIMS
CH9-2	2.319 ± 3	6.720 ± 60	8.9072 ± 52	148.0 ± 1.0	148 ± 10	1365	1360 ± 6	1361	1369	TIMS
CH9-3	2.167 ± 3	6.259 ± 39	8.8638 ± 50	146.1 ± 1.0	42 ± 8	1403	1403 ± 4	1403	1405	TIMS

[a] A, B, and C are replicate samples that we obtained by splitting a 2 g coral sample into multiple pieces.
[b] All errors reported in this table are quoted as 2σ; for $[^{238}U]$, error is for last significant figure.
[c] The measured, uncorrected $(^{230}Th/^{238}U)_{act}$ ratio; 2σ error is for last significant figure.
[d] The measured, uncorrected $(^{234}U/^{235}U)_{atomic}$ ratio; 2σ error is for last significant figure.
[e] $\delta^{234}U = \{[(^{234}U/^{238}U)/(^{234}U/^{238})_{eq}]-1\}\times10^3$, where $(^{234}U/^{238}U)_{eq}$ represents the atomic ratio at secular equilibrium; $\delta^{234}U$(i) represents the initial value, calculated using Eqs. 1 and 2. Decay constants from Cheng et al. [15].
[f] Corrected for a ^{232}Th total procedural blank of 7 ± 5 pg/g.
[g] All dates are reported as year AD. Assigned dates for Modern and NB9 samples are absolute ages derived from the modern coral's oxygen isotopic chronology, error is estimated at ± 0.5 yr; assigned dates for the rest of the samples are derived from the dating constraints posed by Fig. 6, error is estimated at ± 5 years.
[h] Error represents analytical error only.
[i] Date corrected with a $(^{230}Th/^{232}Th)_{atomic}$ of 4.4×10^{-6}, which is the value for materials at secular equilibrium, assuming a bulk Earth crustal $^{232}Th/^{238}U$ ratio of 3.8.
[j] Date corrected with a $(^{230}Th/^{232}Th)_{atomic}$ of 2.0×10^{-5}, which is the highest value required to correct samples in the present study.

nique developed at MIL with a Finnigan MAT Element I [19]. The TIMS and ICP-MS results for splits of the same sample are identical within error, so we conclude that the different measurement techniques do not bias the results presented here (Table 1).

The coral cores were cross-sectioned into 1 cm wide slabs and X-rayed in preparation for the oxygen isotopic analyses, which were carried out with a Finnigan MAT 252 stable isotope mass spectrometer at the Scripps Institution of Oceanography. We milled coral powder for the oxygen isotopic samples at 1 mm increments using a low-speed micro-drill. Given growth rates that average 15 mm/yr, this depth resolution translated to sub-monthly temporal resolution for the coral-based climate reconstruction. We used a well-characterized internal coral standard to check accuracy and precision, which we report as ± 0.05‰ based on more than 500 standards run along with the sam-

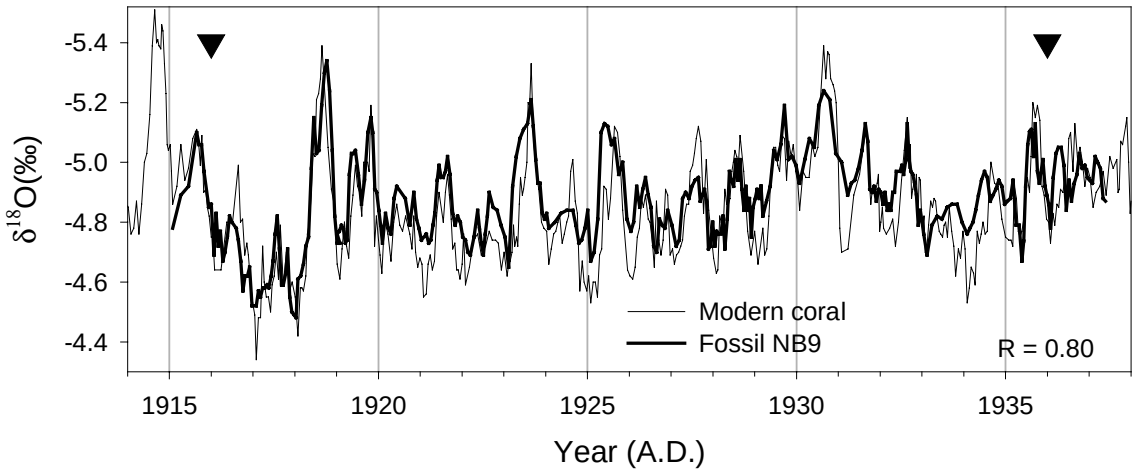

Fig. 2. The overlap of the fossil NB9's $\delta^{18}O$ record with that of the modern coral ($R = 0.80$). The triangles designate the locations that were sampled for U/Th dating in NB9.

ples. We constructed a chronology for the modern coral by matching the most recent portion of the oxygen isotopic record to sea-surface temperature records from the grid-box containing Palmyra for the period 1981–1998 ($R = -0.79$) [20]. In that analysis, we found that the heaviest isotopic value corresponded to January 30th ±43 days (2σ) for a given isotopic year, while the lightest isotopic value corresponded to August 26th ±53 days (2σ). We used these two tie-points to construct a chronology for the entire 112 yr long modern coral record, whose main features are reported elsewhere [12,21]. We used the two tie-points to construct the fossil coral chronologies.

Additionally, we investigated some basic lithologic properties with X-ray diffraction and thin sections on both modern and fossil corals in order to constrain the role of diagenesis in shaping the observed U/Th chemistries.

5. Results

5.1. 20th century corals

The results of the U/Th dating are shown in Table 2. The ^{232}Th concentrations for the modern coral and NB9 are about an order of magnitude higher than ^{232}Th concentrations reported for many reef-building corals (e.g. [2]). We show below that this ^{232}Th is associated with significant $^{230}Th_{nr}$ that makes a sample's calculated U/Th age older than the sample's 'true' age. To determine accurate ages, we must correct for this $^{230}Th_{nr}$ (using Eq. 3), so that a sample's 'corrected' U/Th age is closer to its true age. To make this correction without introducing large errors, we need some knowledge of $(^{230}Th/^{232}Th)_{nr}$.

The uncorrected U/Th dates (Table 2) reveal that fossil coral NB9 grew during the early 20th century. NB9 therefore overlaps the modern coral, whose climate record is absolutely dated (with a maximum error of 5 months at any point) back to 1886 AD through correlation to the instrumental record of climate. By matching the unique patterns of seasonal, interannual, and decadal climate patterns captured in the modern coral and NB9 oxygen isotopic records (Fig. 2), we obtain absolute dates for the fossil NB9. The highest correlation between the $\delta^{18}O$ records of the modern and fossil coral ($R = 0.80$) occurs when the NB9 $\delta^{18}O$ record spans from 1915 to 1937 AD – sliding the fossil sequence younger and older by 1–20 yr yields significantly lower correlations. It is important to note that while the NB9 $\delta^{18}O$ sequence ends at 1915, the absolute dates for samples NB9-2A and B were obtained by counting annual density bands back 5 yr to the location where these samples were taken.

Comparing the uncorrected U/Th ages with the

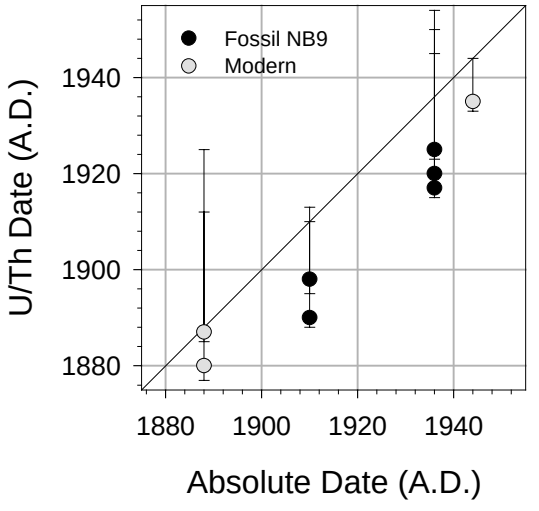

Fig. 3. A comparison of U/Th and absolute dates for the modern and NB9 corals. Lower error bars represent analytical error only (usually 2–3 yr), while upper error bars represent analytical error plus a maximum $^{230}Th_{nr}$ correction using a $(^{230}Th/^{232}Th)_{nr}$ ratio of 2×10^{-5}.

absolute ages for both the modern and the young fossil coral samples reveals that the uncorrected U/Th ages are generally older than the absolute ages (Fig. 3). In principle, this difference can be explained either by the presence of $^{230}Th_{nr}$ and/or the loss of parent ^{238}U and ^{234}U. In the case of modern corals, some evidence exists that the precipitation of petrologically visible secondary aragonite might actually add uranium to the system, not remove it [22]. On the other hand, corals submerged in freshwater for extended periods of time experience uranium loss [23], a possibility that could apply to the subaerially exposed Palmyra fossil corals. However, the modern submerged coral and the fossil subaerially exposed coral are both characterized by similar offsets between their U/Th and absolute ages in addition to high ^{232}Th concentrations. Therefore, we proceed on the assumption that $^{230}Th_{nr}$ is responsible for the U/Th age offsets. We use the absolute ages and the uranium and thorium isotopic analyses to calculate $(^{230}Th/^{232}Th)_{nr}$ using Eqs. 1–3.

For the modern coral, this calculation yields a range of $(^{230}Th/^{232}Th)_{nr}$ ratios with a minimum of 0 (in the case of Modern-1A, where the uncorrected U/Th and absolute ages agree to within analytical error), and a maximum of 2×10^{-5} (in the case of Modern-2, which requires a substantial correction). In Fig. 3, the lower error bar reflects analytical uncertainty alone, while the upper error bar reflects analytical uncertainty plus the $^{230}Th_{nr}$ correction that corresponds to a $(^{230}Th/^{232}Th)_{nr}$ of 2×10^{-5}. Therefore, while the size of the lower error bar remains relatively constant at $\sim$1–2%, the size of the upper error bar for a given sample is proportional to the concentration of ^{232}Th in that sample, given that increased ^{232}Th leads to a larger age correction (Eq. 3).

The $^{230}Th_{nr}$ corrections for the fossil coral NB9 yield $(^{230}Th/^{232}Th)_{nr}$ values that lie within the range defined by the modern coral results. The fact that the $(^{230}Th/^{232}Th)_{nr}$ values overlap for the modern and fossil corals suggests that the fossil coral did not undergo significant uranium loss, despite being exposed to $\sim$400 cm/yr of rainfall for 60 yr. If the fossil coral had experienced significant uranium loss during subaerial exposure, then larger age corrections, and hence higher $(^{230}Th/^{232}Th)_{nr}$ values, would be required to correct its U/Th ages, as compared to the modern coral. Furthermore, the ^{232}Th concentrations in coral NB9 do not exceed the highest ^{232}Th concentration measured in the modern coral (997 pg/g). Thus, there is no evidence that the fossil coral

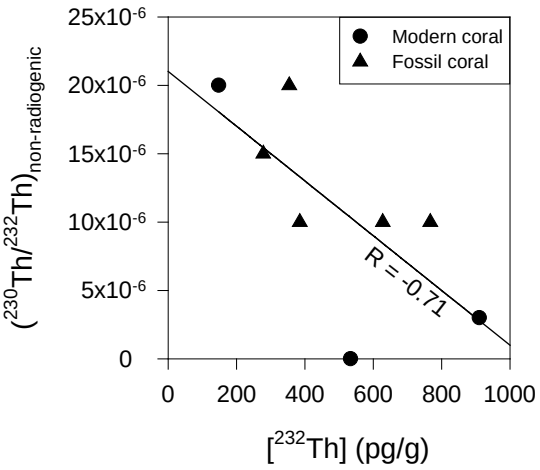

Fig. 4. Plot of ^{232}Th concentration vs. $(^{230}Th/^{232}Th)_{nr}$ for the eight 20th century U/Th samples. The $(^{230}Th/^{232}Th)_{nr}$ plotted here is the minimum value required to match each sample's absolute age within analytical error.

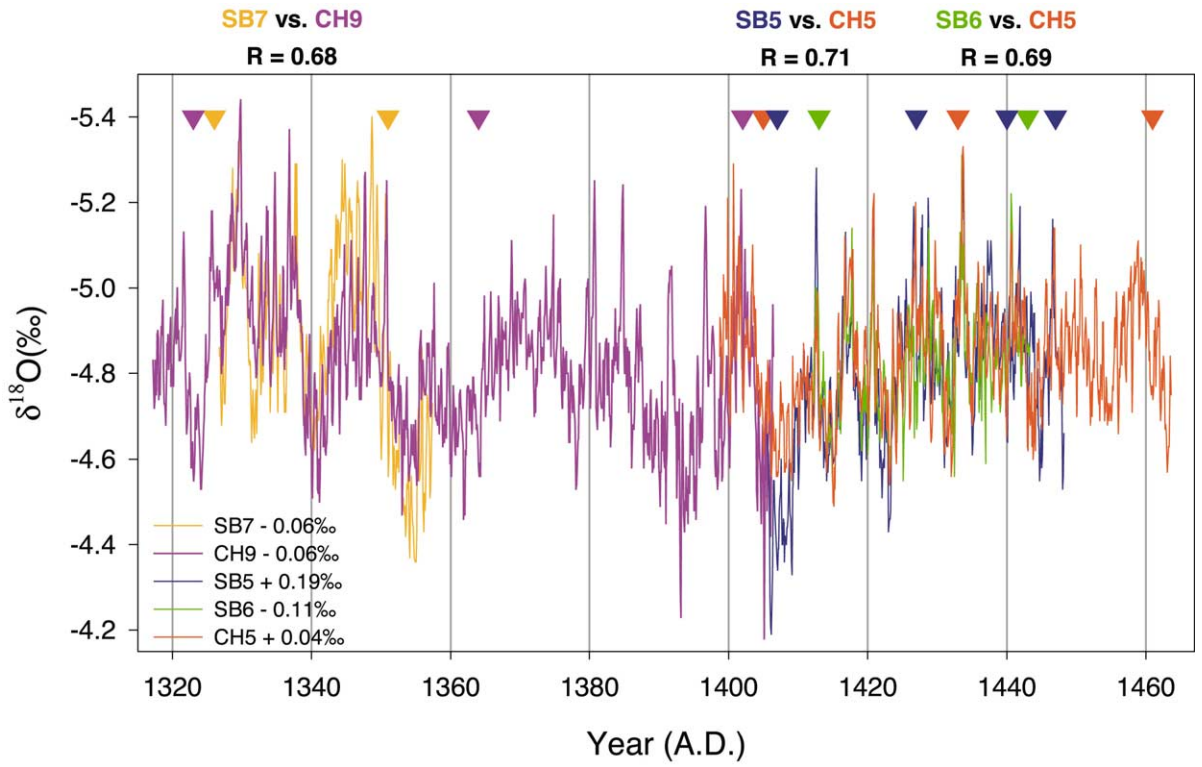

Fig. 5. The proposed overlap of five individual coral $\delta^{18}O$ records from the 14th–15th centuries. Correlation coefficients are shown on top for overlapping $\delta^{18}O$ sequences. The legend reports the offsets in $\delta^{18}O$ that were applied to each coral's $\delta^{18}O$ record to match the sequences as shown here. The triangles designate the locations that were sampled for U/Th dating.

underwent uranium loss and/or ^{232}Th addition during subaerial exposure – any uranium loss and/or ^{232}Th addition likely occurred while it was still living underwater. Within the limits of analytical precision, the young fossil coral remained closed with respect to its U/Th chemistry throughout its history of subaerial exposure.

It is interesting to note that we find that high ^{232}Th concentrations typically correspond to lower $(^{230}Th/^{232}Th)_{nr}$ values in both 20th century corals (Fig. 4). Such a relationship indicates that, at least in open-ocean environments, the addition of non-radiogenic Th may occur along a mixing line between high-[^{232}Th], low-$(^{230}Th/^{232}Th)_{nr}$ and low-[^{232}Th], high-$(^{230}Th/^{232}Th)_{nr}$ end-members. Empirically derived mixing lines between terrestrial sources of $^{230}Th_{nr}$ with low $(^{230}Th/^{232}Th)_{nr}$ values and 'hydrogenous' sources of $^{230}Th_{nr}$ with high $(^{230}Th/^{232}Th)_{nr}$ have previously been invoked to explain variability in $(^{230}Th/^{232}Th)_{nr}$ ratios in speleothems and deep-sea sediments [9,10,24,25]. Taken together, these studies hint that making a first-order approximation of sample $(^{230}Th/^{232}Th)_{nr}$ values by scaling to sample ^{232}Th concentrations may be warranted in certain environments.

5.2. 14th–15th century corals

Five overlapping fossil coral sequences spanning the 14th and 15th centuries provide an opportunity to test whether slightly older fossil corals exhibit U/Th chemistries similar to those observed for 20th century corals. In this case, we compare the 21 U/Th dates derived from the five corals to firm relative dates imposed by the overlap of the five corals' oxygen isotopic records. Ultimately, we derive so-called 'splice' dates for the overlapping coral sequence by minimizing the offsets between the relative and U/Th dates.

The first step involves overlapping the five coral $\delta^{18}O$ records in question, guided by U/Th dates. The samples with the lowest ^{232}Th concentrations provide the best dating constraints for each of the coral heads, whose $\delta^{18}O$ patterns are matched to within the age error of these low-^{232}Th samples. The resulting five-coral composite $\delta^{18}O$ record is shown in Fig. 5. Small coral-to-coral offsets in mean $\delta^{18}O$ are smaller than those observed in modern *Porites* corals from Clipperton Atoll [26] and in no way affect the dating results. By counting the years between U/Th sampling horizons, we obtain firm relative dates for the 21 U/Th samples that are precise to within 1 year. Ideally, the corals' U/Th dates, once corrected for the presence of $^{230}Th_{nr}$, would be entirely consistent with the corals' relative dates, to within analytical error. In this analysis, we allow the $(^{230}Th/^{232}Th)_{nr}$ ratio to vary across the range calculated for the 20th century corals ($0–2\times10^{-5}$). The corals' 21 'splice' dates, reported in year AD in the figures and tables, correspond to a best fit between the relative and the corrected U/Th dates (Fig. 6). The best-fit scenario is consistent with 15 of the 21 U/Th dates. Of the six remaining U/Th dates, four disagree with their assigned splice dates by 8–23 yr – substantial offsets that require explanation.

The observed offsets between the U/Th and splice dates mean that at least one of the following must be true: (1) the overlap between the five coral $\delta^{18}O$ records is inaccurate; (2) a $(^{230}Th/^{232}Th)_{nr}$ higher than 2×10^{-5} is required to correct U/Th dates that are too old; (3) uranium addition underlies the U/Th dates that are too young. With respect to the first possibility, the current configuration of the overlapping $\delta^{18}O$ records seems robust. The accuracy of the chosen overlaps is visually apparent (we cannot shift any one coral record by 1–20 yr to improve the match). Furthermore, in the cases where U/Th and splice dates do not agree, additional U/Th dates from the same coral head do agree with the splice dates. In other words, shifting one of the five corals younger or older to fix one mismatch would create two or three more mismatches. For the two samples whose U/Th dates are too old, invoking a slightly higher $(^{230}Th/^{232}Th)_{nr}$ value (up to 3×10^{-5}) would fix the problem. This seems justified, given

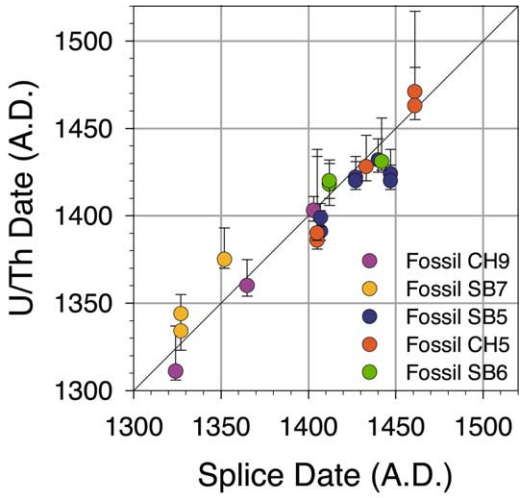

Fig. 6. A comparison of U/Th and 'splice' dates for samples representing the five coral sequences shown in Fig. 5. This figure illustrates the best-fit approach that we use to arrive at 'splice' dates for the composite $\delta^{18}O$ record shown in Fig. 5. One can evaluate the fit for alternative 'splice' dates by sliding the data points in this figure en masse horizontally along the splice-date axis. Error bars follow the conventions used in Fig. 3.

that the spread of $(^{230}Th/^{232}Th)_{nr}$ values for the eight 20th century samples may not capture the full range of $(^{230}Th/^{232}Th)_{nr}$ values in Palmyra corals. Lastly, in the case of the two samples whose U/Th dates are significantly too young, a 2–3% enrichment in uranium through secondary aragonite precipitation would explain the observed age discrepancies. At present, we cannot rule out such trace uranium addition by visual inspection of thin sections. Of course, a 2–3% error in the measurement of coral ^{238}U concentrations could also explain U/Th age offsets of 20 yr. However, the accuracy of TIMS and ICP-MS uranium measurements is monitored daily with uranium standards, and does not exceed $\pm2‰$ (2σ) [15,19].

Unlike the 20th century corals, the 14th–15th century fossil corals suggest that sample ^{232}Th concentrations and $(^{230}Th/^{232}Th)_{nr}$ values are uncorrelated. The fact that an eight-sample subset of the 21 14th–15th century samples can be constructed for which $[^{232}Th]$ and $(^{230}Th/^{232}Th)_{nr}$ are highly correlated ($R=-0.77$) implies that the results from the eight 20th century samples under-

represent the true variability in the $^{230}Th/^{232}Th$ system at our site. Indeed, it is difficult to explain the 14th–15th century $^{230}Th_{nr}$ correction results with simple mixing between wind-blown dust and ambient seawater thorium. Evidently, $^{230}Th_{nr}$ must be introduced into the coral skeleton through more complicated pathways and/or $^{230}Th/^{232}Th$ of the dust and seawater end-members must vary through space and time.

5.3. Absolute dating via proxy–proxy correlations

An alternative method for assigning calendar years to the spliced sequence is to correlate climate patterns in absolutely dated paleoclimate records (tree rings and ice cores) to the climate patterns in the fossil coral records. For this purpose we employ annually resolved, terrestrial proxy records from areas that exhibit teleconnections to the central tropical Pacific, namely the Quelccaya ice core from Peru and assorted tree ring records from the southwestern United States [27,28]. Such records typically reflect climate variability with respect to a single season, so we computed seasonal averages of the Palmyra corals for use in the proxy–proxy comparisons.

Correlation coefficients between the terrestrial proxy records and the Palmyra corals are significant above the 99% confidence interval in the 20th century, when dating errors are negligible. However, proxy–proxy correlations are insignificant in the 14th–15th century, even allowing for generous dating errors in both the coral and terrestrial proxy data. Consequently, we conclude that we cannot use these particular paleoclimate records in a proxy–proxy cross-correlation approach to improve upon the ± 5 yr dating error associated with our best-fit method.

5.4. U concentrations and isotope ratios

With our sampling strategy, it is impossible to detect the small-scale differences in uranium concentrations that might be expected for diagenetic gain or loss of uranium in the Palmyra corals. The reason is that U/Ca ratios could vary by 10–20% in response to the 1–2°C temperature changes that occur on interannual time scales at this site [23,29]. Even so, there is no visible evidence for wholesale leaching or precipitation of secondary aragonite in the Palmyra modern or fossil corals, as determined by analysis of thin sections. Therefore, significant uranium addition by continuous precipitation of secondary aragonite in submerged corals may only be favored in regions where carbonate-saturated groundwaters play an important role in coral pore-fluid chemistry (as documented in the Gulf of Eilat [22], for example).

High $\delta^{234}U(i)$ values are often used as an indicator of diagenetic alteration for fossil corals that are significantly older than those studied here [30–32]. However, the ages of the corals presented here (50–700 yr BP) are extremely young with respect to the half-life of ^{234}U ($\sim$245 000 yr), so $\delta^{234}U(i)$ is not a sensitive indicator of uranium addition or loss in these corals. In fact, we calculate a mean of 145.7 ± 1.6‰ (2σ) for all of our $\delta^{234}U(i)$ measurements, in striking agreement with the best estimate of modern seawater $\delta^{234}U$, 145.8 ± 1.7‰ (2σ) [15].

6. Discussion

Overall, the U/Th chemistries of the Palmyra corals support a limited role for uranium diagenesis in corals younger than 500 yr. This conclusion is supported by the fact that the modern and young fossil corals, characterized by markedly different diagenetic histories, display similar U/Th chemistries. Furthermore, while additional analyses are necessary for a more complete assessment, only two of the U/Th dates for the older fossil corals are consistent with uranium addition.

In both the 20th and 14th–15th century corals, non-radiogenic ^{230}Th is by far the largest source of U/Th dating error. Our results suggest that open-ocean surface corals, both living and fossil, may contain ^{232}Th concentrations up to 1000 pg/g, although many of the samples measured here fall within the few tens of pg/g range established by previous workers [1,2,33]. Given Palmyra's geographic location, these results suggest that open-ocean corals may be relatively protected from, but not immune from, continent-derived

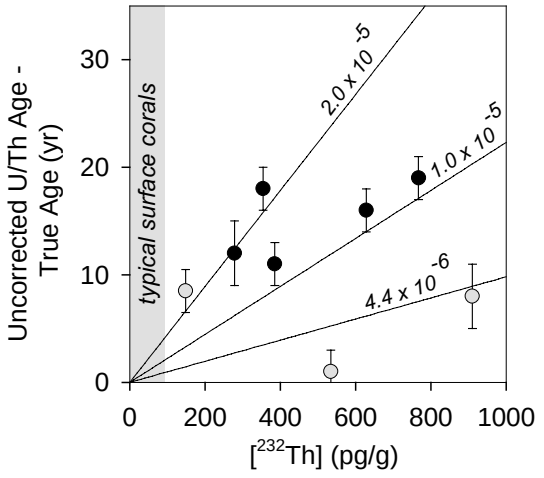

Fig. 7. Difference between uncorrected U/Th age and known, or absolute, age (y-axis) as a function of ^{232}Th concentration (x-axis). The diagonal lines indicate the magnitude of the correction for $^{230}Th_{nr}$ in years as a function of $(^{230}Th/^{232}Th)_{nr}$ ratio and ^{232}Th concentration, calculated assuming a ^{238}U concentration of 2500 μg/g. Our 20th century data (modern = gray circles, fossil = black circles) are plotted to illustrate the range of $(^{230}Th/^{232}Th)_{nr}$ ratios uncovered in this study. Error bars represent analytical error. The typical range of young coral ^{232}Th concentrations is indicated by the vertical shaded bar against the y-axis. Note that even for our highest documented $(^{230}Th/^{232}Th)_{nr}$ (2×10^{-5}), the correction for $^{230}Th_{nr}$ for such low ^{232}Th samples is no greater than 4 yr. Even for the somewhat elevated ^{232}Th concentrations documented above, the correction for $^{230}Th_{nr}$ can be made without introducing large errors, given knowledge of $(^{230}Th/^{232}Th)_{nr}$.

sources of ^{232}Th. The Th-bearing phases in the Palmyra corals have $(^{230}Th/^{232}Th)_{nr}$ ratios that range from 0 ($<4\times10^{-6}$) to 2×10^{-5}, exhibiting a range similar to that of the corals' ^{232}Th concentrations. Our results are in rough agreement with the results from the only other study of $(^{230}Th/^{232}Th)_{nr}$ ratios in surface corals, which documented a range of 0–1.3×10^{-5} in Sumatran corals [4].

If the $(^{230}Th/^{232}Th)_{nr}$ values of these two studies can be generalized, then corrections for $^{230}Th_{nr}$ are insignificant for corals with <100 pg/g ^{232}Th. For young corals with elevated ^{232}Th concentrations of hundreds of pg/g, $^{230}Th_{nr}$ may be significant and analyses of the sort undertaken here may be necessary to achieve dating errors of $\sim1\%$. Diagonal lines in Fig. 7 illustrate the magnitude of the correction for $^{230}Th_{nr}$ (in yr) for a ~100 yr old coral as a function of [^{232}Th] and $(^{230}Th/^{232}Th)_{nr}$ ratio (using Eqs. 1–3 and assuming a ^{238}U concentration of 2500 μg/g). We plot our 20th century results within this framework for reference. In the case of coral samples with less than 100 pg/g ^{232}Th (characteristic of many previously published analyses) corrections range from a few years to less than a year and are negligible. On the other hand, for a coral sample with 1000 pg/g ^{232}Th, the corrections range up to 40 yr, introducing a significant error for a 100 yr old coral but a $<1\%$ error for a 5000 yr old coral.

The range in $(^{230}Th/^{232}Th)_{nr}$ ratios observed at Palmyra is not surprising, given the variability in $^{230}Th/^{232}Th$ values for materials that potentially become integrated into the coral skeletons at Palmyra (see Section 3). In this context, any changes in wave activity, dust loading, and upwelling strength could affect the average $(^{230}Th/^{232}Th)_{nr}$ ratio in the coral. A better understanding of the mechanisms responsible for introducing $^{230}Th_{nr}$ into the Palmyra coral skeletons would require placing constraints on the temporal and spatial variability of Th sources to the corals, and the Th isotopic composition of those sources.

7. Conclusions

We have shown that some young reef-building corals in open-ocean environments can have relatively high concentrations of ^{232}Th, up to about 1000 pg/g. Even for corals with these unusually high ^{232}Th concentrations and very young ages of hundreds of years or less, it is possible to make accurate corrections for the presence of $^{230}Th_{nr}$. We have done so by employing a strategy that utilizes (a) multiple U/Th dates from individual corals and (b) firm relative dating constraints derived from overlapping the corals' $\delta^{18}O$ records to reduce the error on floating fossil coral chronologies from ±20 yr to ±5 yr. We also find that the fossil corals appear to have remained closed systems with respect to their U/Th chemistry for at least 700 yr of subaerial exposure. Our results show that highly precise, accurate U/Th dates can

be obtained for young fossil corals – dates that could potentially provide the basis for an extended, absolutely dated record of tropical climate change prior to the 20th century.

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