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High-precision U-series dating of corals from Western Australia and implications for the timing and duration of the Last Interglacial

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Abstract

U-series ages using methods of thermal ionisation mass spectrometry (TIMS) are reported for Last Interglacial fossil reefs along the stable coastal margin of Western Australia. Thorium isotope ratios were measured with superior precision using methods of charge collection. High levels of precision in the measurement of both uranium and thorium isotopes has reduced the age uncertainty due to analytical errors, excluding the uncertainty in the decay constants, by a factor of four over the precisions reported by many earlier TIMS workers. Uncertainties in $\delta^{234}U(T)$, determined from both $^{230}Th/^{238}U$ and $^{234}U/^{238}U$, are also significantly smaller than previously reported, allowing samples which have undergone diagenetic exchange of uranium and thorium to be more easily identified. Strict criteria were adopted to screen the new Western Australian data. Reliable ages range from 127 to 122 ka. Published TIMS observations from other localities have been assessed using the same strict criteria. When these are combined with glacio-hydro isostatic sea-level models they indicate that the Last Interglacial period occurred from at least 130 to 117 ka. However, these age constraints are largely determined from single data points and need to be verified with additional ages before considering them to be robust estimates for the timing of onset and termination of the Last Interglacial. Globally, the main episode of reef growth appears to be confined to a narrow interval occurring from 127 to 122 ka, in direct agreement with the narrow range in ages obtained from the Western Australian sites. This may indicate that the Last Interglacial was of short duration, extending from 127 to 122 ka only. Alternatively, this interval may reflect a major reef-building event in the middle of a longer duration (130–117 ka) interglacial interval.

1. Introduction

The height, age and stratigraphic relationships of shallow-water coral reefs have been widely used to constrain past sea levels. A particularly important set of reefs are those which formed during the Last Interglacial period, at about 125 ka. This interval represents the last time global sea levels were at or near modern levels, and by inference, the last time ice volumes and global climatic conditions were similar to present day. High-resolution sea-level information obtained from ~ 125 ka reefs has a direct bearing on Last Interglacial ice volumes and hence on the stability of today's major ice sheets [1], rheological properties of the mantle [1], the role of vertical tectonic processes since the Late Pleistocene

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[2], and the validity of the astronomical or Milankovitch model of climate change [3–5]. The Milankovitch model maintains that fluctuations in Pleistocene climate are driven by periodic changes in summer solar insolation in the Northern Hemisphere, caused by variations in the eccentricity of Earth's orbit and the tilt and precession of Earth's rotation axis. Recent insolation predictions for latitude 65°N show a peak between 128 ka [6] and 126 ka [7]. It is usually assumed that this marks the onset of the Last Interglacial period and the oldest corals present in an emergent Last Interglacial reef should have ages that are slightly younger than the 126–128 ka maximum.

The Milankovitch model has been widely used to assign a time scale to marine oxygen isotope curves which, at present, cannot be directly dated using radiometric techniques. However, several recent workers have presented evidence which appear to conflict with the accepted version of the model. In particular, U-series dating and oxygen isotope variations in a core of vein calcite from Devils Hole, Nevada, indicate that full interglacial conditions were reached near 135 ka, some 7–9 kyr before insolation peaked [8], and persisted for significantly longer than many of the orbitally tuned, deep-sea core chronologies suggest e.g. [9,10]. Some recent Last Interglacial sea levels determined from TIMS U-series dating of coral reefs also predate the Northern Hemisphere insolation maximum (e.g., [3,4,11]) whereas other reef localities show no evidence for high sea levels significantly before 128 ka (e.g., [5]). The unresolved question concerning the validity of the Milankovitch model has encouraged some workers to consider mechanisms other than just insolation forcing at 65°N as the driver of Pleistocene climate fluctuations [12,13].

The western margin of Australia is a passive continental margin which is thought to have been tectonically relatively stable since the Late Pleistocene and the fossil reefs can therefore provide critical information on the timing of the Last Interglacial, without requiring models of tectonic movements. Last Interglacial coral reefs feature prominently in the coastal stratigraphy of the Perth and Canarvon Basins. This is probably due to transportation of warm tropical water along the continental margin of Western Australia by the southerly flowing Leeuwin Current in the Late Pleistocene [14].

The recently developed TIMS U-series dating method [15] has significantly improved the precision and reproducibility with which coral reef ages can be determined, compared with earlier α-counting results. Additionally, through the precise determination of the initial $^{234}U/^{238}U$ ratio of the coral and hence seawater at the time of coral growth, the TIMS method has permitted a clearer evaluation of possible mobilisation of uranium and thorium during diagenesis.

Despite the new TIMS results, there remain conflicting estimates of the timing and duration of the Last Interglacial period and the details of sea-level fluctuations within this interval remain unresolved. For example, TIMS ages from the Reef VII complex at Huon Peninsula, Papua New Guinea [4], indicate that the Last Interglacial may have been characterised by two phases of rapid sea-level rise. The first episode may have culminated soon after 134 ka and the second near 118 ka, with apparently relatively little coral growth occurring during the 16 kyr interval separating the two events. In contrast, a detailed TIMS study of two fossil reefs in the Bahamas [3] indicates that the Last Interglacial consisted of a single phase of constant high sea level commencing near 130 ka and terminating near 120 ka. Similarly, a recent study of fossil reefs on Oahu, Hawaii [11] suggests that stable Last Interglacial sea levels may have persisted there from at least 131 to 114 ka.

These discrepancies may, in part, reflect difficulties in assessing the reliability of a ^{230}Th-age, as none of the existing mineralogic, textural and isotopic criteria for evaluating possible open-system behaviour of the U–Th isotopes are self-consistent. Constraints on the initial $^{234}U/^{238}U$ ratio, often reformulated as $\delta^{234}U(T)$ [15,16], are the only direct quantitative indicator of diagenesis of the coral skeleton. The effectiveness of placing tight limits on acceptable $\delta^{234}U(T)$ values is greatly reduced if the quality of the data is poor. For example, if a very strict $\delta^{234}U(T)$ criteria is employed, such that only measurements lying within 2‰ of the value for modern coral are considered acceptable [e.g., $\delta^{234}U(T)_{acceptable} = 149 \pm 2‰$], then the ^{230}Th-age of a coral with a $\delta^{234}U(T)$ value of $156 \pm 6‰$ may be regarded as "reliable". However, if the uncertainty in the measurement is reduced to $\pm 2‰$, the same coral is no longer considered to have a reliable age. Hence, the greater the precision in the $\delta^{234}U(T)$ measurement, the more stringent are the constraints that can be applied to the ^{230}Th-age.

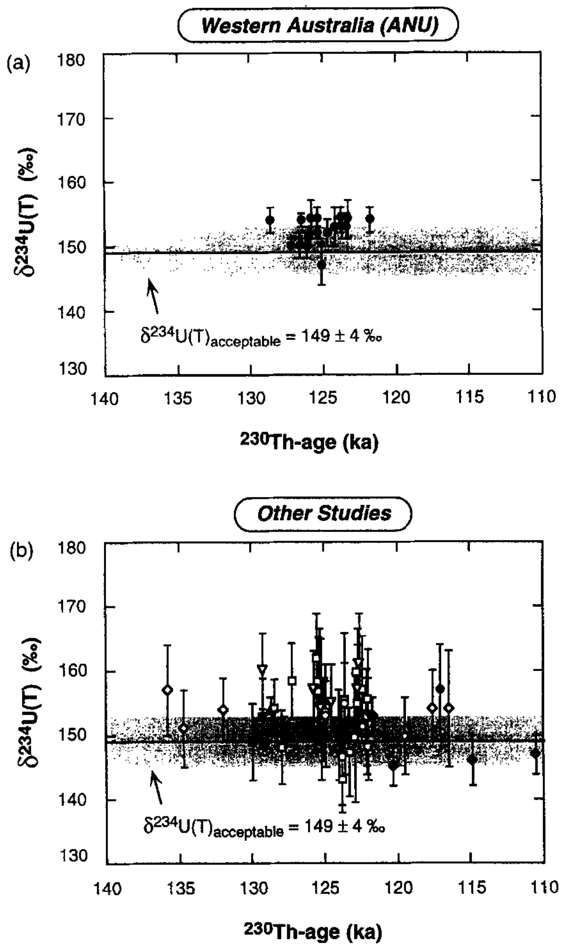

Fig. 1. Uranium isotopic composition $^{234}U/^{238}U$ at the time of coral growth T vs. ^{230}Th-age for (a) reliable samples from Western Australia, analysed at ANU, compared with (b) reliable samples analysed by other laboratories. We determined a $\delta^{234}U$ value of 148.9 ± 0.8‰ for modern coral, indicated by the black horizontal line. Samples with $\delta^{234}U(T)$ values within 4‰ of the modern value (shown by the shaded band) are considered to have ^{230}Th-ages which are accurate to within ± 1000 yr. Only these data are considered "reliable" and are plotted. Clearly, $\delta^{234}U(T)$ values with larger uncertainties are more likely to be considered acceptable as opposed to higher precision data. Therefore, the most precise measurements satisfying the most stringent $\delta^{234}U(T)$ criteria should be considered to give the most reliable ages. The superior precision in $\delta^{234}U(T)$ for the present study compared to previous studies is evident.

We present new U-series ages for Last Interglacial fossil reefs at Rottnest Island and Leander Point from the coast of Western Australia. Using improved techniques in thermal ionisation mass spectrometry (TIMS), we have achieved higher precision in Th isotope measurement. In addition, the level of precision we obtain for the measurement of U isotopes is significantly better than reported by many earlier TIMS workers (e.g., [3,17–19]). Hence, $\delta^{234}U(T)$ values, which are derived from both $^{234}U/^{238}U$ and $^{230}Th/^{238}U$ measurements, have uncertainties which are significantly improved (Fig. 1). When combined with a strict $\delta^{234}U(T)$ criteria, as discussed above, this allows samples which have undergone diagenetic exchange of uranium and thorium to be more easily identified and discarded; the remaining "acceptable" corals should give the most reliable chronology for the Last Interglacial. For ~ 125 ka samples, high levels of precision in the measurement of both U and Th isotopes has led to up to a factor of four reduction in the age uncertainty due to analytical errors, excluding the uncertainty in the decay constants. This corresponds to relative age uncertainties of ± 300 yr ($2\sigma_M$). Absolute age determinations, including the uncertainty in the decay constants, are accurate to within ± 800 yr ($2\sigma_M$). These improvements in age resolution have the potential to resolve the finer details of sea-level change during the Last Interglacial period. Also, Western Australia is characterised by relatively low rainfall over the past several hundred thousand years [20] so that there may be minimal mobilisation of U and Th due to percolation of freshwater through the fossil reefs.

2. Experimental methods

2.1. Sample preparation and chemistry for U-series dating

We preferentially select corals from the Faviidae family for U-series dating, in contrast to species of *Acropora* and *Porites*, as they are characterized by well-developed wall structures of dense aragonite which are generally free of detrital and recrystallized material. These relatively pristine walls can easily be separated from the septa fraction with a diamond drill. X-ray diffraction, thin-section and U–Th isotopic analyses show that the coral septa often contain substantial amounts of secondary calcite and detrital ^{232}Th and therefore are likely to have been diagenetically altered. Two samples were treated with a series of acid washes in an attempt to remove remaining surface contaminants from the coral skeleton.

Approximately 1 g of coral is ultrasonically cleaned in distilled water, dissolved by the step-wise addition of concentrated HNO_3, then spiked with a mixed ^{229}Th–^{233}U tracer. Concentrated H_2O_2 is added to help oxidise any organic matter and the solution dried to ensure complete sample-spike equilibration. Procedures for the separation of U and Th from the sample are based on methods that are similar to those described in Edwards et al. [15].

2.2. Measurement of uranium isotopes

Measurements were made on a Finnigan MAT 261 thermal ionisation mass spectrometer using a double filament assembly with a multi-collector detector system for the simultaneous measurement of ^{238}U, ^{235}U, ^{234}U and ^{233}U ion beams. Faraday cups and electrometers with 10^{11} Ω feed-back resistors are used to measure ^{238}U, ^{235}U and ^{233}U currents (1.5×10^{-11} A, 1×10^{-13} A and $(3–4) \times 10^{-14}$ A, respectively). The low-level ^{234}U ion beam ($\sim 10^{-15}$ A) is measured simultaneously using a secondary electron multiplier (SEM) with a gain of ~ 10,000 and an electrometer with a 10^9 Ω feed-back resistor. The SEM-Faraday cup relative gain is measured at frequent intervals during a run by comparing the measured $^{238}U/^{235}U$ ratio when ^{235}U is located in the SEM and ^{238}U is located in a Faraday cup to that when both ^{238}U and ^{235}U are positioned in Faraday cups.

Measured $^{234}U/^{235}U$ and $^{233}U/^{235}U$ ratios are corrected for isotopic mass fractionation, using the measured $^{235}U/^{238}U$ ratio normalised to the natural value (137.88), with appropriate corrections for the contributions of ^{234}U, ^{235}U and ^{238}U present in the ^{233}U spike. The $^{234}U/^{238}U$ ratios are calculated using the measured $^{234}U/^{235}U$ ratio and the natural $^{238}U/^{235}U$ value. For a single run, $2\sigma_M$ for the $^{234}U/^{238}U$ ratio is typically 1–2‰ and is up to a factor of 10 better than the precision reported from some other laboratories (Fig. 2a).

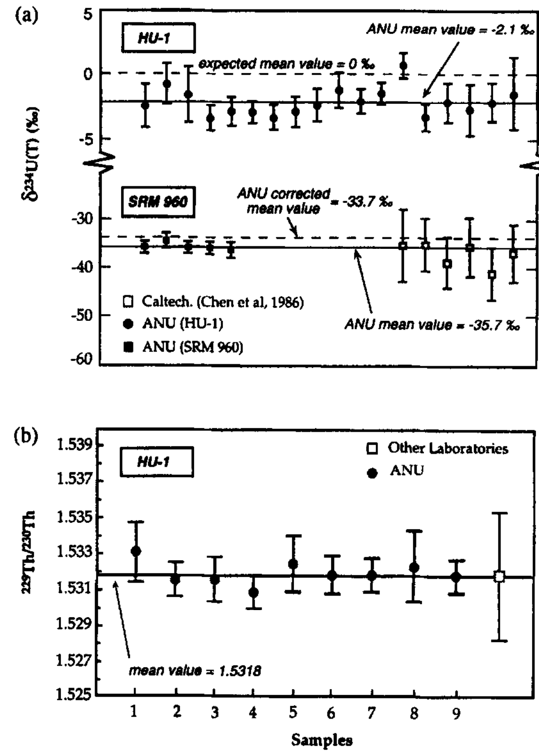

Fig. 2. Repeat measurements of (a) $^{234}U/^{238}U$ and (b) $^{229}Th/^{230}Th$ on standards to determine the external reproducibility of uranium and thorium measurements. $\delta^{234}U(T)$ measurements were performed on the uraninite standard HU-1 (circles) and the NBS SRM 960 uranium standard (squares). Solid symbols denote ANU results. Open symbols denote results in Chen et al. [15]. All errors are $2\sigma_M$. The ANU data for $\delta^{234}U(T)$ are offset from the expected zero value by −2.1‰ which we attribute to a systematic machine bias in Finnigan MAT 261 in the SEM-Faraday cup configuration used for the present experiments. With this correction, $\delta^{234}U(T)$ values for both SRM 960 and modern coral are identical within error to results obtained from other laboratories. Note that the $\delta^{234}U(T)$ values for HU-1 are plotted at a smaller scale than those for SRM 960. The $^{229}Th/^{230}Th$ measurements were performed on a spiked HU-1 solution. The weighted mean $^{229}Th/^{230}Th$ ratio is 1.5318. Solid circles denote ANU results. The error bars are σ_M and are approximately equal to the $2\sigma_M$ statistical uncertainty. The open square shows the best measurement for other laboratories which has been scaled so that $^{229}Th/^{230}Th = 1.5318$. The error bar is $2\sigma_M$.

2.3. Measurement of thorium isotopes

Thorium is present in corals only at ppb levels and is difficult to ionize. Ion beams produced by thermal ionization do not persist for long periods or in sufficient intensity for high-precision measurement using Faraday cups and electrometers with high-value resistors. Previous workers have instead employed peak hopping using a single electron multiplier [15] or a Daley detector [21]. Although these techniques enable measurement of low-intensity ion beams ($< 10^{-14}$ A), they are limited by fluctuations in beam intensity and have poor data-acquisition efficiency.

We have employed a fundamentally new technique to measure thorium isotope ratios [22]. Measurements were made on the 61-cm thermal ionization mass spectrometer in "charge collection" mode. Th sample fractions were loaded on the filament between two layers of graphite and were run using the single-filament technique. In charge mode, the usual high-value feed-back resistor (10^{11} Ω) is replaced with a 20 pico-Farad air-core capacitor. This technique allows the measurement range to be extended below 10^{-14} A, while preserving the advantages of Faraday cup arrays and for the first time it is possible to measure the low-level currents (less than 10^{-16} to 10^{-13} A) of ^{232}Th, ^{230}Th and ^{229}Th simultaneously, in multiple channels with high precision. Typical background currents are $(1–5) \times 10^{-17}$ A and correspond to < 100 units of injected charge per second. These levels are about a factor of 500 lower than for a 10^{11} Ω resistor.

Provided that the ion beam is stable, charge builds up on the capacitor C with time t in a linear fashion and is displayed in analogue mode as a monotonic increase in voltage V with every 1.28 s of peak integration time. Signal integration times for each channel are equal and synchronous. One block of data consists of 60 voltage readings and represents approximately 1.5 min of data acquisition time. The rate of voltage accumulation $\mathrm{d}V/\mathrm{d}t$ is related to the ion current I according to $I = C(\mathrm{d}V/\mathrm{d}t)$. The slope of the best-fit straight line through the data therefore represents the mean current for that 1.5 min interval. 1σ errors in $\mathrm{d}V/\mathrm{d}t$ are estimated from the quality of fit for the 60 data points. Once the background accumulated charge has been removed from the peak signal, the atomic ratio for any isotope pair is directly equal to the ratio of their $\mathrm{d}V/\mathrm{d}t$ values.

Using charge collection, the uncertainty in the ^{229}Th/^{230}Th isotope ratio for one block of data, collected over 1.5 min, is about $\pm 2.5‰$ and is a similar to the best precision in Th isotope ratios previously reported for a whole run [15]. Depending on the duration of the experiment, 20–70 blocks can be measured and for each isotope pair, the grand mean for the run is the weighted average of all block ratios. The internal statistical precision for a 45 min data acquisition period is typically 0.6‰ (σ_M) and can be achieved routinely for 15 pg of ^{230}Th for both standards and thorium separated from corals. This is up to a factor of ten better than uncertainties typically reported from other laboratories [3,5,17,21].

Mass spectrometric duplicate analyses of a spiked aliquot of a solution of Precambrian uraninite (HU-1) that has been shown to be in, or very close to, secular equilibrium for ^{234}U/^{238}U and ^{230}Th/^{238}U ratios [23], were performed to assess the importance of the statistical component in the quoted uncertainty (Fig. 2b). Th loads consisted of 15 pg of ^{230}Th and 25 pg of ^{229}Th. The total thorium loaded on the filament, including ^{232}Th, is about 100 pg. Each of the nine repeat runs have a ^{229}Th/^{230}Th ratio identical, within error, to the mean ^{229}Th/^{230}Th ratio of 1.5318. For measurements conforming to gaussian statistics we expect only two thirds to be identical within errors at the 1σ tolerance level which suggests that the quoted σ_M error overestimates the analytical uncertainty in the measured Th isotope ratios. For this reason, the uncertainties in ^{229}Th/^{230}Th have not been doubled as is usual when errors in the mean are quoted.

At the start of a run, the ^{230}Th current is typically 3×10^{-15} A for approximately 15 pg of ^{230}Th loaded on the filament, but typically grows to more than 6×10^{-14} A as the run proceeds and often remains at this intensity for more than one hour. Th peaks are centred in the Faraday cups by using the 10^{12} Ω resistors. The ionization efficiency ranges from 1 to 6%, a significant improvement over that reported by other laboratories (e.g. [15]). Ionization efficiency remained unchanged when the ^{230}Th load was increased from 15 pg to more than 45 pg. Isotopic mass fractionation of thorium appears insignificant compared with the statistical uncertainty in the measurements throughout the 1–1.5 h duration of a Th-run. We have not observed an appreciable shift in the ^{229}Th/^{230}Th during the course of a run. No evidence was found for reflected beams in the vicinity of the Th isotopes.

2.4. Calibration of the mixed spike

We have used a mixed spike solution of ^{229}Th and ^{233}U for the measurement of naturally occurring thorium and uranium in corals by isotope dilution. Separate ^{229}Th and ^{233}U spikes were obtained in nitrate form from Oak Ridge National Laboratory and New Brunswick Laboratories, respectively. The mixed spike solution is stored in a teflon bottle with a trace amount of HF to prevent thorium from precipitating out of solution. The isotopic composition of the mixed spike was determined to be $^{238}U/^{235}U = 2.8937$, $^{234}U/^{235}U = 2.3378$, $^{233}U/^{235}U = 1259.38$, and $^{229}Th/^{230}Th = 20{,}000$. Last Interglacial samples were spiked so that $^{233}U/^{235}U \sim 0.3$–0.4 and $^{230}Th/^{229}Th \sim 3$–5.

The secular equilibrium standard HU-1 was used to calibrate the $^{233}U/^{229}Th$ ratio of the mixed spike. Because a ratio rather than concentrations are being calibrated, HU-1 has several advantages over gravimetric standards as a spike calibration tool. Firstly, possible systematic biases arising from errors in weighing during the preparation of the gravimetric standard are eliminated. Secondly, systematic biases resulting from errors in the determination of the decay constants are reduced. Mass spectrometric analyses for ten, separately equilibrated, spiked HU-1 solutions gave a mean $^{233}U/^{229}Th$ ratio of 53.18 ± 0.05 $(2\sigma_M)$. The $2\sigma_M$ uncertainty for each experiment ranged from ± 0.6 to ± 1.0‰. Only one of the ten experiments gave a $^{233}U/^{229}Th$ ratio which differed from the mean value by more than 1‰. Several

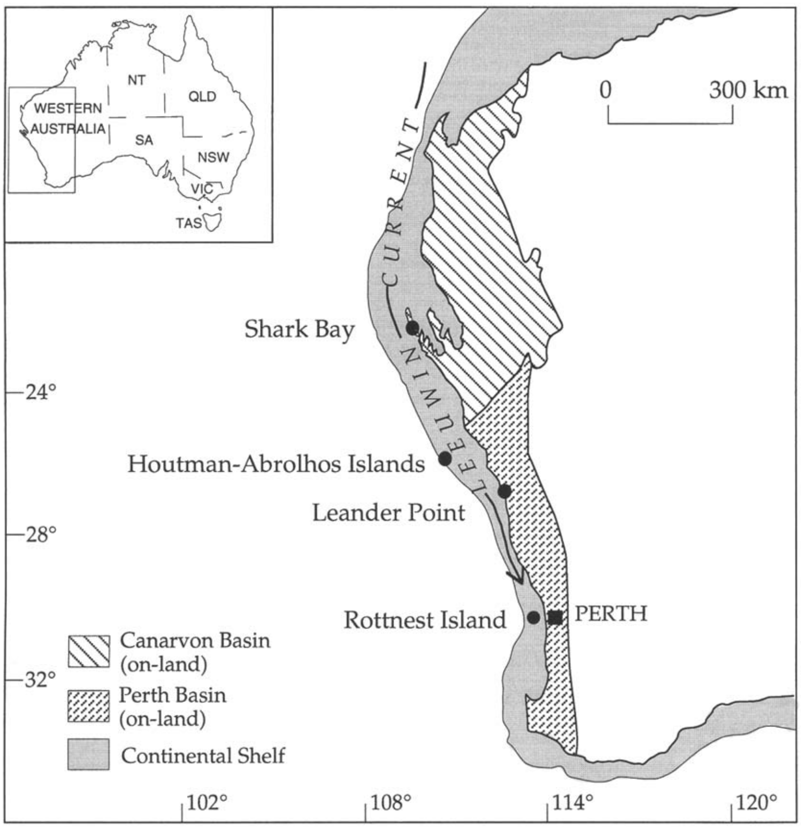

Fig. 3. Location of sites referred to in the text. Rottnest Island is located 18 km off-shore from Perth near the outer edge of the continental shelf at latitude 32°01′S. Leander Point is situated at latitude 29°18′S along the west coast of mainland Australia. For the present study, 7 samples were collected from in-situ coral within the Last Interglacial Rottnest Limestone [26] and a further 10 samples were collected from in-situ coral within the fossil reef complex at Leander Point. The Houtman–Abrolhos Islands, comprise a series of coral reefs located on the outer part of the continental shelf, approximately 70 km off-shore from Leander Point.

solutions were fumed with concentrated $HClO_4$, a process thought to promote sample-spike equilibration [15]. These solutions yielded $^{233}U/^{229}Th$ ratios which were not appreciably different to those which did not undergo $HClO_4$ fuming. Instead, we have regularly used H_2O_2 to achieve equilibration. The concentrations of each spike, although not necessary for determining a ^{230}Th-age, were determined using the absolute values of the National Bureau of Standards (NBS) SRM 960 uranium standard. The spike concentration values are only necessary for determining the concentrations of ^{238}U and ^{232}Th in coral samples. These are are useful, but not quantitative, indicators of the extent of diagenetic exchange of uranium and thorium in the sample.

2.5. Calibration of $^{234}U/^{238}U$ measurements

HU-1 was also used to calibrate $^{234}U/^{238}U$ measurements which provides an independent estimate for machine bias. From 18 experiments, the mean measured $\delta^{234}U$ value for HU-1 is $-2.1 \pm 0.5‰$ ($2\sigma_M$) (Fig. 2a). All but one of the measurements lies within 1‰ of the mean value. These data are offset from the expected zero value for a sample with ^{234}U and ^{238}U in secular equilibrium. This could indicate that HU-1 is not in fact in secular equilibrium. However, two additional and independent checks for $\delta^{234}U$ imply that the offset reflects a machine bias in Finnigan MAT 261 in the SEM-Faraday cup configuration used for the present experiments. Firstly, six measurements of $\delta^{234}U$ for SRM 960 give a mean value of $-35.7 \pm 0.7‰$ ($2\sigma_M$). When corrected for the 2‰ offset, a value of $-33.7 \pm 0.7‰$ ($2\sigma_M$) is obtained which is identical, within error, to the $\delta^{234}U$ value of $-34.1 \pm 1.2‰$ reported for SRM 960 in Edwards et al. [24]. Secondly, we determined a corrected $\delta^{234}U$ value of $148.9 \pm 0.8‰$ ($2\sigma_M$) for modern coral which is not statistically different to the value of $149.7 \pm 1.5‰$ ($2\sigma_M$) established by Gallup et al. [5]. If HU-1 were not in secular equilibrium then we would not expect our offset corrected $\delta^{234}U$ values for both SRM 960 and modern corals to be in agreement with those measured by other laboratories. When calculating ^{230}Th-ages for samples, we therefore normalised our measured $^{234}U/^{238}U$ ratio relative to the corresponding value measured for HU-1. These tests help confirm that HU-1 is in secular equilibrium and is therefore appropriate to use as a calibration standard.

2.6. Procedural blank

The total procedural blank including the filament loading blank is 0.7 ± 0.2 pg of ^{238}U and 5 ± 2 pg of ^{232}Th for filament loads of approximately 1 μg of ^{238}U and 15 pg of ^{230}Th, respectively. We assume that blank levels of ^{234}U are $\sim 10^4$ times less than ^{238}U and those of ^{230}Th are $\sim 10^5$ times less than ^{232}Th. These levels have a negligible effect on U and Th measurements for Last Interglacial samples.

3. Samples

For the present study, 18 corals in apparent growth position within Last Interglacial reefs at Rottnest Island and Leander Point, Western Australia, were sampled for U-series dating. Rottnest Island is located 18 km off-shore from Perth near the outer edge of the continental shelf at latitude 32°01′S (Fig. 3). The Last Interglacial reef complex outcropping at Fairbridge Bluff is referred to as the Rottnest Limestone [25] and represents Australia's southernmost Last Interglacial reef. It forms a thin unit within the Pleistocene–Early Holocene Tamala Limestone [26] which is an extensive aeolian deposit, outcropping from Shark Bay (Fig. 3) to the south coast. The Rottnest Limestone has been dated by α-counting U-series techniques at 100 ± 20 ka [27] and 132 ± 5 ka [28]. The Last Interglacial reef at Leander Point, situated approximately 400 km north of Rottnest Island at latitude 29°18′S (Fig. 3). The ~ 125 ka reef platforms at both sites extend to approximately 3 m above the modern intertidal platforms, indicating that sea level during the Last Interglacial maximum was at least 3 m higher with respect to Rottnest Island and Leander Point than it is today.

4. Results

Species, mineralogy and elevation together with U–Th isotopic data and ^{230}Th ages of coral samples recovered from the Last Interglacial reefs at Rottnest Island and Leander Point are listed in Table 1.

A comparison of the U–Th results between whole coral and wall fractions shows that removal of the septa fraction has had an apparently small effect on the U–Th isotope systematics and a negligible effect on ^{230}Th-age. This indicates that significant U and Th mobilisation has not accompanied the deposition and precipitation of contaminants in the coral skeleton. The U isotopic composition of acid leached and unleached fractions of RN-FB-7 and LP-91-1 agree within errors, indicating that either leaching is inef-

Table 1
U–Th isotope ratios and ^{230}Th of Rottnest Island and Leander Point corals

Sample†	Coral	Elevation (m) #	^{232}Th (ppb) @	^{238}U (ppm)	δ^{234}U(0) (‰) ‡	δ^{234}U(T) (‰) ‡	(^{230}Th/^{238}U)$_{act}$ $	Age (ky) ¶
Rottnest Island								
RN-FB-1 ^{w}	Faviidae	0.21	1.03	3.10	114 ± 1	164 ± 1	0.7792 ± 0.0007	126.6 ± 0.8
RN-FB-2 ^{w}	Faviidae	0.11	1.16	2.84	116 ± 2	167 ± 2	0.7832 ± 0.0010	127.3 ± 0.9
RN-FB-3 ^{w}	Faviidae	0.45	0.10	2.83	107 ± 1	152 ± 1	0.7695 ± 0.0005	**125.5 ± 0.8**
RN-FB-4 ^{w}	Faviidae	1.77	0.12	2.79	105 ± 1	150 ± 2	0.7702 ± 0.0005	**126.2 ± 0.8**
RN-FB-5 a ^{w}	Acropora	1.80	2.60	3.38	116 ± 1	162 ± 2	0.7457 ± 0.0011	116.6 ± 0.8
RN-FB-5 b ^{w}	Acropora	1.80	-	3.26	110 ± 1	150 ± 2	0.7127 ± 0.0018	109.0 ± 0.8
RN-FB-5 c ^{w}	Acropora	1.80	-	3.41	108 ± 1	154 ± 1	0.7686 ± 0.0017	124.9 ± 0.9
RN-FB-6 ^{w}	Faviidae	1.83	0.19	3.15	108 ± 1	154 ± 2	0.7700 ± 0.0007	**125.4 ± 0.9**
RN-FB-7 a ^{w}	Faviidae	2.43	0.30	2.80	105 ± 1	150 ± 1	0.7739 ± 0.0018*	**127.3 ± 1.0**
RN-FB-7 b ^{w}	Faviidae	2.43	0.25	2.75	105 ± 1	151 ± 2	0.7702 ± 0.0005	**126.0 ± 0.8**
RN-FB-7 c ^{s,L}	Faviidae	2.43	-	2.70	105 ± 3	-	-	-
Leander Point								
LP-91-1 a ^{w}	Faviidae	2.16	3.38	2.67	111 ± 1	158 ± 2	0.7682 ± 0.0043*	124.1 ± 1.5
LP-91-1 b ^{s}	Faviidae	2.16	0.33	2.68	105 ± 1	150 ± 2	0.7716 ± 0.0010	**126.6 ± 0.9**
LP-91-1 c ^{s,L}	Faviidae	2.16	-	2.70	108 ± 2	-	-	-
LP-91-2 a ^{w}	Faviidae	0.97	-	3.01	105 ± 1	-	-	-
LP-91-2 b ^{s}	Faviidae	0.97	1.01	2.80	112 ± 1	160 ± 2	0.7754 ± 0.0008	126.1 ± 0.8
LP-93-3 ^{s}	Faviidae	1.67	0.15	2.86	109 ± 1	154 ± 2	0.7590 ± 0.0007	**121.8 ± 0.8**
LP-93-4 ^{s}	Faviidae	1.28	0.84	2.98	108 ± 1	153 ± 2	0.7643 ± 0.0009	**123.7 ± 0.8**
LP-93-5 ^{s}	Faviidae	0.70	0.16	2.99	109 ± 2	154 ± 2	0.7654 ± 0.0010	**123.8 ± 0.9**
LP-93-6 ^{s}	Faviidae	1.80	0.05	2.87	109 ± 2	154 ± 3	0.7638 ± 0.0010	**123.3 ± 0.9**
LP-93-7 ^{w}	Acropora	1.81	0.37	3.68	126 ± 1	177 ± 2	0.7688 ± 0.0007	121.0 ± 0.8
LP-93-8 ^{s}	Faviidae	0.68	0.02	2.76	108 ± 2	153 ± 3	0.7660 ± 0.0010	**124.2 ± 0.9**
LP-93-9 ^{s}	Faviidae	1.09	0.02	2.76	115 ± 1	163 ± 2	0.7715 ± 0.0007	124.2 ± 0.8
LP-93-10 ^{s}	Faviidae	2.20	0.07	2.89	108 ± 2	153 ± 2	0.7632 ± 0.0008	**123.4 ± 0.9**
Living Coral								
Huon-1	Porites	-	-	-	148 ± 2	148 ± 2	-	-
Huon-2	Porites	-	-	-	149 ± 2	149 ± 2	-	-
GBR-1	Porites	-	-	-	149 ± 1	149 ± 1	-	-

fective in removing surface contaminants or all surface contaminants in RN-FB-7 and LP-91-1 were effectively removed by drilling out the septa prior to acid leaching. Similar conclusions have been reached previously from leaching experiments on Last Interglacial corals from Curacao, Haiti and Barbados [29].

4.1. Reliability of ^{230}Th-ages

Samples that have (1) > 99% aragonite, (2) minimal calcite infilling or replacement, (3) approximately 3 ppm total uranium, consistent with concentrations in modern corals, and (4) low ^{232}Th concentrations, are likely to have remained a closed system with respect to uranium and thorium since the time of coral growth. X-ray diffraction and thin section analyses show that the calcite content of all sampled corals, except LP-91-1a, is less than 1%. Concentrations of ^{238}U range from 2.7 to 3.7 ppm, comparable to levels reported by other workers for Last Interglacial corals [17,19,29,30] and consistent with ^{238}U concentrations in modern corals [15,29,31]. Six samples have ^{232}Th concentrations which exceed the 0.5 ppb limit specified by corals from oceanic islands [3,15]. Anomalously high ^{232}Th concentrations were also recorded in Pleistocene and Holocene corals from the Abrolhos Islands [19]. No attempt has been made to correct the influence of high ^{232}Th content on the ^{230}Th-age, as the correction is generally less than the error in the age.

Extensive efforts have been made to correlate petrography, trace element concentrations and stable isotopes with diagenesis [4,32] but criteria derived from these measurements appear unable to detect low levels of U–Th mobilisation. Therefore, following earlier workers (e.g., [3,15]), we consider $\delta^{234}U(T)$ to be the quantitative test of alteration of the coral skeleton. Recent isotopic studies of Last Interglacial corals [5,11], together with steady state models allowing for the total input and residence time of uranium in the oceans [16,33], indicate that at 125 ka, the marine $^{234}U/^{238}U$ ratio was probably not significantly different to that of today, although small (several permil) shifts can not be ruled out. Therefore, it is generally assumed that fossil corals with $\delta^{234}U(T)$ values coincident with those for modern open ocean water have undergone minimal diagenetic exchange of uranium and thorium and hence have ^{230}Th-ages which reflect the true age of the sample [3–5,11,15,19]. We determined a $\delta^{234}U$ value of 148.9 ± 0.8‰ ($2\sigma_M$) for living coral (Table 1) by averaging the $\delta^{234}U$ values of two modern samples from Huon Peninsula, Papua New Guinea, together with one modern sample from the Great Barrier Reef, Australia. This is identical to the $\delta^{234}U$ value obtained by other workers for modern coral [3,5,11,30] and to the $\delta^{234}U$ range of 140–150‰ measured for present-day seawater [16].

Recent TIMS U-series data from fossil corals in Barbados [5] show a rough linear trend between

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Notes to Table 1:

† *a*, *b* and *c* indicate replicate analyses on different coral fragments from the same sample. *w* and *s* denote whole coral and wall fractions respectively. Acid leached fragments are denoted by *L*.

Elevations for Rottnest Island and Leander Point samples are in m above the modern intertidal platform.

@ Error in ^{232}Th is dominated by the uncertainty in the ^{232}Th blank correction (5 ± 2 ρg).

‡ $\delta^{234}U = \{[(^{234}U/^{238}U)/(^{234}U/^{238}U)_{eq}] - 1\} \times 10^3$. $(^{234}U/^{238}U)_{eq}$ is the atomic ratio at secular equilibrium and is equal to $\lambda_{238}/\lambda_{234} = 5.472 \times 10^{-5}$ where λ_{238} and λ_{234} are the decay constants for ^{238}U and ^{234}U, respectively. $\delta^{234}U(O)$ is the measured value, the initial value is given by $\delta^{234}U(T) = \delta^{234}U(O)e^{\lambda 234 T}$, where T is the age in years. Acceptable samples have $\delta^{234}U(T)$ values overlapping the range 149 ± 4‰.

$ $(^{230}Th/^{238}U)_{act} = (^{230}Th/^{238}U)/(\lambda^{238}/\lambda^{230})$.

* Th analysed on Finningan MAT 261, resulting in larger errors in $(^{230}Th/^{238}U)_{act}$.

¶ ^{230}Th-ages are calculated iteratively using:

$$1 - \left[\frac{^{230}Th}{^{238}U}\right]_{act} = e^{-\lambda_{230}T} - \left(\frac{\delta^{234}U(O)}{1000}\right)\left(\frac{\lambda_{230}}{\lambda_{230} - \lambda_{234}}\right)\left(1 - e^{(\lambda_{234} - \lambda_{230})T}\right)$$

where T is the age in years and λ_{230} is the decay constant for ^{230}Th. $\lambda_{238} = 1.551 \times 10^{-10}\ yr^{-1}$; $\lambda_{234} = 2.835 \times 10^{-6}\ yr^{-1}$; $\lambda_{230} = 9.195 \times 10^{-6}\ yr^{-1}$. Uncertainty in the age is largely due to errors in the decay constants which contribute 800 yr to $2\sigma_M$. Ages in bold type are considered "reliable".

increasing δ^{234}U(T) and ^{230}Th-age. In addition, no δ^{234}U(T) measurements lower than the modern marine value were documented. These data are consistent with a diagenetic model whereby ^{234}U and ^{230}Th are added continuously to the coral skeleton, assuming an initial δ^{234}U identical to modern seawater [5]. The model indicates that a sample with a $\delta^{234}U(T)$ exceeding the marine value by four units has a ^{230}Th-age about 1000 yr older than its true age. More than 60 Last Interglacial samples from Western Australia (this study, unpubl. ANU results, and data from the Abrolhos Islands [19,34]) support the diagenetic model of Gallup et al. [5]. Although exceptions do occur (e.g., LP-93-7), a broadly linear relationship exists between ^{230}Th-age and $\delta^{234}U(T)$ and no initial δ^{234}U have values less than the modern marine value. It thus seems likely that similar diagenetic processes are occurring in both Western Australia and Barbados and following Gallup et al. [5], we consider that samples with $\delta^{234}U(T) = 149 \pm 4$ have ^{230}Th-ages accurate to within ± 1000 yr. Only samples satisfying this strict $\delta^{234}U(T)$ criteria are considered to have reliable ^{230}Th-ages.

Despite placing strict limits on acceptable $\delta^{234}U(T)$, we recognise that the δ^{234}U(T) criterion, although essential, is not entirely self consistent. Samples which are clearly unreliable can give acceptable δ^{234}U(T) values. For example, Chen et al. [3] measured acceptable $\delta^{234}U(T)$ in samples which give inconsistent age results. Similarly, in this study, ^{230}Th-ages for three separate fragments of RN-FB-5, a species of *Acropora*, span a 16 kyr interval, but two of the fragments have acceptable $\delta^{234}U(T)$ values. The lack of reproducibility of these data is most likely due to the presence of visible amounts of non-carbonate detrital material in the pore spaces of the coral skeleton which is correlated with anomalously high levels of ^{232}Th. In addition, departures of $\delta^{234}U(T)$ from the modern seawater value need not indicate significant mobilisation of the U–Th isotopes. For example, results from this study show that ^{230}Th-ages for samples with anomalous $\delta^{234}U(T)$ values, are not appreciably different to those for samples with acceptable $\delta^{234}U(T)$.

Excluding RN-FB-5, 18 ^{230}Th-ages are reported for the Rottnest Island and Leander Point reefs. Of these, twelve pass all the abovementioned criteria employed to investigate possible open system behaviour of U and Th. "Acceptable" ages range from 127 to 122 ka.

TIMS U-series ages are available for two other Last Interglacial reefs along the west coast of Australia; the Houtman–Abrolhos Islands, which comprise a series of coral reefs located on the outer part of the continental shelf, approximately 70 km offshore from Leander Point, and Shark Bay (Fig. 3). We have determined ages for eight corals sampled by L.B. Collins and Z.R. Zhu from reefs at the latter site. Of these, five are considered reliable and range from approximately 129 to 125 ka. Ten ages have been reported for the Last Interglacial reefs on the Houtman–Abrolhos Islands [19,34], of which two (117 and 124 ka) are considered reliable when they are screened by the above criteria. In particular, the 134 ka age of a single coral recovered from drill core which has been argued to constrain the timing of onset of the Last Interglacial highstand, is excluded.

5. Discussion

Last Interglacial sea-level signatures, for sites that lie far from the margins of the penultimate glacial maximum ice sheets, will not be the same as those for sites closer to the main ice centres because of the isostatic response of the lithosphere and mantle to ice and melt water loading and unloading [1]. Even the far-field sites, such as those along the continental margin of Western Australia, will exhibit some spatial variability in the sea-level response, but this can be neglected for the present purpose. Model predictions, assuming that ice volumes during the Last Interglacial were the same as they are today, show that at far-field sites, sea levels first reach their present level shortly before melting ceases and that highstands develop early in the interglacial interval, similar to the Late Holocene highstands observed along many far-field continental margins [35,36]. If ice volumes during the Last Interglacial were less than those of today then sea levels at that time would lie above these isostatic predictions. In contrast, at intermediate-field sites, particularly the more northern sites in the Caribbean, sea level is predicted to first reach the present level some time after ice melting has ceased and the maximum highstands do not occur until late in the interglacial interval [1].

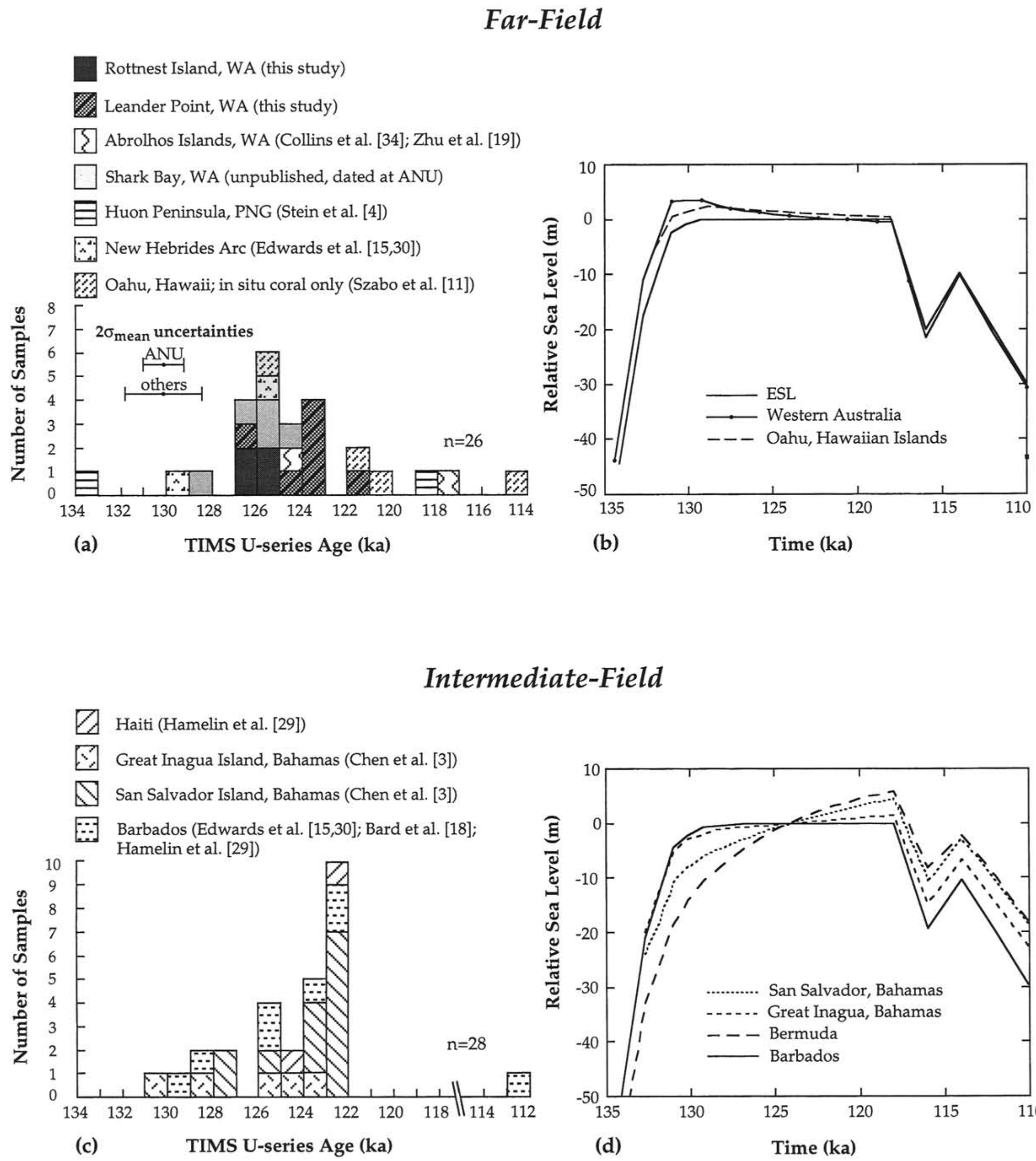
Therefore, the main phase of coral growth near present sea level is expected to occur later at intermediate-field sites than at far-field continental margin sites.

5.1. Implications for the timing and duration of the Last Interglacial from all available data

Fig. 4a summarises all reliable ages for the far-field sites that are currently available in the literature. Relative sea-level predictions for these locations are illustrated in Fig. 4b. The oldest reliable age for the far-field is 134 ka, determined for Reef VIIb at the rapidly uplifting Huon Peninsula, Papua New Guinea [4]. The stratigraphic position of this coral relative to other reliably dated corals from Huon Peninsula, indicates it most likely grew while sea level was rising rapidly prior to the start of the interglacial interval. If a rate of sea-level rise equivalent to that in the V19-30 oxygen isotope record [10] is assumed, then it is likely that the Last Interglacial commenced not more than 2–3 kyr later. In Western Australia, the occurrence of a 129 ka coral, more than 2 m above present-day sea level, at the tectonically stable region of Shark Bay indicates that sea-level highstands with amplitudes of at least 2–3 m above modern sea level had been attained by 129 ka. A comparable age is reported for the oldest part of the New Hebrides reef [15,30]. The above three far-field results therefore constrain the timing of onset of the Last Interglacial period to soon after 134 ka but before 129 ka. In addition, the Shark Bay data are in agreement with model predictions for the far-field which show highstands of several metres amplitude at the start of the interglacial interval. The height and age data for the Rottnest Island and Leander Point corals indicate that sea levels were at least 2 m above present sea level from 127 to 122 ka. The youngest coral from the Western Australian coast has an age of 117 ka and is located approximately 2 m above present-day sea level on the Abrolhos Islands [21,31], indicating that +2 m relative sea levels persisted there until at least 117 ka. This suggests that ocean volumes may have been greater during the Last Interglacial compared with those of today [1], although for the purpose of the present study, where absolute sea-level heights are not of paramount interest, the interglacial interval has been represented by zero eustatic sea level in Fig. 4. The youngest ages for in-situ coral from Oahu, Hawaii [11] are only slightly younger than the youngest coral from Western Australia. The earliest time at which the Last Interglacial ended may therefore be constrained to 117–114 ka.

Reliable ages and relative sea-level predictions for intermediate-field sites are shown in Fig. 4c and d, respectively. Because of the isostatic response of the lithosphere and mantle to the unloading of the North American ice sheet, the Last Interglacial must have commenced before the oldest ^{230}Th-age from the southernmost site in the Caribbean region, that is Barbados. Both Barbados and Great Inagua, Bahamas, are predicted to exhibit similar sea-level signatures over the duration of the interglacial (Fig. 4d) and, as expected, the oldest corals from both sites are identical within error, clustering about 129–130 ka [3,15] (Fig. 4c). These observations constrain the timing of onset of the interglacial to before 130 ka. At San Salvador, Bahamas, some 350 km north of Great Inagua, present sea level is predicted to first occur sometime after it is attained at Great Inagua and Barbados, in agreement with the possible younger age of 128 ka age for the oldest coral [3]. Relative sea-level predictions for San Salvador and Great Inagua show a well-developed highstand of several metres amplitude toward the end of the interglacial. The observational data are insufficient to verify this prediction; the ages do suggest that reef growth becomes increasingly prolific as the interglacial progresses but, apart from one dated sample at 112 ka [18], coral growth ends somewhat abruptly near 122 ka, approximately 7 kyr before the interglacial is predicted to end from the far-field data.

The observational evidence from Last Interglacial coral reefs from both far-and intermediate-field sites is thus in good agreement with the relative sea-level model predictions, assuming that the melting of the penultimate glacial maximum was complete and that interglacial conditions lasted from before 130 ka to at least 117 ka. This interval predates the Milankovitch insolation peak at 126–128 ka and suggests that either the orbital forcing model and its input parameters require some modifications or Pleistocene glacial–interglacial cycles are not driven by periodic changes in the Earth's orbital parameters alone. How then do we explain the tight clustering of ages between 127 and 122 ka that is observed in Western Australia as well as the abrupt termination of reef growth at 122 ka in the intermediate-field, in terms of a long-duration interglacial period? The period of prolific reef growth from 127 to 122 ka, occurring at Rottnest Island and Leander Point, when sea levels were still at least ~ 2 m above present-day levels, may correspond to one of the short-lived sea-level maxima that typically characterise the Last Interglacial period in marine $\delta^{18}O$ records (e.g.,



[10]). However, reef growth is also dependent on ocean surface temperatures, and an alternative explanation is that the interglacial was characterised by a single prolonged highstand, but with ocean temperatures along, for example, the western margin of Australia being warm enough for widespread coral growth only during the middle part of the interglacial interval. In this case the range in coral ages should become progressively larger and the timing of onset of reef growth should be progressively earlier for sites closer to the equator as ocean surface temperatures increase. Although the total number of reliably dated samples is small, some progression in the ages of onset and termination of reef growth is observed from Rottnest Island to Shark Bay. For example, the Rottnest Island reef appears to have formed in a short interval from 127 to 125 ka. No reefs formed on Rottnest Island during the Holocene and the implication is that ocean temperatures around the island were approximately 2° warmer than present-day temperatures during this interval only. At Leander Point, approximately 400 km north of Rottnest Island, a larger range in ages is observed; reef growth appears to have commenced by 127 ka and terminated near 122 ka. This apparent trend is supported by preliminary results for a single Shark Bay sample where the start of reef growth is shifted back to 129 ka.

5.2. *Implications for the timing and duration of the Last Interglacial from Rottnest Island and Leander Point data*

The new Western Australian observations provide constraints on the timing of onset and duration of the Last Interglacial and are more reliable than previously published data. Reliable ages from Rottnest

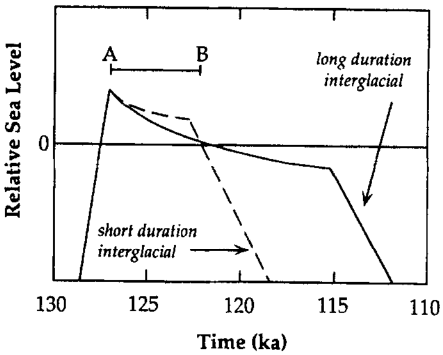


Fig. 5. Schematic illustration of sea-level change relative to the Western Australian continental margin for a long and short duration interglacial, assuming similar ice volumes to present day. The interval $A-B$ corresponds to the period of formation of the now emergent reefs at Rottnest Island and Leander Point. The dashed line corresponds to the inferred sea-level curve from the observational data. The solid line represents an alternative curve, for a faster mantle relaxation time and longer duration interglacial, in which any reefs that formed during the latter part of the interval would now lie a few metres below sea level.

Island and Leander Point range from 127 to 122 ka. If we assume that sea levels relative to the continental margin of Western Australia were at or above present-day levels throughout the entire duration of the interglacial interval, then the implication is that the Last Interglacial extended from 127 to 122 ka only (dashed curve in Fig. 5). This is of shorter duration than previously determined from the coral record but is in close agreement with the timing and duration obtained from orbital tuning of the marine oxygen isotope records (e.g [9,10]) and hence with the predictions of the Milankovitch model. The large spread in apparent ages observed for other Last Interglacial reef sites may then be due to subtle diagenetic alteration in the coral skeleton, giving rise

Fig. 4. Distribution of "reliable" ^{230}Th-ages for Last Interglacial reef sites in the (a) far-field and (c) intermediate-field. The data point at 133–134 ka for Huon Peninsula is the weighted mean of three ages for two corals in close proximity to one another, which most likely grew while sea level was rising rapidly prior to the start of the interglacial interval. Model predictions for relative sea level for (b) far-field sites in Western Australia and (d) intermediate-field sites in Bermuda, Barbados and the Bahamas. For the purpose of comparing differences in model predictions between far-and intermediate-field sites we have adopted one relative sea-level curve for the entire coastal margin of Western Australia since differences in model predictions between Rottnest Island, Leander Point, Shark Bay and the Abrolhos Islands are minimal. Similar relative sea levels are predicted for Huon Peninsula and the New Hebrides Arc. The Last Interglacial is represented by approximately zero eustatic sea level (ESL) from 131 to 117 ka. The Earth model corresponds to that determined from glacio-hydro-isostatic studies of Holocene sea levels [25]. The ice model is discussed in Lambeck and Nakada [1] and includes contributions from both hemispheres.

to anomalous ^{230}Th-ages which cannot be readily distinguished from those assessed as reliable due to large errors on $\delta^{234}U(T)$ measurements. When the Last Interglacial TIMS data from other laboratories are screened by the strict $\delta^{234}U(T)$ criterion adopted in this study, close to 80% of the ages cluster between the 127 and 122 ka limits defined by the Rottnest Island and Leander Point data (Fig. 1b), providing support for the argument of a short-duration interglacial interval.

Alternatively, sea levels relative to the tectonically stable margin of western Australia, may have been at or above present levels during the early part of the Last Interglacial only, as some far-field model predictions indicate that the latter part of the interglacial may be recorded in fossil reefs which are currently a few metres below present sea level (solid curve in Fig. 5). ^{230}Th-ages for the emergent reefs from stable far-field localities will therefore provide a good estimate for the timing of onset of the interglacial interval but may give an apparently short duration. In this context, the Rottnest Island and Leander Point data indicate that the Last Interglacial commenced near 127 ka and terminated perhaps several thousand years after 122 ka. However, this explanation seems unlikely since sea levels relative to the western Australian coast, as indicated by LP-93-3, were still at least ~ 2 m above present levels at 122 ka when prolific reef growth ended.

6. Conclusions

The new observations for Rottnest Island and Leander Point are more reliable than previously published results for several reasons. Firstly, we have selected corals in growth position from localities that are characterised by apparently low levels of diagenesis and relative tectonic quiescence so that the relationship between the reef and coral heights and relative sea level is well constrained. Secondly, using charge collection thermal ionisation mass spectrometry, we have achieved superior precision in Th isotope measurement. For the first time Th-ion beams have been measured simultaneously, in multiple channels, with ionisation efficiencies of up to 6%. Hence, Th isotope ratios are routinely measured up to a factor of ten more precisely than reported by other laboratories. Thirdly, the level of precision in U isotope measurement is typically 1–2‰, significantly better than reported by many earlier TIMS workers. The combined improvements in the measurement of both U and Th isotopes has led to up to a factor of four reduction in the age uncertainty due to analytical errors, excluding the uncertainty in the decay constants, potentially allowing the finer details of sea-level change during the Last Interglacial to be resolved. In addition, $\delta^{234}U(T)$ values, determined from both $^{230}Th/^{238}U$ and $^{234}U/^{238}U$, have uncertainties which are significantly smaller than previously reported. When these superior $\delta^{234}U(T)$ measurements are screened with a strict $\delta^{234}U(T)$ criteria, such that $\delta^{234}U(T)_{acceptable} = 149 \pm 4$, the remaining acceptable values give the most reliable, as well as the most precise, chronological data for the Last Interglacial period.

However, no single criterion appears to be completely reliable for assessing the veracity of an age and the possibility remains that some individual corals may still have undergone subtle diagenetic alteration, giving rise to anomalous ^{230}Th-ages, without disturbing their $\delta^{234}U(T)$ values. For this reason, we prefer not to place undue emphasis on single ages within any one thousand year time interval. It is important to emphasise that when all "reliable" chronological data are considered, constraints on the timing and duration of the Last Interglacial are largely determined from such single data points (Fig. 4). It will be necessary to verify these single ages with additional data before considering them as robust estimates for the timing of onset and termination of the Last Interglacial.

The main feature of Fig. 4 is that, apart from a few outliers, the major episode of reef building, in both the far-and the intermediate-field, is confined to a very narrow interval occurring from 127 to 122 ka. This is highlighted by the data from Rottnest Island and Leander Point.

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