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Research paper

Testing the precision and accuracy of the U–Th chronometer for dating coral mortality events in the last 100 years



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ABSTRACT

To assist with our understanding of reef dynamics prior to modern monitoring programs and recent observations of coral decline, a robust dating technique is required to place coral mortality events and historical changes in community structure in an accurate chronological framework. In this study we adopted a refined Uranium-Thorium (U-Th) isotope measurement protocol using multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) for rapid, precise and accurate age determination of a large branching Acropora coral death assemblage from an inshore reef of the Great Barrier Reef (GBR) where the timing of mortality is independently constrained. To achieve this, we developed a vigorous sample cleaning/treatment procedure to remove most non-carbonate detritus from the coral skeleton, and a correction scheme that accounts for initial ²³⁰Th sources in the dead coral skeletons. Using this method, the 230 Th ages (with 2σ errors of 1–5 years) from 41 individual dead Acropora branches precisely bracket the timing of a documented ~100% loss of hard coral cover, primarily Acropora, that was caused by increased sea-surface temperatures during the 1997-1998 mass bleaching event. Our results demonstrate the applicability of U-Th dating in accurately determining the timing of previous disturbance events in coral reef communities, as well as identifying potential drivers. This approach provides a powerful tool to researchers and managers in assessing the current status of reefs and identifying areas vulnerable to degradation where long-term monitoring data are absent or too recent. © 2014 Elsevier B.V. All rights reserved.

1. Introduction

Current observations and future predictions of the status of corals reefs appear grim in the face of anthropogenic disturbance and climate change (Hughes et al., 2003; Pandolfi et al., 2011). However, on the Great Barrier Reef (GBR), Australia, there is still considerable debate as to whether inshore reefs are degraded or not (Hughes et al., 2011; Sweatman and Syms, 2011), partly due to a lack of understanding of coral community structure and disturbance history beyond the time period of long-term monitoring (Pandolfi et al., 2003; Roff et al., 2013).

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Palaeoecological studies provide a means to examine past changes in coral community structure and historical mortality events (Pandolfi and Greenstein, 1997), but a well-established chronology is also required to determine the absolute timing of these events. While a large number of studies have quantitatively described historical changes in coral communities (e.g. Greenstein et al., 1998; Pandolfi and Jackson, 2006), only a few studies have isotopically dated samples at high enough resolution and with consistently low uncertainties to be able to link mortality events with specific drivers (e.g. Cramer et al., 2012; Pandolfi et al., 2006; Roff et al., 2013; Yu et al., 2012a, 2012b, 2006).

 $^{238}U^{-230}$ Th disequilibrium (U–Th) dating, which utilises the $^{238}U^{-234}U^{-230}$ Th decay chain, has proven to be a reliable method for determining the age of Pleistocene to Recent carbonate deposits. While the majority of studies have focused on dating

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samples thousands of years old, recent analytical advancements has led to dating coral samples as young as a few years with a precision of up to $\pm 1-2$ years (Clark et al., 2012; McCulloch and Mortimer, 2008; Roff et al., 2013; Shen et al., 2008; Yu et al., 2012a, 20012b, 2006; Zhao et al., 2009). However, the uncertainty in hydrogenous and detrital sources of ²³⁰Th [also termed 'nonradiogenic' (²³⁰Th_{nr}), initial (²³⁰Th₀) or secondary ²³⁰Th sources that were not generated by the *in situ* decay of U] incorporated into the coral skeleton during skeletogenesis and after death, respectively, can result in inaccurate age estimates [for review see Zhao et al. (2009)]. This is especially true for young coral samples where the proportion of 230 Th₀ is significantly greater than the radiogenic ²³⁰Th component, or for corals from inshore reef settings where non-carbonate terrestrial input is much greater compared to offshore settings; the former having ²³²Th levels typically a few orders of magnitude higher than the latter (cf. Burley et al., 2012; Clark et al., 2012; Roff et al., 2013; Yu et al., 2012a, 2012b). As the 230 Th₀ in a sample cannot be separated from the radiogenic 230 Th during measurement, it can only be corrected for using the measured ²³²Th level in conjunction with the initial ²³⁰Th/²³²Th ratio in the sample (expressed here as ${}^{230}\text{Th}/{}^{232}\text{Th}_0$). As $^{230}\text{Th}/^{232}\text{Th}_0$ may vary between sites or even between samples, a bulk-Earth activity value of 0.82 (atomic value \sim 4.4 \times 10⁻⁶) with a large arbitrarily assigned uncertainty of $\pm 50-100\%$ has been commonly assumed to correct for the $^{230}Th_0$ contribution. Although this assumption was proven to be acceptable for most coral samples from inshore settings (Clark et al., 2012; Shen et al., 2008), the large associated uncertainty makes the age uncertainty of the corrected ²³⁰Th age for young corals too large to be meaningful (see Zhao et al., 2009).

The 230 Th/ 232 Th₀ ratio in a sample with detrital and carbonate components is generally constrained using isochron diagrams from multiple sub-samples of coeval material. This approach works if all the initial/detrital ²³²Th and ²³⁰Th are from the detritus, the 234 U/ 238 U and 230 Th/ 232 Th of the detrital component are the same for all sub-samples, and the system has remained closed (Bischoff and Fitzpatrick, 1991; Richards and Dorale, 2003). However, sources of 230 Th₀ in corals can be highly variable (Clark et al., 2012; Cobb et al., 2003; Shen et al., 2008; Yu et al., 2006) and it is likely that corals from coastal environments contain two (or more) sources of ²³⁰Th₀: both detrital particulates and hydrogenous ²³⁰Th. In that regard, inshore reef corals would be comparable to lake carbonates (Haase-Schramm et al., 2004; Lin et al., 1996) and deep-sea corals (Cheng et al., 2000a). Where the $^{230}\mathrm{Th}/^{232}\mathrm{Th}$ values of the detrital and hydrogenous components are dissimilar, the inability to account and correct for both sources can introduce substantial biases to the ²³⁰Th age. To improve both the precision and accuracy of corrected ²³⁰Th ages, it is necessary to be able to correct for ²³⁰Th₀ based on a well-constrained site- or sample-specific 230 Th/ 232 Th₀ ratio (Clark et al., 2012; Shen et al., 2008).

For this study, the remains of a large number of dead branching *Acropora* corals were used to verify the accuracy of the U–Th dating method and the application of a sample-specific ²³⁰Th₀ correction scheme to determine the time of death. The *Acropora* 'death assemblage' (Pandolfi and Greenstein, 1997) sampled is at Pandora Reef, an inshore reef from the central GBR, Australia, for which both short-term observations (DeVantier et al., 1997) and long-term coral monitoring data are available (Done et al., 2007; Sweatman et al., 2005). These data allowed our estimates of the timing of coral death to be compared with those noted during direct field observations. A close match would facilitate the dating of the time of death of corals on the vast majority of reefs over long temporal scales (viz. pre-1980s) for which direct time-series observations of corals do not exist.

2. Materials and methods

2.1. Study site

Pandora Reef (18°48′S, 146°26′E; 750 m long \times 200 m wide; Fig. 1) lies 17 km from the mainland and is episodically reached by flood plumes from the Burdekin and Herbert Rivers and other smaller tributaries adjacent to the region. Branching *Acropora* colonies were found in high abundance on the fore reef slope (sites P1 and t5) from 1985 to the start of 1998, and lesser abundance on the back reef slope (sites t1-4, V1 and V2) which was characterised by other coral genera including *Goniopora*, *Turbinaria*, and *Porites* (Done et al., 2007).

In 1997-1998 a severe El Niño event created a heat wave causing widespread bleaching along the entire length of the GBR from mid-December to early March (Berkelmans et al., 2004; Marshall and Baird, 2000). In the Palm Islands, bleaching was first reported on the 10 February 1998 and reduced coral cover by more than 50% in Acropora-dominated communities and up to 100% at some exposed reef flats (Gralton, 2002; Marshall and Baird, 2000). At Pandora Reef, bleaching was first reported at the beginning of March (Suzuki et al., 2003), affecting approximately 80% of hard corals down to a depth of 10 m. Members of the family Acroporidae were the most affected, with almost a complete loss reported across the entire fore-reef [(Done et al., 2007; Sweatman et al., 2005); Fig. 5c]. At the time of sampling in May 2008, a large expanse of dead Acropora branches, attributable to the 1998 bleaching event, formed a consolidated matrix at the sedimentwater interface along the entire south-west flank of Pandora Reef and was overgrown by macroalgae (Fig. 2a). This is in contrast to the high coral cover (>50%) reported in 1994, when branching Acropora colonies were present at all three [shallow (1-3 m), midslope (4-6 m) and deep (9-11 m)] depth ranges (DeVantier et al., 1997).



Fig. 1. Map showing location of the Palm Islands in the central region of the Great Barrier Reef and an enlarged map of Pandora Reef. Sites marked as V1, V2, t1-4 and t5 (green lines) are the locations of video transects surveyed by AIMS from 1992 onwards. Site P1 (blue circle) is a photo transect monitored by T. Done from 1980 to 2005. Red bar represents the location where dead baraching *Acropora* were sampled in this study. This site was also surveyed by DeVantier et al. (1997) in February and April 1994, however, quantitative data are unavailable. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 2. a) Photograph of *Acropora* death assemblage overgrown by macroalgae taken in May 2008 from the south-west flank of Pandora Reef; b) Example of a dead arborescent *Acropora* branch collected for U–Th dating; c) Example of coral skeletal sample after pressurised cleaning with H_2O_2 . Note the successful removal of detrital material. Scale represented by 5 mm grid paper.

2.2. Coral collection and sampling

Dead Acropora coral rubble samples were collected from the leeward back reef environment at Pandora Reef at a depth of 4–5 m (Fig. 1). Five grab samples (~5 L each) of dead coral rubble were collected by hand at the sediment—water interface and placed in calico bags at random points along each of four 20 m transects laid parallel to the reef flat (T1–T4; Table 1); thus totalling 20 grab



Fig. 3. ²³⁰Th/²³²Th versus ²³⁸U/²³²Th isochron for five coeval sets of sub-samples obtained from annual growth bands of dead *Porites* skeletons collected from the Palm Islands region, central Great Barrier Reef. Inset shows the isochron-inferred ²³⁰Th/²³²Th₀ ratios (y-intercepts with 2σ errors) of the detrital component (average 0.61 ± 0.01 (1 σ)).

samples with a spatial coverage of >80 m. The contents of the bags which predominately contained dead Acropora branches, were dried and individual branches with an intact branch tip or firstorder branch (Bottjer, 1980) representing the most recent growth were selected from each of the 20 grab samples (Fig. 2b). Approximately 0.5-1 g of material was sampled from the cleanest part along the length of the branch, but within 16 cm (average sampling location was 5.0 cm) of each branch tip, using a diamond blade saw from 41 individual branches (Table 1). This ensured enough high quality material, free from alteration, for U–Th dating. Given high linear extension rates in branching Acropora corals typical of turbid, inshore, sheltered environments [from an average ~ 17.9 cm vr⁻¹ (Crabbe and Smith, 2005) up to 33.3 \pm 4.2 cm yr⁻¹(Diaz-Pulido et al., 2009)], an average sampling location ~5.0 cm from the branch tip would ensure that the site of skeletogenesis where sampling took place was within 0.3 yrs (or ~4 months) of the time of colony death.

Each sample was crushed to a ~1 mm grain size fraction and soaked overnight in a pre-cleaned glass beaker containing ~10% H₂O₂. It was then rinsed with Milli-Q water and centrifuged for 15 min at 4000 rpm in a pre-cleaned Teflon beaker containing enough ~10% H₂O₂ to cover the sample. Samples were again rinsed with Milli-Q water and ultra-sonicated several times until the water was clear. The excess water was then decanted and the sample dried on a hotplate at 40 °C. This rigorous cleaning procedure ensures that detrital contaminants containing high concentrations of ²³²Th are removed from the pore spaces of the skeletal matrix. The quality of each sample was then inspected under a binocular microscope and approximately 500 mg of the cleanest skeletal material selected for U–Th dating (Fig. 2c).

2.3. U–Th chemistry procedures

All U–Th chemistry and analytical procedures were performed under ultra-clean conditions at the Radiogenic Isotope Laboratory, the University of Queensland. Approximately 0.03 ml of a 229 Th– 233 U mixed tracer (229 Th– 233 U-spike #2) was added to each pre-cleaned Teflon beaker using a pipette and the weight recorded. The spike solution was then dried down completely at 60 °C on a hot-plate, after which ~0.5 g of sample material was added to the spiked beaker. The sample/spike was then dissolved in doubledistilled 70% HNO₃, co-precipitated with Fe(OH)₂ and U and Th separated using the standard ion-exchange column chemistry



Fig. 4. Mixing diagram of $1/^{232}$ Th (ppb) plotted against 230 Th/ 232 Th ($\pm 2\sigma$) activity ratios using mean values obtained from live *Porites* (230 Th/ 232 Th = 1.08 \pm 0.19, 232 Th = 0.95 ppb; green line), live *Acropora* (230 Th/ 232 Th = 3.5 \pm 0.8, 232 Th = 0.15 ppb; blue line), and isochron-derived detrital ratio (similar to Burdekin River sediments) obtained from dead *Porites* (230 Th/ 232 Th = 0.61 \pm 0.02, $1/^{232}$ Th = 0.9pb; orange line). Isotopic data from all components show a negative correlation between two end-members: a detrital (sediment) phase and a hypothetical hydrogenous (seawater) phase (large open circle with a question mark) that is yet to be constrained. Following correction for 230 Th₀ using a two-component mixing model, the measured 230 Th/ 232 Th ratios obtained from the dead *Acropora* samples (grey circles) shift towards and fall on the mixing line (grey triangles with error bars not shown for ease of interpretation), with most lying close to the sediment and live *Porites* values (see enlargement). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

procedure slightly modified after Edwards et al. (1987). Following separation, the U and Th solutions were dried overnight at 80 °C then redissolved using 1 ml of 2% HNO₃. The amount of uranium solution to be measured was calculated based on pre-screened signals from a more dilute U–Th solution to ensure that the ²³⁸U signal in the final solution did not exceed the capacity of the Faraday cups. For 500 mg of coral sample, approximately 20 µl U solution was taken from the 1 ml stock solution and transferred to a pre-cleaned 3 ml polypropylene tube to achieve a final concentration of ~7 ppb U. The entire 1 ml solution of Th was also added and made up to 3 ml using 2% HNO₃. Tubes were then centrifuged at 4000 rpm for 20 min to remove any suspended material (mainly a trace amount of leaked resin) from the solution. All 41 samples were subsequently measured for U and Th isotopes using a Nu Plasma Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) to ascertain the timing of mortality, following a protocol first reported by Zhou et al. (2011).

2.4. MC-ICP-MS measurement

The U–Th mixed solution was injected into the MC-ICP-MS through a DSN-100 desolvation nebulizer system with an uptake rate of around 0.12 ml per minute. The U–Th isotopic ratio measurement protocol, which was first reported in Zhou et al. (2011), is similar to that previously described by Hellstrom (2003), but with minor modification to the detector configuration, viz. the U–Th

isotopes were measured in two, instead of three sequences as used in Hellstrom (2003) (Table S1).

In our protocol, the two secondary electron multiplier (or SEM) gains are calculated from the SEM and Faraday signal ratios of 233 U and 229 Th (shaded grey in Table S1) that are collected in the two sequences, respectively. In addition, a deceleration lens behind SEM2 was used to increase the abundance sensitivity by 10 times (to reduce 238 U tailing at mass 237 to <0.5 ppm), so that the 232 Th tailing effect at mass 230 is small enough for accurate correction (the tailing effect was corrected using the geometric mean of background measurements at masses 229.5 and 230.5), regardless of the size of the 232 Th/ 230 Th ratio in the sample. For instance, even for samples with a 232 Th/ 230 Th abundance ratio of ~100,000, the 232 Th tailing contribution to the 230 Th signal is typically less than 3%. Each sample took about 25 min to measure. Measurements of samples, standards, and carryover memories were performed fully automatically using a modified Cetac ASX-110 autosampler.

A 'drift monitoring' solution was made by adding ²²⁹Th and ²³³U spikes separately into a dilute solution of a uranium oxide impurity standard New Brunswick Laboratory-6 (NBL-6) from the USA. The 'drift monitoring' was repeatedly measured after every six unknown samples, and the results were used to correct for long-term drift in a number of parameters such as ion counter gain (gain values were interpolated from bracketing the 'drift monitors' if ²²⁹Th and/or ²³³U signals in the samples were too small), and minor bias in the ²³⁰Th/²³⁸U and ²³⁴U/²³⁸U ratios during each session (the



Fig. 5. The effects of each correction scheme on ²³⁰Th ages. a) Uncorrected b) bulk Earth c) live coral d) isochron-derived detrital component from dead *Porites* corals, and e) two-component corrected ²³⁰Th ages versus ²³²Th (ppb). If the bulk Earth- and live coral-based ²³⁰Th/²³²Th₀ values were used for correction, the corrected ages show a positive relationship with measured ²³²Th (r = 0.8670, P = <0.0001 and r = 0.9702,

bias was mainly caused by the imperfect signal peak shapes and alignments). The working U concentration in the 'drift monitor' when this set of samples were analysed, was ~6 ppb, with ²³⁸U, ²³³U and ²²⁹Th signal sizes typically around ~3 V (V), ~8 mV and ~2 mV at typical machine sensitivities, respectively, and has been precisely calibrated against the secular equilibrium Harwell Uraninite, HU-1 (Stirling et al., 1995; Zhao et al., 2001; Hellstrom, 2003).

2.4.1. Spike measurements

A mixed ²²⁹Th–²³³U tracer designed for dating young coral samples was added to each of the Teflon beakers prior to digestion. The isotopic compositions of this mixed tracer were determined to be: ²³⁸U/²³³U = 4.90 × 10⁻⁴ ± 0.74%, ²³⁴U/²³³U = 2.37 × 10⁻³ ± 0.15%, ²³⁵U/²³³U = 1.25 × 10⁻⁴ ± 0.30%, ²³²Th/²²⁹Th = 1.22 × 10⁻⁴ ± 1.6% and ²³⁰Th/²²⁹Th = 4.78 × 10⁻⁵ ± 0.5%. The spike concentrations are ²³³U = 1.42029 × 10⁻² nm g⁻¹, ²³⁸U = 6.96390 × 10⁻⁶ nm g⁻¹, ²²⁹Th = 1.25476 × 10⁻³ nm g⁻¹, ²³²Th = 1.53356 × 10⁻⁷ nm g⁻¹.

2.4.2. Blank correction

For MC-ICP-MS measurements of samples with very young ages, one of the main contributors to age error is the carry-over memory between samples being measured. However, this was alleviated by flushing the system prior to a new sample being measured for 15 min with 5% Aqua regia followed by 2% HNO₃ to prevent any cross contamination or 'memory' effect. In the clean-up stage all isotopes were monitored and raw counts measured on their respective detectors to ensure no carry-over memories from previous samples. Long-term monitoring of carryover memories over 20 months shows that ²³⁰Th memory is less than 0.1 counts per second (cps) most of the time, which is negligible for most samples. ²³⁰Th signals in the samples range from 20 to 50 cps, about 200–500 larger than the carry-over memory. The memories for all other isotopes are also negligible.

The total procedural ²³⁰Th blank was determined to be $1.18 \pm 0.24 \times 10^{-10}$ nmol or 0.27 ± 0.05 fg (N = 10); contributing an average 0.09 yr to the ²³⁰Th ages of the samples in this study. The procedural blanks for ²³⁸U and ²³²Th were averaged at $1.4 \pm 0.9 \times 10^{-5}$ nmol (or 3.3 ± 2.2 pg) and $3.0 \pm 1.9 \times 10^{-6}$ nmol (or 0.69 ± 0.41 pg), respectively, which are negligible for coral samples containing typically ~3 ppm U. These values are much lower than the procedural blanks measured using thermal ionisation mass spectrometry (TIMS), where high blank contributions were considered to be a result of more complex column chemistry and the colloidal graphite used to load the sample onto the filaments (Clark et al., 2012). Procedural ²³⁰Th and ²³²Th blanks were extracted from the samples in the Microsoft Excel spreadsheet used for U–Th age calculation.

2.5. Initial 230 Th (230 Th₀) correction

After MC-ICP-MS measurements, U–Th ages were calculated using the Isoplot/Ex version 3.0 program (Ludwig, 2003b). In order

P = <0.0001, respectively). However, if the isochron-derived detrital ²³⁰Th/²³²Th₀ value and our two-component mixing model ²³⁰Th/²³²Th₀ value are used there is no correlation: (r = -0.2288 P = 0.1502 and r = -0.2361 P = 0.1372, respectively), suggesting that the presence of ²³²Th (a proxy for the amount of initial ²³⁰Th in a sample) has been appropriately corrected for. Dashed line represents the timing of the 1997–1998 bleaching event. Grey line represents an uncertainty of 0.3 years in order to account for systematic bias towards slightly older ages due to the material used for dating being taken from areas of the coral skeleton that was deposited in the months leading up to the coral colony's death. This value was based on average linear extension rates in branching *Acropora* corals typical of turbid, inshore, sheltered environments (~17.9 cm yr⁻¹; Crabbe and Smith, 2005) and taking into consideration the average sampling location with respect to the distance from the tip of the branch which was determined to be ~5.0 cm.

40

Table 1

T.R. Clark et al. / Quaternary Geochronology 23 (2014) 35-45

MC-ICP-MS²³⁰Th ages for dead branching Acropora samples collected from the death assemblage at Pandora Reef, central Great Barrier Reef.

Sample name ^a	Sampling range (cm) ^b	Sample wt.(g)	U (ppm)	²³² Th (ppb)	(²³⁰ Th/ ²³² Th) _{meas}	(²³⁰ Th/ ²³⁸ U)	$\delta^{234}U^{c}$	Uncorr. ²³⁰ Th age (AD)	Time of chemistry
PanS1T1.2	5.9	0.52861	3.1264 + 0.0024	2.3013 + 0.0048	1.336 ± 0.030	0.0003241 ± 0.0000069	147.0 + 1.0	1979.8 ± 0.7	2010.7
PanS1T1.3	13.4	0.59958	3.0211 + 0.0013	0.9677 + 0.0023	2.362 ± 0.042	0.0002494 + 0.0000040	146.7 + 1.2	1986.9 ± 0.4	2010.7
PanS1T1.4	7.5	0.55553	3.1885 + 0.0015	3.1981 + 0.0039	1.271 + 0.021	0.0004202 + 0.0000066	147.2 + 1.2	1970.7 + 0.6	2010.7
PanS1T1.5	13.0	0.50465	3.0345 + 0.0011	1.7522 + 0.0023	1.607 + 0.027	0.0003059 + 0.0000049	147.0 + 1.2	1981.6 + 0.5	2010.7
PanS1T1.7	9.7	0.64273	3.1385 + 0.0020	0.26074 + 0.00038	6.75 ± 0.19	0.0001848 ± 0.0000054	145.4 + 1.2	1993.1 + 0.5	2010.7
PanS1T1.8	16.5	0.53981	3.0306 + 0.0019	1.3201 + 0.0018	1.841 + 0.032	0.0002643 + 0.0000045	145.9 + 1.1	1985.5 + 0.4	2010.7
PanS1T1.9	11.7	0.61789	3.2140 + 0.0018	3.8856 + 0.0051	1.050 + 0.017	0.0004185 + 0.0000065	146.8 + 1.0	1970.8 + 0.6	2010.7
PanS1T1.10	12.1	0.56444	3.1802 + 0.0013	0.9354 + 0.0014	2.407 + 0.043	0.0002333 + 0.0000040	145.5 + 1.0	1988.5 + 0.4	2010.7
PanS1T1.11	13.8	0.73242	3.1275 + 0.0015	3.4069 + 0.0031	1.108 + 0.017	0.0003977 + 0.0000059	145.0 + 0.9	1972.8 + 0.6	2010.7
PanS1T1.12	9.5	0.51359	3.1434 + 0.0023	3.3456 + 0.0062	1.192 + 0.021	0.0004181 + 0.0000070	146.1 + 0.9	1970.9 + 0.7	2010.7
PanS1T2.1	8.0	0.61479	3.1976 + 0.0014	0.6177 + 0.0011	3.890 + 0.060	0.0002477 + 0.0000036	146.8 + 1.2	1987.1 + 0.3	2010.7
PanS1T2.3	5.4	0.58241	3.1133 + 0.0016	3.1317 + 0.0032	1.169 + 0.019	0.0003876 ± 0.0000063	146.5 + 1.2	1973.8 + 0.6	2010.7
PanS1T2.4	10.8	0.51142	3.2295 + 0.0016	1.5958 + 0.0015	1.707 + 0.031	0.0002779 + 0.0000050	146.5 + 1.2	1984.2 + 0.5	2010.7
PanS1T2.5	12.4	0.54179	2.9378 + 0.0017	0.45549 + 0.00067	4.055 + 0.086	0.0002072 + 0.0000043	147.5 + 1.0	1991.0 + 0.4	2010.7
PanS1T2.6	8.0	0.57222	2.9336 + 0.0015	0.43871 + 0.00060	4.079 ± 0.098	0.0002010 + 0.0000048	146.7 + 1.1	1991.6 + 0.5	2010.7
PanS1T2.7	8.9	0.50702	3.3822 + 0.0014	1.7674 + 0.0016	1.616 + 0.035	0.0002784 ± 0.0000060	146.0 + 0.9	1984.2 + 0.6	2010.7
PanS1T2.8	6.8	0.53453	2.9511 + 0.0015	3.6596 + 0.0031	1.155 + 0.018	0.0004719 + 0.0000073	146.3 + 0.8	1965.7 + 0.7	2010.7
PanS1T2.9	5.9	0.55237	3.2718 + 0.0015	0.9260 + 0.0013	2.730 + 0.062	0.0002546 ± 0.0000056	145.7 + 1.2	1986.4 + 0.5	2010.7
PanS1T2.10	7.4	0.57864	2.9315 + 0.0020	1.1244 + 0.0012	2.029 + 0.047	0.0002565 + 0.0000060	145.9 + 1.4	1986.3 + 0.6	2010.7
PanS1T2.11	8.0	0.51666	3.1868 ± 0.0016	1.9400 ± 0.0020	1.495 ± 0.028	0.0002999 ± 0.0000056	146.3 ± 1.0	1982.1 ± 0.5	2010.7
PanS1T2.12	6.4	0.65912	3.2470 + 0.0016	2.6622 + 0.0026	1.283 + 0.019	0.0003466 + 0.0000050	146.0 + 1.2	1977.7 + 0.5	2010.7
PanS1T3.3	11.8	0.53734	3.2765 + 0.0016	0.49495 + 0.00057	4.211 + 0.078	0.0002096 + 0.0000039	147.0 + 1.2	1990.9 + 0.4	2010.9
PanS1T3.4	14.5	0.55105	3.1239 + 0.0015	1.5117 + 0.0036	1.768 + 0.027	0.0002819 + 0.0000039	146.6 + 0.8	1984.1 + 0.4	2010.9
PanS1T3.5	8.0	0.53053	3.1027 + 0.0014	0.30429 + 0.00060	6.51 + 0.16	0.0002104 + 0.0000050	146.7 + 0.8	1990.9 + 0.5	2010.9
PanS1T3.6	5.2	0.51268	3.2460 + 0.0020	0.50127 + 0.00076	4.378 ± 0.083	0.0002228 + 0.0000041	146.2 + 0.9	1989.7 + 0.4	2010.9
PanS1T3.7	9.2	0.56672	3.2631 ± 0.0020	0.81785 ± 0.00090	2.641 ± 0.058	0.0002182 ± 0.0000048	147.0 ± 1.2	1990.1 ± 0.5	2010.9
PanS1T3.8	15.5	0.52514	3.2931 + 0.0016	0.60433 + 0.00049	3.512 + 0.065	0.0002124 + 0.0000040	147.0 + 0.8	1990.7 + 0.4	2010.9
PanS1T3.11	14.5	0.53172	3.3683 + 0.0016	2.3049 + 0.0017	1.423 + 0.020	0.0003210 + 0.0000046	147.2 + 1.0	1980.3 + 0.4	2010.9
PanS1T3.13	5.7	0.51982	3.2520 + 0.0018	3.2679 + 0.0035	1.168 + 0.021	0.0003867 + 0.0000069	146.2 + 0.7	1974.1 + 0.7	2010.9
PanS1T3.15	7.5	0.63710	3.3315 + 0.0014	2.7712 + 0.0032	1.229 + 0.018	0.0003368 + 0.0000048	146.4 + 0.8	1978.8 + 0.5	2010.9
PanS1T4.1	12.7	0.54322	3.2816 ± 0.0015	0.54822 ± 0.00055	4.136 ± 0.064	0.0002277 ± 0.0000035	147.7 ± 0.9	1989.2 ± 0.3	2010.9
PanS1T4.3	12.0	0.54173	3.1571 ± 0.0013	1.4899 ± 0.0013	1.746 ± 0.032	0.0002716 ± 0.0000050	147.5 ± 0.9	1985.1 ± 0.5	2010.9
PanS1T4.4	9.7	0.51410	3.2284 + 0.0010	0.85858 + 0.00081	2.718 + 0.047	0.0002382 + 0.0000041	147.9 + 0.8	1988.2 + 0.4	2010.9
PanS1T4.5	13.5	0.52256	3.1620 + 0.0013	1.0616 + 0.0021	2.207 + 0.042	0.0002442 + 0.0000044	146.7 + 0.9	1987.6 + 0.4	2010.9
PanS1T4.6	6.2	0.54909	3.1463 + 0.0016	0.34359 + 0.00039	5.93 ± 0.10	0.0002133 + 0.0000036	147.8 + 1.2	1990.6 + 0.3	2010.9
PanS1T4.7	14.0	0.54121	3.0927 + 0.0021	0.81946 + 0.00083	3.059 ± 0.058	0.0002671 + 0.0000051	148.9 ± 0.9	1985.5 + 0.5	2010.9
PanS1T4.9	10.8	0.79178	3.2502 + 0.0019	0.8433 + 0.0014	2.792 + 0.044	0.0002387 + 0.0000036	146.8 + 1.0	1988.2 + 0.3	2010.9
PanS1T4.10	7.5	0.70195	3.2810 ± 0.0015	1.5303 ± 0.0013	1.792 ± 0.028	0.0002755 ± 0.0000042	146.9 ± 1.1	1984.7 ± 0.4	2010.9
PanS1T4.12	9.0	0.59781	3.2461 ± 0.0015	0.63972 ± 0.00085	3.424 ± 0.059	0.0002224 ± 0.0000037	146.9 ± 0.9	1989.7 ± 0.4	2010.9
PanS1T4.13	11.0	0.65207	3.2151 ± 0.0014	0.58939 ± 0.00080	3.560 ± 0.059	0.0002151 ± 0.0000035	146.5 ± 1.1	1990.4 ± 0.3	2010.9
PanS1T4.17	8.3	0.54801	3.2038 ± 0.0015	2.0043 ± 0.0035	1.479 ± 0.028	0.0003050 ± 0.0000054	145.9 ± 1.0	1981.8 ± 0.5	2010.9

Ratios in parentheses are activity ratios calculated from atomic ratios using decay constants of Cheng et al. (2000b). All values have been corrected for laboratory procedural blanks. All errors reported as 2σ . Uncorrected ²³⁰Th age (a) was calculated using Isoplot/EX 3.0 program (Ludwig, 2003b), where *a* denotes year.

^a For the sample nomenclature, S1 refers to Site 1 where the samples were collected at Pandora Reef. T1–T4 refers to transects 1–4 which were each 20 m in length. The number after the decimal refers to the individual *Acropora* branch dated from that particular transect.

^b Sampling range where material for U–Th dating was collected with respect to distance from the tip of the branch, or end of the branch where the tip has broken off, in centimetres. To ensure a 0.5–1.0 g sample size free from alteration, it was not possible to sample from a single location.

 $^{c} \delta^{234} U = [(^{234} U/^{238} U) - 1] \times 1000.$

to assess whether the presence of $^{230}\text{Th}_0$ can be reliably corrected for, four different corrections schemes using likely $^{230}\text{Th}/^{232}\text{Th}_0$ ratios were tested: a bulk Earth, isochron-derived (detrital), live coral (hydrogenous) and site-specific model $^{230}\text{Th}/^{232}\text{Th}$ value that accounts for both detrital and hydrogenous sources. The corrected ^{230}Th ages were then compared with the 'true', independently constrained age of the coral death assemblage to evaluate which scheme returns the most accurate ^{230}Th age.

2.5.1. Bulk Earth-based correction scheme

The 230 Th age data was corrected using the bulk-Earth activity value of 0.82 (atomic ratio ~4.4 \times 10 $^{-6}$) with an arbitrarily assigned uncertainty of ±50–100% (Richards and Dorale, 2003).

2.5.2. Live coral (hydrogenous)-based correction scheme

The live coral (hydrogenous) correction was based on 12 230 Th/ 232 Th₀ ratios obtained from live *Porites* colonies collected from Pandora, Havannah and Fantome Island in the Palm Islands region, central GBR (Clark et al., 2012), which have a weighted

mean activity ratio of 1.08 \pm 0.08 corresponding to an atomic ratio of 5.85 \pm 0.52 \times 10⁻⁶. This value, with a more conservative uncertainty of \pm 20% (to encompass the full range of variation in the 12 230 Th/ 232 Th₀ ratios), was subsequently used to correct for 230 Th₀ in the 41 *Acropora* samples in the present study.

2.5.3. Sediment (detrital)-based correction scheme

Pandora Reef is periodically reached by plumes from the Burdekin River and nearby streams (Done et al., 2007; McCulloch et al., 2003), and as a result, the detrital ²³⁰Th/²³²Th component in the *Acropora* samples is best represented by a mean Th/U ratio of 4.8 \pm 0.9 based on 44 sediment samples from the Burdekin River catchment area measured in our laboratory (Cooper et al., 2006). Assuming secular equilibrium, this corresponds to an activity ratio of 0.65 \pm 20% (atomic ratio of 3.53 \pm 0.71 \times 10⁻⁶). This is further supported by isochron-inferred ²³⁰Th/²³²Th ratios determined using local dead *Porites* corals. U–Th analyses of five *Porites* samples of coeval material (i.e. material that was formed at the same time but with different ²³²Th/²³⁸U) defined ²³⁸U/²³²Th vs ²³⁰Th/²³²Th

isochrons with intercepts on the ²³⁰Th/²³²Th axis giving a weighted mean initial ²³⁰Th/²³²Th activity ratio of 0.61 \pm 0.02 (2 σ), which is within error of the Burdekin sediment value (Fig. 3). This isochronderived mean initial ²³⁰Th/²³²Th ratio, with a more conservative uncertainty of \pm 20% as reflected by the Burdekin sediments, is considered to approximate the value of the detrital component and is therefore used in the age correction.

2.5.4. Two-component mixing correction scheme

A new 230 Th/ 232 Th₀ correction ratio [(230 Th/ 232 Th)_{mix}] was developed to account for two major isotopically distinctive components (or end-members) contributing 230 Th₀ to the 230 Th age of the coral sample: 1) an insoluble Th component adsorbed to terrestrially derived sediments or particulates that were incorporated into the skeleton either post-mortem (major) or during coral growth (minor), and 2) a soluble or hydrogenous Th component present in the water column that was incorporated into the skeleton during growth. As both insoluble (detrital) and soluble (hydrogenous) Th components could be incorporated into the skeletal matrix during growth, initial Th in live coral skeletons itself is also a mixture of both components; therefore live corals should theoretically fall onto the binary mixing line between the two abovementioned end-members. Because of this, the (²³⁰Th/²³²Th)_{mix} ratio can be calculated for each sample using the following mixing equation:

$$\begin{pmatrix} 2^{30}\text{Th} \\ \overline{2^{32}\text{Th}} \end{pmatrix}_{\text{mix}} = \left(\begin{pmatrix} 2^{32}\text{Th}_{\text{live}} \\ \overline{2^{32}\text{Th}_{\text{dead}}} \end{pmatrix} \times \begin{pmatrix} 2^{30}\text{Th} \\ \overline{2^{32}\text{Th}} \end{pmatrix}_{\text{live}} \right) \\ + \left(\begin{pmatrix} \frac{2^{32}\text{Th}_{\text{dead}} - 2^{32}\text{Th}_{\text{live}} \\ \overline{2^{32}\text{Th}_{\text{dead}}} \end{pmatrix} \times \begin{pmatrix} 2^{30}\text{Th} \\ \overline{2^{32}\text{Th}} \end{pmatrix}_{\text{sed}} \right)$$
(1)

where ²³²Th_{dead} is the measured ²³²Th value (ppb) in the individual dead coral sample of interest. ²³²Th_{live} is 0.95 ppb, being the mean ²³²Th value of live *Porites* coral samples in and near the study area (N = 12) with a corresponding ²³⁰Th/²³²Th_{live} activity ratio of 1.08 ± 20% (atomic ratio of 5.85 × 10⁻⁶ ± 20%). This value is representative of the isotopic composition of the mixture of the detrital/hydrogenous components incorporated in the live coral skeleton during growth, and is isotopically closer to the soluble Th end-member. ²³⁰Th/²³²Th_{sed} activity ratio is representative of the terrestrially-derived insoluble Th component incorporated either post-mortem or as particulates during coral growth. This ratio is calculated to be 0.61 ± 20% (atomic ratio 3.53 × 10⁻⁶ ± 20%), based on *y*-intercept values of ²³⁰Th/²³²Th vs ²³⁸U/²³²Th isochrons defined by local dead *Porites* corals, with a conservative uncertainty of 20% to account for the variability in the region. The influence of detrital ²³⁴U/²³⁸U was considered totally negligible in our samples and was therefore not incorporated in our age calculations (See Supplementary Fig. S1).

3. Results and discussion

U–Th dating of inshore-reef coral mortality events that have occurred over relatively recent timescales (less than 200 yrs) is extremely challenging due to the presence of high and variable levels of 230 Th₀ present both in the water column and adsorbed to fine sediments or particulates (Clark et al., 2012; Cobb et al., 2003; Robinson et al., 2004; Shen et al., 2008; Yu et al., 2006). Physical separation of extraneous sources of Th from that produced by the *in situ* decay of 238 U is virtually impossible. Despite using a rigorous H₂O₂ cleaning method to help remove the bulk of sediments adhered to the coral skeleton (which is reflected by a reduction in 232 Th levels), 232 Th concentrations in the measured samples still

averaged 1.5 \pm 1.1 ppb, which is significantly higher than concentrations found in live coral skeletons (Fig. 4) and on average 50-100times higher than branching corals from the central Pacific (e.g. Burley et al., 2012; Weisler et al., 2006). The presence of high levels of 232 Th is an indication of high 230 Th₀ still present in the cleaned samples. For such young coral samples (<200 years old), high levels of 230 Th₀ can cause the measured 230 Th age to be highly inaccurate, if not meaningless. The effect high concentrations of detrital Th (as reflected by elevated ²³²Th levels) can have on the U–Th data can be seen in Fig. 5a. By comparing ²³²Th concentrations obtained from each sample with their respective uncorrected ²³⁰Th age, a negative correlation exists between increasing ²³²Th and increasing uncorrected age of the sample (Fig. 5a). As expected, where ²³²Th concentrations are high, the uncorrected ²³⁰Th ages are much older than the 'true' ages of the samples, indicating that there is a significant contribution of 230 Th₀ to their 230 Th ages. Using the skeletons of 41 Acropora corals obtained from the death assemblage whose time of death was independently constrained by long-term observations, we were able to assess the effectiveness of four different ²³⁰Th₀ correction schemes in accounting for the presence of 230 Th₀ in the coral samples. How close the corrected 230 Th ages matched the timing of the 1998 bleaching event was the measure of success.

The conservative bulk-Earth ²³⁰Th/²³²Th activity ratio of 0.82 (atomic ratio 4.4×10^{-6}) (Richards and Dorale, 2003) is considered suitable for the correction of the ²³⁰Th₀ component in corals where the dominant source of detrital Th is terrestrially derived [e.g. Clark et al. (2012); and Shen et al. (2008)]. However, the large arbitrarily assigned uncertainty of 50–100% can result in excessively large age uncertainties, which, although acceptable for dating events/processes thousands of years ago, will render the corrected ages meaningless. The positive correlation between the corrected ²³⁰Th age and ²³²Th concentration (Fig. 5b), suggests that the ²³⁰Th age data are overcorrected. The resulting poor age precision precludes the identification of a specific episode of mortality (Fig. 6; Table 2).

Using live corals of known age (Clark et al., 2012; Cobb et al., 2003; Shen et al., 2008) and ambient seawater thorium isotopic measurements (Shen et al., 2008), site-specific 230 Th/ 232 Th₀ ratios can be constrained by comparing the 230 Th ages of the corals with their 'true' or absolute ages and can be considered close to representing the hydrogenous 230 Th/ 232 Th, as initial Th in live corals is mainly derived from (but not limited to) seawater during growth (Shen et al., 2008). Using this method, 12 samples of known age obtained from Porites corals provided an alternative correction for ²³⁰Th₀ (Clark et al., 2012). Yet despite using the average live *Porites* value, the corrected ²³⁰Th ages still show a positive trend with ²³²Th, suggesting that this correction scheme again over-corrected the ²³⁰Th₀ component in the dead coral skeletons (Fig. 5c; Table 2). The significantly higher ²³²Th concentrations in the dead *Acropora* coral samples [1.5 \pm 1.1 ppb (N = 41)] compared to their living counterparts $[0.15 \pm 0.18 \text{ ppb} (N = 7)]$, suggests that much of the Th in these corals was incorporated after death (Fig. 4). A live coral 230 Th/ 232 Th₀ ratio is thus not suitable for 230 Th₀ correction, as it does not take into consideration the contribution of an insoluble Th component with a contrasting 230 Th/ 232 Th₀ ratio. The isochron-derived 230 Th/ 232 Th₀ activity ratios [weighted

The isochron-derived ²³⁰Th/²³²Th₀ activity ratios [weighted mean 0.61 \pm 0.02 (2 σ)] obtained from multiple individual *Porites* coral samples from Pandora Reef and adjacent islands, most likely reflect ambient sediment values and the primary source of detrital ²³⁰Th in our samples (Fig. 3). These low ²³⁰Th/²³²Th ratios are within error of those calculated from 44 trapped sediment samples from the Burdekin River catchment (0.65 \pm 20%; Cooper et al., 2006) whose plumes periodically reach Pandora Reef (Done et al., 2007). Moreover, when all the U and Th isotopic data are plotted in ²³⁰Th/²³²Th –²³⁸U/²³²Th space (Fig. 7), all values fall on a straight



Fig. 6. a) Annual Burdekin River discharge in mega litres for the years 1981–2005 measured at the Burdekin River station at Clare site 120006B (source: Queensland Department of the Environment and Resource Management); b) Maximum annual sea surface temperatures for 1×1 grid at 146.5°E, 18.5°S (Source: NOAA Reyn and SmithOlv2); c) Percent coral cover of the genus *Acropora*. Video footage was collected by the Australian Institute of Marine Science at sites V1, V2, t1-4 and t5. Photographs were taken by Done et al. (2007) at site P1 (see Fig. 1) d) Relative probability plot of 41 U–Th ages obtained from dead *Acropora* corals corrected for 230 Th/ 232 Th₀ using a two-component (blue), isochron-derived detrital component (similar to Burdekin River sediments) (red), live coral (green) and Bulk Earth (yellow) value. The height and width of the curve represents the number of samples that date to the same time period and associated error, respectively. Orange vertical bars represent bleaching years, light

line, with the intercept at the y-axis corresponding to an 230 Th/ 232 Th₀ activity ratio of 0.64 \pm 0.04 which is analytically indistinguishable from the values constrained using the other two independent methods described above. However, the use of the isochron derived 230 Th/ 232 Th ratio of 0.61 \pm 20% for correction vielded ²³⁰Th ages that centre around a peak of ~1994 (Fig. 6d; Fig. S2b; Table 2; Table 3); which does not match the timing of the observed 1997/1998 mortality event. Although coral mortality did occur as a result of multiple flood plumes and elevated SSTs in 1994 at Pandora Reef, overall mortality at this time was reported to be less than 1% of the total cover for this genus (DeVantier et al., 1997). The 1994 disturbance event is therefore unlikely to have been responsible for the normally distributed age population produced by 41 coral fragments collected over a distance of more than 80 m. The failure of this correction scheme is due to the fact that the hydrogenous ²³⁰Th component with higher ²³⁰Th/²³²Th was ignored. Similar results have also been shown for Lake Lahontan carbonates, where samples containing hydrogenous 230 Th and corrected using isochron derived 230 Th/ 232 Th $_0$ ratios were also too old (Lin et al., 1996).

The most accurate correction scheme accounted for the two isotopically contrasting components contributing ²³⁰Th₀: 1) detrital ²³⁰Th from terrestrially-derived insoluble Th components incorporated into the coral skeleton either during growth (minor) or post-mortem (major), and 2) hydrogenous ²³⁰Th adsorbed on detritus or present in the water column and directly incorporated into the aragonite matrix (Haase-Schramm et al., 2004; Lin et al., 1996). For living reef corals, the primary source of 230 Th₀ is mainly from the dissolved fraction in seawater (Shen et al., 2008), although the presence of a minor detrital component in seawater cannot be ruled out at inshore settings. Live corals may scavenge a small amount of the detrital component present in the seawater column in particulate/colloidal forms as it switches from a passive autotroph to active heterotroph. It is likely that the minor detrital component in live corals is variable among different species and in different environmental settings: higher in Porites compared to Acropora (Clark et al., 2012; this study), and higher in inshore compared to offshore settings (cf. Burley et al., 2012; Roff et al., 2013; Yu et al., 2012a, 2012b). In this regard, the mean 230 Th/ 232 Th ratios obtained from both live *Porites* and *Acropora* should theoretically fall on a binary mixing line between the hydrogenous (seawater) and detrital (sediment) end-members. Postmortem, corals can no longer actively exclude sediments which become adsorbed into the porous skeletal matrix of the coral (Lasker, 1980). As a result, both sources of Th₀ need to be corrected independently using the binary mixing model in order to achieve accurate U-Th ages.

In this study, live *Porites* and isochron-derived ²³⁰Th/²³²Th₀ ratios were used to approximate the isotopically distinctive hydrogenous and detrital end-membersin the dead *Acropora* corals, respectively. When the mean ²³⁰Th/²³²Th ratios for the live *Porites*, live *Acropora* and isochron-derived detrital values from dead *Porites* are plotted in a ²³⁰Th/²³²Th versus 1/²³²Th diagram, all four types of samples fall on a binary mixing line between two end-members: a hypothetical seawater (hydrogenous) component and a terrestrial (detrital) component (Fig. 4). Interestingly, the live *Acropora* coral ²³⁰Th/²³²Th ratios fall between those determined for the dissolved and particulate fraction of seawater analysed from continental shelf settings in the western Pacific and eastern Indian Ocean (Shen et al., 2008), suggesting that this value may be an accurate representation

blue bars represent years of major flooding and grey bars represent cyclone events. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

MC-ICP-MS²³⁰Th ages for dead Acropora coral samples collected in May 2008 corrected using the bulk Earth, live coral, sediment and two-component²³⁰Th/²³²Th₀ correction value

Sample name	Uncorr. ²³⁰ Th age (AD)	Bulk Earth (AD) ^a	Live coral (AD) ^b	Sediment (AD) ^c	Two-component (AD) ^d
PanS1T1.2	1979.8 ± 0.7	1998.9 ± 9.6	2004.9 ± 5.0	1993.9 ± 2.9	1998.4 ± 3.8
PanS1T1.3	1986.9 ± 0.4	1995.2 ± 4.2	1997.8 ± 2.2	1993.1 ± 1.3	1997.7 ± 2.2
PanS1T1.4	1970.7 ± 0.6	1997 ± 13	2004.8 ± 6.8	1989.9 ± 3.9	1994.3 ± 4.8
PanS1T1.5	1981.6 ± 0.5	1996.5 ± 7.5	2001.2 ± 4.0	1992.6 ± 2.3	1997.3 ± 3.2
PanS1T1.7	1993.1 ± 0.5	1995.2 ± 1.2	1995.9 ± 0.8	1994.7 ± 0.6	1999.2 ± 1.3
PanS1T1.8	1985.5 ± 0.4	1996.8 ± 5.7	2000.3 ± 3.0	1993.9 ± 1.7	1998.5 ± 2.6
PanS1T1.9	1970.8 ± 0.6	2002 ± 15	-ve age	1994.0 ± 4.7	1998.4 ± 5.5
PanS1T1.10	1988.5 ± 0.4	1996.1 ± 3.8	1998.5 ± 2.0	1994.1 ± 1.2	1998.5 ± 2.1
PanS1T1.11	1972.8 ± 0.6	2001 ± 14	2009.9 ± 7.4	1993.7 ± 4.2	1998.2 ± 5.1
PanS1T1.12	1970.9 ± 0.7	1998 ± 14	2007.1 ± 7.3	1991.2 ± 4.1	1995.7 ± 5.0
PanS1T2.1	1987.1 ± 0.3	1992.1 ± 2.5	1993.7 ± 1.4	1990.8 ± 0.8	1995.2 ± 1.7
PanS1T2.3	1973.8 ± 0.6	2000 ± 13	2008.0 ± 6.9	1993.0 ± 3.9	1997.6 ± 4.8
PanS1T2.4	1984.2 ± 0.5	1997.0 ± 6.4	2001.0 ± 3.4	1993.7 ± 2.0	1998.1 ± 2.8
PanS1T2.5	1991.0 ± 0.4	1995.0 ± 2.0	1996.2 ± 1.1	1993.9 ± 0.7	1998.7 ± 1.6
PanS1T2.6	1991.6 ± 0.5	1995.4 ± 2.0	1996.6 ± 1.1	1994.4 ± 0.7	1999.2 ± 1.6
PanS1T2.7	1984.2 ± 0.6	1997.7 ± 6.8	2001.9 ± 3.6	1994.2 ± 2.1	1998.4 ± 2.9
PanS1T2.8	1965.7 ± 0.7	1998 ± 16	2007.9 ± 8.5	1989.5 ± 4.8	1994.3 ± 5.7
PanS1T2.9	1986.4 ± 0.5	1993.8 ± 3.7	1996.1 ± 2.0	1991.9 ± 1.2	1996.2 ± 2.0
PanS1T2.10	1986.3 ± 0.6	1996.2 ± 5.0	1999.3 ± 2.7	1993.6 ± 1.6	1998.4 ± 2.5
PanS1T2.11	1982.1 ± 0.5	1997.9 ± 7.9	2002.8 ± 4.2	1993.8 ± 2.4	1998.2 ± 3.3
PanS1T2.12	1977.7 ± 0.5	1999 ± 11	2005.6 ± 5.6	1993.4 ± 3.2	1997.7 ± 4.0
PanS1T3.3	1990.9 ± 0.4	1994.9 ± 2.0	1996.1 ± 1.1	1993.8 ± 0.7	1998.1 ± 1.5
PanS1T3.4	1984.1 ± 0.4	1996.6 ± 6.3	2000.5 ± 3.3	1993.3 ± 1.9	1997.8 ± 2.8
PanS1T3.5	1990.9 ± 0.5	1993.4 ± 1.4	1994.2 ± 0.8	1992.7 ± 0.6	1997.3 ± 1.4
PanS1T3.6	1989.7 ± 0.4	1993.7 ± 2.0	1994.9 ± 1.1	1992.6 ± 0.7	1997.0 ± 1.5
PanS1T3.7	1990.1 ± 0.5	1996.6 ± 3.3	1998.6 ± 1.8	1994.9 ± 1.1	1999.2 ± 1.9
PanS1T3.8	1990.7 ± 0.4	1995.4 ± 2.4	1996.9 ± 1.3	1994.2 ± 0.8	1998.5 ± 1.6
PanS1T3.11	1980.3 ± 0.4	1998.1 ± 8.9	2003.6 ± 4.7	1993.4 ± 2.7	1997.6 ± 3.5
PanS1T3.13	1974.1 ± 0.7	2000 ± 13	2008.2 ± 6.9	1993.3 ± 3.9	1997.6 ± 4.8
PanS1T3.15	1978.8 ± 0.5	2000 ± 11	2007.1 ± 5.7	1994.7 ± 3.2	1999.0 ± 4.1
PanS1T4.1	1989.2 ± 0.3	1993.6 ± 2.2	1994.9 ± 1.2	1992.4 ± 0.7	1996.7 ± 1.5
PanS1T4.3	1985.1 ± 0.5	1997.3 ± 6.1	2001.1 ± 3.2	1994.1 ± 1.9	1998.5 ± 2.7
PanS1T4.4	1988.2 ± 0.4	1995.1 ± 3.5	1997.3 ± 1.8	1993.3 ± 1.1	1997.7 ± 1.9
PanS1T4.5	1987.6 ± 0.4	1996.3 ± 4.4	1999.1 ± 2.3	1994.1 ± 1.4	1998.5 ± 2.2
PanS1T4.6	1990.6 ± 0.3	1993.4 ± 1.5	1994.3 ± 0.8	1992.7 ± 0.5	1997.2 ± 1.4
PanS1T4.7	1985.5 ± 0.5	1992.4 ± 3.5	1994.5 ± 1.9	1990.6 ± 1.1	1995.1 ± 2.0
PanS1T4.9	1988.2 ± 0.3	1994.9 ± 3.4	1997.0 ± 1.8	1993.1 ± 1.1	1997.5 ± 2.0
PanS1T4.10	1984.7 ± 0.4	1996.7 ± 6.1	2000.5 ± 3.2	1993.6 ± 1.8	1997.9 ± 2.7
PanS1T4.12	1989.7 ± 0.4	1994.8 ± 2.6	1996.4 ± 1.4	1993.5 ± 0.8	1997.8 ± 1.7
PanS1T4.13	1990.4 ± 0.3	1995.2 ± 2.4	1996.6 ± 1.3	1993.9 ± 0.8	1998.3 ± 1.6
PanS1T4.17	1981.8 ± 0.5	1998.0 ± 8.1	2003.1 ± 4.3	1993.8 ± 2.5	1998.2 ± 3.3
1 1 230Th (AT					

Th age (AD) was calculated using Isoplot/EX 3.0 program (Ludwig, 2003b).

Corrected ²³⁰Th ages were calculated using.

^a Bulk Earth value = $0.82 \pm 50\%$ (atomic value ~ $4.4 \times 10^{-6} \pm 50\%$). ^b Region specific ²³⁰Th/²³²Th₀ value for the Palm Islands derived from live *Porites* of known age = $1.083 \pm 20\%$ (atomic value of $5.7 \times 10^{-6} \pm 20\%$).

Burdekin River sediment value derived from 40 ICP-MS measurements = 0.61 \pm 20% (atomic value 3.53 \times 10⁻⁶ \pm 20%).

^d Two-component correction value calculated using Equation (1).

of seawater values (although this is yet to be confirmed). While values obtained from live Acropora corals would better reflect the hydrogenous end-member in the dead Acropora samples dated in this study, they were not used in the equation for two reasons: 1) concentrations of ²³²Th in the live *Acropora* samples are extremely low $(0.15 \pm 0.18 \text{ ppb})$ and difficult to measure accurately; 2) it is also difficult to independently constrain the 'true' age of a sample from an Acropora branch in order to determine ²³⁰Th/²³²Th₀ without cross-referencing with annual growth bands (from X-rays) or elemental cycles (using ICP-MS). The assumption that we are sampling from within one year of growth is based on a few observational studies of annual extension rates for Acropora colonies from the inshore GBR and other turbid reef environments (Crabbe and Smith, 2005; Diaz-Pulido et al., 2009). Moreover, directly determining seawater 230 Th/ 232 Th ratios may also be an inaccurate estimate of the hydrogenous 230 Th/ 232 Th₀ component in the dead coral skeleton as seawater ²³²Th concentrations are highly variable over spatial and short-term temporal scales. For example, dissolved ²³²Th concentrations measured from coastal environments by Shen et al. (2008) were on average ~80 times higher than the bulk ²³²Th concentrations (excluding organics) in seawater from the Bahamas (0.00828 versus 0.00010 ppb, respectively). Shen et al. (2008) also found higher concentrations of ²³²Th at high tide compared to low tide. Thus an understanding of site-specific seawater ²³²Th concentrations is needed. In addition, it is difficult to know the ²³²Th concentrations of the local seawater at the time when the coral died.

When the measured 230 Th/ 232 Th data for the dead Acropora corals are corrected using the two-component mixing equation (Eqn. (1)), the isotopic variations shift towards this mixing line, with most of them falling between the detrital end-member and the live Porites 230 Th/ 232 Th ratio. The 230 Th age population corrected using the two-component mixing scheme also becomes normally distributed, as can be seen when the data are plotted as a relative probability plot [that incorporates both the mean and 2sigma uncertainties of the individual dates using the Isoplot Program (Ludwig, 2003a,b)]. The peak value of this distribution is ~1998 AD (weighted mean 1998.2 ± 0.3 AD, MSWD = 1.1) (Fig. 6d and Table 3), which is within error of the timing of the 1998 mass bleaching event (Fig. 6; Fig. S2a; Table 2). At this time, long-term



Fig. 7. U and Th isotope measurements for 41 dead Acropora samples that reportedly died as a result of the 1998 bleaching event show a linear relationship in (²³⁰Th/²³²Th)- $(^{238}U)^{232}Th)$ space. This plot likely reflects a mixing line and is not a true isochron (due to multiple sources of $^{230}Th_0$). The *y*-intercept is equivalent to the $^{230}Th/^{232}Th_0$ in the detrital phase. This was determined to be $0.64 \pm 0.04 (1\sigma)$, which is similar to isochron derived 230 Th/ 232 Th_{nr} values from dead *Porites* colonies (0.61 ± 0.01 (1 σ)), as well as ICP-MS measurements of Burdekin River sediments (Th/U = 4.8 ± 1.0 or 230 Th/ 232 Th = 0.65 ± 0.2).

monitoring datasets from sites similar in community composition less than 500 m away (Fig. 1) and regional scale observations documented almost 100% mortality in Acropora (Done et al., 2007; Maynard et al., 2008; Sweatman et al., 2005). In contrast, the probability distribution patterns of corrected ²³⁰Th ages derived from all the other three correction schemes are all skewed to some extent, suggesting those schemes are insufficient to correct for 230 Th₀ components in the samples.

Had the death assemblage been derived from a number of events (e.g. cyclones, predators or freshwater inundation during previous decades), we would not have expected such a tightly constrained age estimate (Edinger et al., 2001). For example, if the death assemblage included the skeletal material from mortality events spanning 5-10 generations, using the mean age of reproductive maturity for Acropora species as the generation time (Van Oppen et al., 2000) [i.e. 3-8 years (Csaszar et al., 2010; Wallace, 1999)], a random sample of skeletons would be between 15 and 80 years old (Van Oppen et al., 2000). This is a much greater range than the two-component corrected 2σ age range observed for our 230 Th ages (11.6 ± 1.9 to 16.4 ± 5.8 years, or 1999.3 ± 1.9 to 1994.3 \pm 5.8 AD). Having said that, there appears to be a slight tailing in the U-Th data in the relative probability plot of Fig. 6d and the individual age plot of Figure S3, implying minor mortality that may have occurred during the 1994 bleaching/flood event. However, this has little impact on the well-defined 1998 AD age peak recorded in Fig. 6d.

Table 3

Summary statistics of ²³⁰Th ages derived from 41 dead branching *Acropora* corals.

Correction used	Mean age (A.D.)	Median age (A.D.)	S.D.	Age range (A.D.)	Weighted mean age ± 2σ (A.D.) ^a	MSWD ^b
Uncorrected	1983.9	1985.5	7.0	1965.7-1993.1	1985.8 ± 1.8	671
Bulk Earth	1996.5	1996.5	2.3	1992.1-2002.1	1994.7 ± 0.5	0.8
Live coral	2000.1	1999.2	4.6	1993.7-	1996.2 ± 0.7	7.4
Sediment	1993.2	1993.6	1.3	1989.5-1994.9	1993.3 ± 0.3	3.8
Two-component	1997.7	1997.9	1.2	1994.3-1999.2	1997.8 ± 0.3	1.1

Weighted mean calculated using Isoplot/Ex (Ludwig, 2003b).

^b MSWD = Mean Square of Weighted Deviates. The MSWD is the sum of squares of weighted residuals divided by the degrees of freedom. MSWD values greater than unity (i.e. >1) indicate either underestimated analytical errors, or the presence of non-analytical 'geological' scatter (Ludwig, 2003b).

4. Conclusions

The congruence between the ²³⁰Th age data corrected using the two-component equation and the documented catastrophic loss of Acropora both at Pandora Reef and over a much broader scale as a result of the 1998 bleaching event affirms that it is possible to use the U-Th method to accurately date recently dead coral skeletons from the death assemblage. For dead corals (including both massive and branching growth forms) from inshore reef settings, it is necessary to correct for both hydrogenous (dissolved) and detrital ²³⁰Th incorporated during growth as well as post-mortem (see also Clark et al., 2014). This approach can then be used as a powerful tool for researchers and managers to identify mortality events and estimate rates of recovery in a historical context. For example, following the 1997–1998 bleaching event, it was predicted that it would not be until 2008-2010 that coral on the shallow fore-reef could recover to its 1981 status (Done et al., 2007). Our observations in 2008 and 2009 suggest that recovery severely lags behind this prediction due to an apparent failure of coral recruitment at Pandora Reef. However, for the vast majority of coral reefs there are no such long-term ecological data. On those reefs, high-precision U–Th dating with high sample throughput can now be used with surety on degraded coral reefs to determine when the reefs were damaged and hence ascertaining not only the drivers but also the time that has been available for their recovery post-disturbance; critical issues that have received insufficient attention in coral reef science (Hughes et al., 2010). 'Time for recovery' is a key variable in evaluating a damaged reef's recovery performance against established benchmarks (Done et al., 2010). As much as it provides the timescale, high-precision U-Th dating thus has important applications that extend beyond scientific understanding and into the realm of coral reef policy and management.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http:// dx.doi.org/10.1016/j.quageo.2014.05.002.

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