Variation of initial ^{230}Th/^{232}Th and limits of high precision U-Th dating of shallow-water corals

Chuan-Chou Shen,[1,*] Kuei-Shu Li,[1] Kerry Sieh,[2] Danny Natawidjaja,[2] Hai Cheng,[3] Xianfeng Wang,[3] R. Lawrence Edwards,[3] Doan Dinh Lam,[4] Yu-Te Hsieh,[1] Tung-Yung Fan,[5] Aron J. Meltzner,[2] Fred W. Taylor,[6] Terrence M. Quinn,[6,7] Hong-Wei Chiang,[1] K. Halimeda Kilbourne [7,8]

[1]Department of Geosciences, National Taiwan University, Taipei, Taiwan, ROC

[2]Tectonics Observatory, California Institute of Technology, Pasadena, California, USA

[3]Department of Geology and Geophysics, University of Minnesota, Minnesota, USA

[4]Institute of Geology, Vietnamese Academy of Science and Technology, Hanoi, Vietnam

[5]National Museum of Marine Biology and Aquarium, Pingtung, Taiwan, ROC

[6]Institute for Geophysics, John A. and Katherine G. Jackson School of Geosciences, University of Texas at Austin, Austin, Texas, USA

[7]College of Marine Science, University of South Florida, St. Petersburg, Florida, USA

[8]Physical Sciences Division R/PSD1, NOAA, Earth System Research Laboratory, 325 Broadway, Boulder, Colorado 80305, USA

* Corresponding author: National Taiwan University

(e-mail) river@ntu.edu.tw

(tel) 886-2-3366-5878; (fax) 886-2-3365-1917.

Abstract

One hundred eighty U-Th data, including 23 isochrons on 24 pristine modern and Holocene corals and 33 seawater samples, were analyzed using sector-field mass spectrometry to understand the variability of initial $^{230}Th/^{232}Th$ ($^{230}Th/^{232}Th_0$). This dataset allows us to further assess the accuracy and precision of coral ^{230}Th dating method. By applying quality control, including careful sampling and subsampling protocols and the use of contamination-free storage and workbench spaces, the resulting low procedural blanks give an uncertainty in age of only ± 0.2-0.3 yr for 1-2-g of coral sample. Using site-specific $^{230}Th/^{232}Th_0$ values or isochron techniques, our study demonstrates that corals with an age less than 100 years can be ^{230}Th-dated with precisions of ± 1 yr. Six living subtidal coral samples were collected from two continental shelf sites, Nanwan off southern Taiwan in the western Pacific and Son Tra off central Vietnam in the South China Sea; one coral core was drilled from an open ocean site, Santo Island, Vanuatu, in the western tropical Pacific; and modern and fossil intertidal coral slabs, 17 in total, were cut from six sites around the islands of Simeulue, Lago, North Pagai and South Pagai of Sumatra in the eastern Indian Ocean. The results indicate that the main source of thorium is the dissolved phase of seawater, with variation of $^{230}Th/^{232}Th_0$ depending on local hydrology. With intense input of terrestrial material, low $^{230}Th/^{232}Th_0$ atomic ratios of 4.9 × 10^{-6} and 3.2 × 10^{-6} with a 10% variation are observed in Nanwan and Son Tra, respectively. At the Santo site, we find a value of 5.6 × 10^{-6} at 4 horizons and one high value of 24 × 10^{-6} in a sample from AD 1974.6 ± 0.5, likely due to the upwelling of cold water during a La Niña event between AD 1973 and 1976. The natural dynamics of $^{230}Th/^{232}Th_0$ recorded in the intertidal corals at sites in the Sumatran islands are

complicated so that this value varies significantly from 3.0 to 9.4 $\times$ 10^{-6}. Three of the 141 modern coral ^{230}Th ages differ from their true ages by -23 to +4 years, indicating the presence of detrital material with anomalous ^{230}Th/^{232}Th values. Duplicate measurement of coeval subsamples is therefore recommended to verify the age accuracy. This improved high precision coral ^{230}Th dating method raises the prospects of refining the age models for band-counted and tracer-tuned chronologies and of advancing coral paleoclimate research.

1. Introduction

Geochemical and isotopic compositions in shallow-water coralline aragonite have been widely used as proxies to reconstruct paleoclimatic conditions (e.g., Wellington and Dunbar, 1995; Shen et al., 1996; Beck et al., 1997; Gagan et al., 1998; McCulloch et al., 1999; Hendy et al., 2002; Culter et al., 2003; McCulloch et al., 2003; Shen et al., 2005a; Thompson and Goldstein, 2005). Since the 1990s, weekly to monthly resolution coral climate records over decadal, centennial, or millennial time scales have been published (e.g., Gagan and Chivas, 1995; Crowley et al., 1997; Evans et al., 1998; Linsley et al., 2000; Urban et al., 2000; Hendy et al., 2002; Cobb et al., 2003a; McCulloch et al., 2003; Kilbourne et al., 2004; Sun et al., 2004; Nyberg et al., 2007). For centuries-old modern and young fossil corals, age uncertainties of $\pm$ 1 to 5 yrs limit our ability to accurately determine the timing of climatic events and the precisely cross-correlate coral records to other high resolution proxy records.

Coral chronology can be determined by bulk annual density band counting (e.g., Dodge and Brass, 1984; Cole and Fairbanks, 1990; Gagan et al., 1995; Crowley et al., 1997; Linsley et al., 1999; Kilbourne et al., 2004), or by tuning tracer records using methods adapted from dendrochronology and sediment core chronostratigraphy (e.g., Linsley et al., 1999; Cobb et al., 2003b; Hendy et al., 2003). Difficulty arises when using these methods for precise absolute chronological control in the following cases: (1) living corals with banding discontinuities caused by climatic and/or tectonic anomalies (e.g., Sieh et al., 1999; Hendy et al., 2003; Natawidjaja et al., 2004), (2) corals living in the equatorial oceans with intrinsically small seasonal climatic cycles (as low as 0-2 ^{o}C of seasonal temperature change), and (3) dead coral heads and fossils without absolute ages and/or mortality event-related age markers (Glynn et al.,

1983; Yu et al., 2004),

The [230]Th dating method provides an ideal absolute chronological tool for coral because of high uranium levels of 2-3 parts per million (ppm) in their skeletons (Bateman, 1910; Barnes et al., 1956; Broecker and Thurber, 1965; Kaufman and Broecker, 1965; Edwards et al., 1987; Edwards et al., 1988; Edwards et al., 2003). The [230]Th age equation, including initial [230]Th/[232]Th, as reported by Edwards et al. (2003) (modified from Bateman (1910) and Broecker (1963)):

$$\frac{^{230}Th}{^{238}U} = 1 + \left(\left(\frac{^{232}Th}{^{238}U}\right)\left(\frac{^{230}Th}{^{232}Th}\right)_i - 1\right)e^{-\lambda_{230}t}$$

$$+ \frac{\delta^{234}U_m}{1000}\left(\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\right)\left(1 - e^{(\lambda_{234}-\lambda_{230})t}\right), \qquad (1)$$

where all isotope ratios are activity ratios, the λ's are decay constants, t is the [230]Th age, and $(^{230}Th/^{232}Th)_i$ is the initial [230]Th/[232]Th ratio. The [234]U/[238]U ratio has been formulated into δ-notation, which denotes the fractionational enrichment in the [234]U/[238]U ratio at secular equilibrium in parts per thousand. The observed value is given by $\delta^{234}U_m = \{[(^{234}U/^{238}U)_m/(^{234}U/^{238}U)_{eq}]-1\} \times 10^3$ where $(^{234}U/^{238}U)_m$ is the measured activity ratio and $(^{234}U/^{238}U)_{eq}$ is the ratio of 1 at secular equilibrium (Edwards et al., 1987). As $(^{232}Th/^{238}U)$ can be measured, an accurate [230]Th age can be determined if $(^{230}Th/^{232}Th)_i$ is known.

The first high accuracy young coral [230]Th dating techniques with precisions of ± 3 yrs were developed using thermal ionization mass spectrometry by Edwards et al. (1988). Their study suggested the [230]Th$_0$ is not significant within errors of ± 3 yrs in typical shallow-water corals with [232]Th levels less than 100 parts per trillion (ppt) and low initial [230]Th/[232]Th $(^{230}Th/^{232}Th_0)$ of 2 to 4 × 10^{-6} (atomic ratio, hereafter). However, development of a coral [230]Th dating method with a precision better than ± 1 to 2 yrs is difficult with uncertainties mainly affected by the thorium levels and the

uncertainty of the correction for initial ^{230}Th (^{230}Th$_0$) (Zachariasen et al., 1999; Cobb et al., 2003b).   For example, uncertainties of 25%, 50%, and 100% for a ^{230}Th/^{232}Th$_0$ value of 4 $\times$ 10^{-6}, give approximate age errors of $\pm$ 3, $\pm$ 5, and $\pm$ 10 yrs, respectively, for a decades-centuries-old coral with 2.4 ppm uranium and 1000 ppt thorium.

The initial thorium in the coralline structure is influenced both by scavenging processes between seawater and the coral skeleton, and by thorium association with detrital material incorporated into the crystal matrix (Cobb et al., 2003b; Edwards et al., 2003).   Cobb et al. (2003b) proposed different potential sources of coral ^{230}Th$_0$, including (1) wind-blown dust with a ^{230}Th/^{232}Th ratio of 4 $\times$ 10^{-6}; (2) surface seawater containing dissolved and particulate thorium with a ^{230}Th/^{232}Th ratio of 5-10 $\times$ 10^{-6}; (3) deep seawater Th with a ^{230}Th/^{232}Th ratio of up to 2 $\times$ 10^{-4} (Moran et al., 2002); and (4) carbonate sands with ^{230}Th/^{232}Th ratios as high as 1 $\times$ 10^{-2}.   Different sources and the non-conservative property of thorium in seawater (e.g., Broecker et al., 1973) could result in diverse coral ^{230}Th/^{232}Th$_0$ ratios.   The range of ^{230}Th/^{232}Th$_0$ values observed in intertidal corals at different sites near Sumatra is 6.5 $\pm$ 6.5 $\times$ 10^{-6} (Zachariasen et al., 1999).   High values of 10 to 20 $\times$ 10^{-6} are documented in Palmyra living corals (Cobb et al., 2003b).   Modern Bahamas corals have 60 to 80 $\times$ 10^{-6} of ^{230}Th/^{232}Th$_0$ values, intermediate between those of normal surface water and of bank-top water (Robinson et al., 2004).   Clearly, the ^{230}Th$_0$ incorporated into a growing aragonitic skeleton needs to be well constrained or its uncertainty may lead to significant error in the ^{230}Th ages of young corals.

The precision and accuracy of coral ^{230}Th dating techniques are affected by not only ^{232}Th content and variability of ^{230}Th$_0$, but also by the ^{230}Th introduced during sample collection and treatment.   The previously published lowest procedural ^{230}Th blank, 0.0015 $\pm$ 0.0015 femto-mole (fmol) (Cheng et al., 2000), is equivalent to an uncertainty of $\pm$ 0.4 yr.   Coupled with instrumental constraints (Goldstein and

Stirling, 2003), the best coral U-Th dating procedures yielded an accuracy and precision of $\pm$ 2 to 3 yrs (after Edwards et al., 1987; Edwards et al., 2003).

Three novel approaches were taken in this study to better understand the natural variability of coral $^{230}Th/^{232}Th_0$ in the oceans and to establish a high precision ^{230}Th dating method. First, chemical and analytical techniques were refined. The procedural ^{230}Th blank was reduced to as low as 0.0008 fmol, corresponding to an error of $\pm$ 0.2 yr for a 1-g coral sample. Analytical uncertainty in ^{230}Th using inductively coupled plasma sector field mass spectrometry (ICP-SF-MS) techniques (Shen et al., 2002) was reduced to an equivalent age uncertainty of about $\pm$ 0.2 to 0.3 yr. Second, concentrations and isotopic compositions of dissolved and particulate thorium were analyzed to distinguish the contributions of the two phases to the coral skeletal matrix. Finally, the variability of $^{230}Th/^{232}Th_0$ at different hydrological settings was evaluated. Subtidal and intertidal modern and fossil corals, 24 in total, from the continental shelf sites of Nanwan in the western Pacific and Son Tra in the South China Sea, the open ocean site of Santo in the western tropic Pacific, and six sites among the Sumatran islands in the eastern Indian Ocean, were analyzed to investigate the influences of terrestrial input, ^{230}Th ingrowth from the underlying substrate, and different water masses on $^{230}Th/^{232}Th_0$. (4) Comparisons of the corals' ^{230}Th ages and independently-derived absolute ages, as well as isochron plots, were used to constrain spatial and temporal variations of $^{230}Th/^{232}Th_0$ values on annual to millennial timescales at the selected sites.

2. Materials and methods

2.1. *Field collection*

Seawater and massive *Porites* coral samples were collected at sites with different hydrological settings from Nanwan, Taiwan, Son Tra Island, central Vietnam, southern Espiritu Santo Island, Vanuatu, and Sumatran islands, (Fig. 1, Figs. A1-A4).

2.1.1. Nanwan

Nanwan ($21^{o}55$'N, $120^{o}47$'E) is a semi-enclosed basin on the southern tip of Taiwan (Fig. A1).   Two subtidal coral heads, NW0310 and NW0402, each 30-40 cm in diameter, were collected from the water intake channel of the nuclear power plant located at the northeastern corner of Nanwan in October 2003 and February 2004 at depths of 4 m and 2 m, respectively.

A hydrological diagram of seawater Sr/Ca versus (vs.) salinity shows that Nanwan water is composed mainly of three endmembers: 25% offshore surface water, 75% tidally-induced upwelled subsurface cold water from depths of 100-200 m offshore, and an additional 0-2.5% fresh water in the summer (Lee et al., 1997; Shen et al., 2005b).   In order to understand the influences of upwelled seawater and tide height on the local seawater ^{232}Th level and ^{230}Th/^{232}Th ratio, 21 consecutive 500-ml samples of filtered seawater were collected from the water intake channel of the Third Nuclear Power Plant, Nanwan, on July 31 and August 1, 2004, at a depth of 1 m (Fig. A1, Table A1).   Seawater was filtered with an acid-cleaned 0.45-μm acetate cellulose filter and stored acidified in an acid-cleaned polyethylene bottle by adding 0.5-1 g 14 N HNO$_3$ in the field (Fig. A1).   Eight samples of suspended particulate

matter, filtered from 5 L seawater with 0.45-μm acetate cellulose filters, were collected between October 28 and 30, 2004, at the same position where filtered seawater samples were collected (Fig. A1, Table A2). Suspended particulate matter samples were stored in acid-cleaned polyethylene bags at 5 ^{o}C in a refrigerator before chemical analysis. The same seawater sampling process was applied at other sites. One calibrated underwater thermometer with precision of 0.05 ^{o}C was placed next to the NW0402 coral sampling site to record sea surface temperature and monitor the timing of cold upwelled water intrusion. Tide height data monitored at Houbihu, 2 km south of the sample collection site (Fig. A1), are from the Central Weather Bureau.

2.1.2. Son Tra and Santo

One living 35-cm *Porites* coral head, ST0506, and 2 filtered 1-L seawater samples from a depth of 4 m below sea level were collected at Son Tra Island, a near-shore island in central Vietnam (16°13'N, 108°12'E) on June 14, 2005 (Fig. A2). The site is located at the north tip of Vung Da Nang Bay with terrestrial material influxes mainly delivered to the southern corner of the bay by the Han River. Two 1-L filtered seawater samples were also collected from a depth of 2 m at the same site.

The Santo subtidal coral core, 92MC, is 130 cm in length and 8 cm in diameter. It was drilled from a *Porites lutea* head living at a depth of 1.5 m at an open ocean site, with notable lack of riverine influence, in the passage between Malo Island and Espiritu Santo Island (15°42'S, 167°12'E), in the western tropical Pacific Ocean, in June 1992 (Kilbourne et al., 2004) (Fig. A3).

2.1.3. Sumatran islands

The Sumatran islands are located along the Sunda subduction zone, an area associated with frequent earthquakes (e.g., Briggs et al., 2006; Subarya et al., 2006). The subduction zone and Sumatran fault separate the Indian and Australian plates from the Southeast Asian plate (Fitch, 1972; McCaffrey, 1991).   Modern and fossil *Porites* slabs with thicknesses of 7 to 9 cm were cut with a chain saw from microatolls on shallow reefs within the intertidal zone at sites near Lewak (2^o56'N, 95^o48'E) and Gusong Bay (2^o23'N, 96^o20'E) on Simeulue Island; Lago (0^o03'N, 94^o32'E) in the Batu Islands; North Pagai Island (2^o35'S, 100^o06'E); and Bulasat ($3°07.6$'S, $100°18.7$'E), and Saumang ($3°07.7$'S, $100°18.6$'E) on South Pagai Island (Fig. A4). One slab, LWK05, was collected from Lewak, and two slabs, GSG05YNG and GSG05OLD, were collected from Gusong Bay between the 1st and 3rd of June 2005; the two sites are 80 km apart, on the northern tip and the southern southwest coast of Simeulue Island, respectively.   Two contiguous slabs, LG99A1 and LG00A1, were collected from the northeastern flank of Lago in 1999 and 2000, respectively (Natawidjaja et al., 2004).   The Lago site is a landward pool characterized by a wide intertidal flat with coral rubble on a carbonate-enriched sandy substrate.   Two slabs, NP00A1 and NP00A2, were collected from the eastern coast of North Pagai.   Nine samples were collected from microatolls at Bulasat and Saumang on the southwestern coast of South Pagai.   Following the same sampling procedure as at Nanwan, two 500-ml filtered seawater samples were collected at depths of 1 m and 2 m at the coral collection site in North Pagai in January 2004.

2.2. Sample storage

General storage rooms for Quaternary carbonates can have ^{230}Th blanks higher

than 0.002 fmol, which could bias dates for coral samples by as much as 30 yrs (Table A3). This cross contamination, mainly from fine powder of Quaternary samples with high ^{230}Th content, may not be easy to remove from a porous coral skeleton, even using ultrasonic cleaning methods. Subsequently, we now bag coral samples individually and isolate young and old corals from each other in our storage rooms. In addition, we isolate coral samples from other types of natural carbonate samples, such as speleothem and tufa. These procedures effectively keep storage-related ^{230}Th background levels to 2×10^{-4} fmol or less.

2.3. Coral subsampling

In this study, modern corals were subsampled in a separate class-100 laminar-flow clean working bench. For Nanwan, Son Tra, and Santo coral slabs, subsamples 0.5-1.5 cm in width were cut using a surgical blade, along growth layers, based on X-ray photographs. The widths are equivalent to 0.5 to 1 year of coral growth. For Sumatran coral slabs, subsamples were collected by either cutting with a blade or drilling with a hole-drill bit 1 cm in diameter. The drilled cylindrical subsamples were 0.8 cm in diameter. Weights of coral subsamples ranged from 0.3 to 3 g (Tables A3-A12). All were ultrasonicated with deionized water 4 to 5 times until there was no visible powder or detrital material, and then dried at 70 ^{o}C. To avoid possible cross contamination in the pretreatment process, all steps were performed on class-100 laminar flow benches.

2.4. Chemistry

U-Th chemistry was performed in the Geochemistry Technology Laboratory of the Department of Geosciences, National Taiwan University (NTU) and in the Minnesota Isotope Laboratory of the Department of Geology and Geophysics,

University of Minnesota (UMN). Chemistry for seawater samples was conducted following the procedures described in Moran et al. (2002) and Shen et al. (2003). Coral subsamples were prepared with chemical methods similar to those described by Edwards et al. (1988) and Shen et al. (2002; 2003). Samples were spiked with a ^{229}Th-^{233}U-^{236}U tracer. Uranium and thorium were separated with Fe coprecipitation and anion-exchange chromatography. The uranium and thorium aliquots were dissolved in 1% HNO_3 + 0.005 N HF for instrumental measurements (Shen et al., 2002).

2.5. Instrumental analysis

Determinations of uranium and thorium isotopic compositions and concentrations were performed using two ICP-SF-MS: a Thermo Electron ELEMENT II, housed at the Department of Geosciences, NTU, and a Finnigan ELEMENT, at the Minnesota Isotope Laboratory, UMN, with analytical techniques described by Shen et al. (2002). Spiked NBL-112A standard solution was measured to correct for multiplier intensity bias every day (Cheng et al., 2000). Pairs of the separated thorium and uranium fractions were sequentially analyzed by ICP-SF-MS. Both thorium and uranium mass biases were normalized to a ^{236}U/^{233}U atomic ratio of 1.010572 ± 0.00050 (Cheng et al., 2000). The accuracy of measurements of different thorium standards within error of accepted values suggests that there is no significant mass bias difference between thorium and uranium (Shen et al., 2002). Typical ^{230}Th$^+$ ion beam intensities were just tens of counts per second (cps) for corals younger than 20 years and for filtered seawater and suspended particulate material. Sources of background noise include multiplier dark noise, memory blanks, tailing of ^{238}U$^+$ and ^{232}Th$^+$, and isobaric spectral interferences. The dark noise of the multiplier (0.05 to 0.1 cps), instrumental memory blanks (typically 0.1-0.2 cps at m/z = 230, for

example), and background from the $^{238}U^+$ and $^{232}Th^+$ tails were subtracted during off-line data processing.   Spectral interferences of 0-10 cps at m/z = 229 to 237 could be generated from polyatomic organics and/or complexes, which cause a high noise/signal ratio of 20-50% for low ^{230}Th analyses.   This contamination was effectively removed by oxidation treatment with perchloric acid in chemistry, reducing the interferences to less than 0.1 cps at m/z = 230 (Shen et al., 2002). Resultant analytical uncertainty in ^{230}Th corresponded to an age uncertainty of 0.2 to 0.3 yrs.   All errors given are two standard deviations (2σ) unless otherwise noted.

2.6. Blanks

Procedural blanks were 0.02 ± 0.01 pmol ^{238}U, 0.003 ± 0.003 pmol ^{232}Th, and 0.0008 ± 0.0008 fmol ^{230}Th for coral.   The low ^{230}Th procedural blank corresponds to an age uncertainty of ± 0.1-0.2 yr.   The isotopic composition in the spike solution was carefully re-quantified.   The value of the $^{230}Th/^{229}Th$ ratio in the ^{229}Th-^{233}U-^{236}U spike solution is 0.000050 ± 0.000002.   The uncertainty corresponds to an error of $\pm$ 0.1-0.2 yr for 1 to 2-g coral samples.   The improved chemical procedure results in an overall age error of only ± 0.2-0.3 yr.   Procedural blanks of ^{238}U, ^{232}Th, and ^{230}Th for filtered seawater samples had the same as the values for corals.   For the particulate fraction, the use of an acetate cellulose filter caused a relatively high procedural ^{232}Th blank of 0.06 ± 0.03 pmol.

3. Results and discussion

3.1. Uranium and thorium data

U-Th data for all seawater and coral samples are listed in Tables A1 to A12. Uncertainties in the ^{235}U, ^{234}U, ^{232}Th, and ^{230}Th data are calculated at the 2σ level and include corrections for blanks, multiplier dark noise, abundance sensitivity, and errors associated with quantifying the isotopic composition in the spike solution. The ^{238}U level was calculated from measurement of ^{235}U and the assumed natural $^{238}U/^{235}U$ atomic ratio of 137.88 (Cowan and Adler, 1977; Steiger and Jager, 1977).

The decay constants used are 9.1577×10^{-6} yr^{-1} for ^{230}Th and 2.8263×10^{-6} yr^{-1} for ^{234}U (Cheng et al., 2000), and 1.55125×10^{-10} yr^{-1} for ^{238}U (Jaffey et al., 1971). Ages are corrected for $^{230}Th_0$ by estimating $^{230}Th/^{232}Th_0$ with 3-D ($^{232}Th/^{238}U$-$^{230}Th/^{238}U$-$^{234}U/^{238}U$) isochron techniques and with seawater data for coral sample ST0506 from Son Tra. The initial $\delta^{234}U$ value is calculated from the measured $\delta^{234}U$ value using the corrected ^{230}Th age. Intercepts, isochron ages, and isochron plot errors were calculated with an Excel macro, *Isoplot* 3.00, by K.R. Ludwig of the Berkeley Geochronology Center, California, USA (Ludwig and Titterington, 1994; Ludwig, 2003).

3.2. Seawater uranium and thorium at Nanwan and other sites

Dissolved ^{238}U concentration is 3.1 ± 0.1 ppb for Nanwan seawater (Fig. 2, Table A1) with a salinity of 33.8-34.0 (H. J. Lee, pers. comm.). Our observations support the conservative behavior of the ^{238}U concentration in this coastal ocean (Chen et al.,

1986; Robinson et al., 2004), although Taiwan experiences a high erosion rate of 3-6 mm/yr and supplies 300-500 Mt/yr of suspended sediment to the ocean (Dadson et al., 2003). Particulate ^{238}U concentration ranges from 0.6 to 2.0 ppt (Fig. 3, Table A2), representing 0.02-0.06% of the respective dissolved concentrations.

At Nanwan, the measured dissolved δ^{234}U of 147.4 ± 2.0 (Fig. 2, Table A1) is within the open-ocean interval of 146 ± 2 (Chen et al., 1986). δ^{234}U of suspended particulate matter is 34 ± 36 in Nanwan seawater (Fig. 3, Table A2). The calculated δ^{234}U of bulk (or unfiltered) seawater is 146.1-146.5, which is not significantly different from dissolved δ^{234}U values. With an analytical precision of 1-2‰, the influence of suspended particulate matter on the dissolved δ^{234}U value is not observable. The dissolved δ^{234}U is 146.6 in North Pagai and 145.0 in Son Tra (Table A1), both within the range observed in the open ocean (Chen et al., 1986).

Consecutive measurements of dissolved ^{232}Th range from 4-7 ppt at low tide to 0.7-3 ppt at high tide (Fig. 2, Table A1), 10-100 times higher than the open ocean bulk values from the central Pacific (Roy-Barman et al., 1996). The averaged particulate ^{232}Th of 3.6 ppt (Fig. 3) is 150% of the dissolved fraction. This proportion is higher, by 10-30%, than those at open-ocean sites in the Labrador Sea and Atlantic (Moran et al., 2002). This is likely the result of high thorium fluxes associated with terrestrial material input at Nanwan. The dissolved ^{230}Th/^{232}Th ratio, $4.0 ± 0.5 × 10^{-6}$, is slightly higher than that ($3.0 ± 0.7 × 10^{-6}$) of the particulate fraction (Fig. 3). It indicates that thorium is coming not only from terrestrial sources, but also from the open ocean which has high ^{230}Th/^{232}Th (Roy-Barman et al., 1996) or is derived from the ingrowth of ^{230}Th by uranium decay (e.g., Robinson et al., 2004).

There is no observable periodic trend of ^{230}Th/^{232}Th ratios for either the dissolved fraction or suspended particulate matter (Figs. 2 and 3). For neither fraction do ^{230}Th/^{232}Th ratios correlate with tide height or cold water intrusion, indicating that

nearshore water ^{230}Th/^{232}Th is not affected by either tide height or upwelled subsurface cold water. Plots of ^{230}Th/^{232}Th vs. 1/^{232}Th for both the dissolved and suspended particulate fractions support the observation of no additional distinguishable sources of thorium at Nanwan (Fig. 4).

3.3. Uranium in corals

^{238}U concentrations range from 2.4 to 2.7 ppm for modern *Porites* coral samples in Nanwan, Son Tra, and Santo (Tables A3-A6). *Porites* coral skeletal Ca content is 38.2-38.5% by mass in the South China Sea and western Pacific (Sun et al., 1999). The calculated molar coral U/Ca ratio, [U/Ca]$_{coral}$, is 1.0-1.2 $\times$ 10^{-6}. Seawater Ca concentration is 10.3 mmol/g at a salinity of 35 (Chen, 1990; Shen et al., 1996; 2005b). We calculate a molar U/Ca ratios for Nanwan water to be 1.3 $\times$ 10^{-6}. The distribution coefficient of U between *Porites* coral and seawater at the three sites is estimated as: D[U/Ca] = [U/Ca]$_{coral}$ / [U/Ca]$_{sea}$ = 0.80-0.90, consistent with the values reported by Swart and Hubbard (1982) and Shen and Dunbar (1995). For all corals in Nanwan, Son Tra, Santo, and the Sumatran islands, molar U/Ca ratios are within 30%, agreeing with previous observations (Shen and Dunbar, 1995; Min et al., 1995).

For Nanwan corals, δ^{234}U averages 147.7 $\pm$ 3.2, matching the local dissolved value of 147.6 $\pm$ 2.7 (Fig. 2). δ^{234}U in corals of Son Tra and North Pagai also matches the seawater value of 145-147 (Table A1). There is no distinguishable difference of coral δ^{234}U from dissolved value at the sites with tremendous input of terrestrial material, which supports the conclusion that marine uranium is incorporated into the coral skeleton without isotopic fractionation and that the initial δ^{234}U is a reliable parameter for estimating diagenesis (Edwards et al., 2003).

3.4. Thorium in corals

3.4.1. Distribution coefficient

Coral skeletal ^{232}Th content is a function of oceanographic setting. For *Porites* corals, it increases from 10s-100s ppt in the open ocean, such as at the Santo site, to 100-1000s ppt on the continental shelf, such as at the Son Tra and Nanwan sites, characterized by input of high-^{232}Th terrestrial material. For the intertidal corals at sites in the Sumatran islands with similar geological settings, skeletal ^{232}Th level varies from 100s-10000s ppt. Even at the same site, such as Gusong Bay, ^{232}Th content in *Porites* GSG05OLD is 3000-7800 ppt, 10 times higher than that in *Porites* GSG05YNG (Table A8). Tenfold variation in skeletal ^{232}Th levels of 1000s-10000s ppt is also observed in eight modern and Holocene corals at Bulasat, South Pagai. Unlike uranium that has a long residence time of 300-500 kyr (Dunk et al., 2002) and displays conservative behavior (Ku et al., 1977; Chen et al., 1986), thorium is non-conservative and a residence time of 0.1-0.7 yr in surface water (Broecker et al., 1973; Okubo, 1982). These features, along with the fact that different sources of thorium have various ^{230}Th/^{232}Th ratios (Cobb et al., 2003b), result in a wide range of coral ^{232}Th levels in different hydrological settings.

Thorium partitioning, similar to uranium (Cross and Cross, 1983), appears to be species dependent. The molar Th/Ca ratio is $1.0 \pm 1.2 \times 10^{-9}$ in the dissolved fraction of seawater and $2.2 \pm 2.2 \times 10^{-10}$ in *Porites* in Nanwan. A distribution coefficient, D[Th/Ca], of ~ 0.2 suggests *Porites* excludes thorium during growth (Edwards et al., 2003). Based on our measurements and previous reports (Edwards et al., 1987; Zachariasen, 1998; Zachariasen et al. 1999; Zachariasen et al., 2000; Cobb et al., 2003b; Edwards et al., 2003; Natawidjaja et al., 2004; Robinson et al.,

2004), *Acropora* corals have high D[Th/Ca] values $\geqq$ 1. Thorium is effectively excluded by *Goniastrea* with an estimated D[Th/Ca] of 0.02 or less.

3.4.2. Dissolved phase of seawater and detrital materials

Three isochrons with ages of 4.1 ± 1.2, 5.1 ± 3.0 and 15.56 ± 0.56 yrs, for two Nanwan coral slabs, NW0310 and NW0402, are shown in Fig. 5a. All regression y-intercept values, with uncertainties of 1-2 × 10^{-6}, overlap with each other at 5 × 10^{-6} (Fig. 5a). An excellent fit to straight lines illustrates that there is no significant difference of the isotopic composition of associated thorium in the skeletal lattice between the three growth bands of the two coral heads in the same hydrographic environment. All intercepts are consistent with a value of 4.0 ± 0.5 × 10^{-6} in the dissolved fraction of seawater (Fig. 2) and higher than the value of 3.0 ± 0.7 × 10^{-6} in the suspended particulate matter (Fig. 3). A $^{230}Th/^{232}Th_0$ value of 4.7 ± 1.0 × 10^{-6} for growth banding of a modern coral, NP00A1 (Fig. 5f), from North Pagai captures the dissolved values of 4.0-4.3 × 10^{-6} at that site. At the Son Tra site, using a dissolved $^{230}Th/^{232}Th$ value of 3.20 ± 0.32 × 10^{-6} as the initial thorium isotopic composition, the ^{230}Th ages of 3 bands of the Son Tra *Porites*, ST0506, match the absolute ages (Table A5).

Robinson et al. (2004) showed the seawater $^{230}Th/^{232}Th$ value should be used for $^{230}Th_0$ correction with coral and bulk seawater data. The similarity between the dissolved $^{230}Th/^{232}Th$ values and the respective initial ratios in corals at Nanwan, North Pagai, and Son Tra, further supports that the idea that $^{230}Th_0$ is mainly from the dissolved fraction of seawater. The data from all three sites show that the ^{230}Th age should be corrected for $^{230}Th_0$ using the dissolved $^{230}Th/^{232}Th$ ratio. Precise and accurate correction for $^{230}Th_0$ content in coral can be achieved by understanding the spatial and temporal variability of $^{230}Th/^{232}Th_0$; this can be accomplished with

isochron techniques as described in sections 3.5 and 3.6.

Three discordant ages were observed from isochrons (Fig. 5) and the 1:1 line of the ^{230}Th age vs. growth band age plot (Fig. 6). The ^{230}Th age of one subsample, NW0310-5#3 (Table A4), is 3.8 ± 1.0 yrs older than the other three coeval subsamples. Two ^{230}Th/^{232}Th-^{234}U/^{232}Th data points, 92MC-1#4 (Table A6) and NP00A1-1#5 (Table A10), do not lie on isochrons (Figs. 5b and 5f). The ^{230}Th age of 92MC-1#4 is 5.0 ± 1.4 yrs older than the isochron age of 14.0 ± 1.1 yrs, constructed with the other 3 coeval subsamples. The ^{230}Th age of NP00A1-1#5 is 23 ± 6 yrs younger than the isochron age of 8.1 ± 1.9 yrs, established with 4 coeval subsamples (Table A10). The discordant ages are proposed to be caused by detrital materials with different thorium concentrations and ^{230}Th/^{232}Th ratios that were incorporated into the growing lattice during crystallization. The positive age bias for NW0310-5#3 and 92MC-1#4 could be due to carbonate detritus with low thorium concentration and a high ^{230}Th/^{232}Th ratio. Terrestrial particulates with a high thorium level and a low ^{230}Th/^{232}Th ratio of 1-2 × 10^{-6} could result in the negative age bias of NP00A1-1#5.

3.5. Variability of ^{230}Th/^{232}Th$_0$ in coral skeletons

3.5.1. Subtidal corals at continental shelf sites: Nanwan and Son Tra

3.5.1.1. Nanwan. The ^{230}Th/^{232}Th$_0$ values derived from three isochrons of two modern Nanwan corals, 5.2 ± 1.1 × 10^{-6} and 4.86 ± 0.27 × 10^{-6} for NW0310 and 5.1 ± 1.9 × 10^{-6} for NW0402, collected 60 m apart, are consistent with each other (Fig. 5a). Plots of ^{230}Th age, calculated with the ^{230}Th/^{232}Th$_0$ value of 4.86 ± 0.27 × 10^{-6} from NW0310-6, vs. banding age for 3 subsamples of NW940617 and 5 layers of NW0310, including bulk and three crushed fractions of < 840 μm, 840-1410 μm, and

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1410-2380 μm (Table A4), are shown in Fig. 6. All points, except for NW0310-5#3 with high ^{230}Th/^{232}Th-detritus, plot within error of a 1:1 line for both bulk subsamples and crushed fractions, indicating that the ^{230}Th/^{232}Th$_0$ ratios on different growth layers vary less than $\pm$ 0.27 $\times$ 10^{-6}. A single ^{230}Th/^{232}Th$_0$ value can be used to determine different ^{230}Th ages on annual to decadal timescales in Nanwan. The results and measurements of seawater thorium isotopic composition at Nanwan (Figs. 2 and 3) indicate that inter-annual variability of the ^{230}Th/^{232}Th ratio in the dissolved fraction is not resolvable, even though there is a large daily intrusion of cold upwelled subsurface water mass at the Nanwan site (Lee et al., 1997; Shen et al., 2005b).

3.5.1.2. Son Tra. All 6 subsamples of 3 layers of the modern Son Tra coral lie on a 1:1 line in Fig. 6 using a seawater ^{230}Th/^{232}Th value of 3.20 $\pm$ 0.32 $\times$ 10^{-6} as the initial value, indicating no significant annual change of the initial thorium isotopic composition at this site. There is a distinguishable difference in ^{230}Th/^{232}Th$_0$ values of 1.66 $\pm$ 0.42 $\times$ 10^{-6} between Son Tra and Nanwan (Fig. 7), suggesting that a terrestrial source with low thorium isotopic composition and high ^{232}Th levels of 1000s ppt is more dominant at Son Tra.

Located on the continental shelf with a large influx of inland material, the Nanwan and Son Tra sites in the western Pacific yield isochron-inferred ^{230}Th/^{232}Th$_0$ ratios of 3-5 $\times$ 10^{-6} (Fig. 5), consistent with an average crustal value (Richards and Dorale, 2003). Results of seawater analyses and isochrons, and the consistency of ^{230}Th and absolute ages, show that the variability of ^{230}Th/^{232}Th ratios in the dissolved fraction of seawater and initial values in corals is $\pm$ 0.25-0.35 $\times$ 10^{-6}, which is within 10% of the mean value at the two local sites. These small temporal and spatial variations suggest that the crust-derived mean value of 4-5 $\times$ 10^{-6} with an arbitrary uncertainty of 50% or 100% is practical for the continental shelf region in the western Pacific. The observation of resolvable differences in the ^{230}Th/^{232}Th$_0$ values between Nanwan

and Son Tra indicates that a site-specific ^{230}Th/^{232}Th$_0$ ratio is important for high precision and accurate ^{230}Th dating.

3.5.2. Subtidal corals at open ocean site: Santo

Data of ^{230}Th age vs. growth band age for subsamples of the Santo 92MC coral, except for subsample 1#4, and subsamples 2#1 through 2#4 (Table A6), reside on a 1:1 line using the isochron-inferred ^{230}Th/^{232}Th$_0$ value of 5.62 ± 2.05 × 10^{-6} from the coeval subsamples on layer 92MC-1 (Fig. 5b).   This initial value at this open-ocean site, with minor terrestrial influence, is higher than 3.2 × 10^{-6} measured at the continental shelf site of Son Tra (Fig. 5).   The discordant age of subsample 1#4 was discussed earlier in section 3.4.2.   The isochron plot of layer 92MC-2 shows a much higher ^{230}Th/^{232}Th$_0$ ratio of 23.8 ± 4.7 × 10^{-6} and the isochron age of 29.2 ± 1.3 yrs matches the absolute date (Table A6).   If we use the initial value inferred from the layer 92MC-1, however, the ^{230}Th ages of subsamples of 2#1-2#4 are 2.9-8.7 years older than the absolute age of 30.0 ± 0.5 yrs.

The high ^{230}Th/^{232}Th$_0$ value of layer 92MC-2 in AD 1974.6 ± 0.5 (growth band age) could be attributed to substantial upwelling of cold water in the Eastern Pacific during a predominant La Niña episode between AD 1973 and 1976.   In terms of the magnitude of the Southern Oscillation Index, the three La Niña years, AD 1974, 1975, and 1976, are historically ranked as the 1st, 15th, and 4th, respectively during the interval from 1951 to 1996 (Clark et al., 2001).   A monthly sea surface temperature anomaly (SSTA) of -2 ^{o}C over the Niño-3.4 region (5°S-5°N, 170-120°W; http://www.cpc.noaa.gov/data/indices/) in the central equatorial Pacific was observed in both La Niña episodes of AD 1973/74 and 1975/76.   During a similar SSTA during the 1998/99 La Niña event, shoaling of the thermocline in the eastern

equatorial Pacific was induced by a strong westward near-surface (0-15 m) current of 1 m/s (Grodsky and Carton, 2001; Bonjean and Lagerloef, 2002). A high seawater ^{230}Th/^{232}Th value of 20-200 $\times$ 10^{-6} was measured at a depth of 25 m at the Aloha Station in the central Pacific (22°45'N, 158°00'W) in September 1994 (normal El Niño/Southern Oscillation (ENSO) condition; Roy-Barman et al., 1996). Notably, in the 1944 La Niña event, a high ^{230}Th/^{232}Th$_0$ ratio of 22-25 $\times$ 10^{-6}, calculated with U-Th data from the Modern-2 coral in Table 2 of Cobb et al. (2003b), was also observed at Palmyra Island, in the central tropical Pacific (5°51'N, 162°8'W). The high ^{230}Th/^{232}Th$_0$ value of 24 $\times$ 10^{-6} found in layer 92MC-2 of the Santo coral could likely be attributed to cold upwelled water delivered by the persistent westward current. An alternate possibility might be local changes in current circulation and/or wind strength and direction at the Santo site that were tied to ENSO.

In Santo coral 92MC, an initial isotopic ratio of 5.6 ± 2.1 × 10^{-6} is observed in 4 of 5 bands and a high value of 23.8 ± 4.7 × 10^{-6} observed in AD 1974-1975 during a strong La Niña episode. The fluctuation of ^{230}Th/^{232}Th$_0$ values at Santo, an open ocean site in the western tropical Pacific, could be related to mixing of water masses. Also taking into consideration the case at Palmyra Island in the central tropical Pacific (Cobb et al., 2003b), isochron techniques are suggested for determining accurate and precise ^{230}Th dates. High-^{230}Th/^{232}Th thorium, delivered by the upwelled water, can be taken up by the coral skeleton. The magnitude of ^{230}Th/^{232}Th$_0$ variation at these regions should therefore be understood in advance for high precision coral ^{230}Th dating.

3.5.3. Intertidal corals at sites in the Sumatran islands

3.5.3.1. Gusong Bay and Lewak of Simeulue. At the Gusong Bay site of

southwestern Simeulue, there is no significant difference between four isochron-inferred $^{230}Th/^{232}Th_0$ values, $7.35 \pm 0.65 \times 10^{-6}$ and $7.24 \pm 0.70 \times 10^{-6}$ from modern coral GSG05YNG, and $6.59 \pm 0.97 \times 10^{-6}$ and $6.97 \pm 2.86 \times 10^{-6}$ from fossil GSG05OLD (Fig. 5d). Using an initial value of $7.35 \pm 0.65 \times 10^{-6}$, six additional ^{230}Th age vs. band counting age points lie on a 1:1 line (Fig. 6; Table A8). Gusong Bay data show that the $^{230}Th/^{232}Th_0$ ratio remains steady at this site. A similar case of a steady but lower $^{230}Th/^{232}Th_0$ value of $3.01 \pm 0.47 \times 10^{-6}$, inferred from a LWK05 coral isochron (Fig. 5c; Table A7) and supported by the concordance between the banding ages and the ^{230}Th ages calculated using the initial value (Fig. 6), is observed at the Lewak site in northern Simeulue, 80 km from the Gusong Bay site. The discrepancy between the initial values at the 2 sites, on the same island, clearly shows the $^{230}Th/^{232}Th_0$ ratio is dependent on local hydrology.

3.5.3.2. *Lago.* A $^{230}Th/^{232}Th_0$ ratio of $9.4 \pm 1.2 \times 10^{-6}$ is observed for band LG00A1-1 of coral LG00A1, from the northeastern side of Lago island (Fig. 5e). An age of AD 1935.1 ± 0.9, determined with this initial value, is identical to a band age of AD 1935.0 ± 0.5 on the slab (Fig. 5e). Calculated with the same $^{230}Th/^{232}Th_0$ value, a date of AD 1942.1 ± 1.6 is calculated, identical to the absolute age (AD 1942.5 ± 0.5), for horizon LG99A1-3 of a contiguous coral slab, LG99A1, from the same coral head (Table A9). This indicates that the high $^{230}Th/^{232}Th_0$ value did not change after the earthquake in December 1935 (Mw = 7.7; Rivera et al., 2002). The carbonate-enriched sandy substrate could supply ^{230}Th and a high $^{230}Th/^{232}Th$ ratio source to coral skeletons (Robinson et al., 2004). A different initial value of $7.08 \pm 0.86 \times 10^{-6}$ is found using an isochron from an AD 1996 layer, LG99A1-1. This low initial value could likely be attributed to either a different thorium isotopic composition in seawater or a lower proportion of ^{230}Th to the coral skeleton.

3.5.3.3. *North Pagai.* The isochron-derived $^{230}Th/^{232}Th_0$ ratios for one modern

coral, NP00A1, and one 475-year-old fossil, NP00A2, sampled near Simanganya village along the northeastern coast of North Pagai are indistinguishable (Fig. 5f). The value of $4.7 \pm 1.0 \times 10^{-6}$ is identical to that of the dissolved fraction of ambient seawater, $4.0\text{-}4.3 \times 10^{-6}$, supporting the finding that the dissolved seawater thorium is the main source of thorium to the coral skeleton. Dissolved ^{232}Th concentrations at the North Pagai site range from 9.5-13.8 ppt, which is even higher than those at Nanwan. Coupled with low ^{230}Th/^{232}Th ratios of $4.0\text{-}4.3 \times 10^{-6}$, this indicates that the site in North Pagai also experiences a high flux of terrestrial matter.

3.5.3.4. Bulasat and Saumang of South Pagai. The Bulasat and Saumang sites have similar hydrological settings and are 10 km apart on the southwestern coast of South Pagai. The ^{230}Th/^{232}Th$_0$ ratios of two modern coral slabs, BLS02A5 and SMG02A1, are $4.9 \pm 1.4 \times 10^{-6}$ and $5.02 \pm 0.65 \times 10^{-6}$, respectively (Figs. 5g and 5h). The identical ^{230}Th/^{232}Th$_0$ values of the two slabs suggest that there is no spatial variation of thorium isotopic composition at the sites in South Pagai. Temporal variation of ^{230}Th/^{232}Th$_0$ values at the Bulasat site is characterized with 8 isochrons going back 2.5 thousand years (Figs. 5g and 7a). Despite 2-sigma uncertainties of $1.4\text{-}3.8 \times 10^{-6}$, the graphs show that the means of the initial values ranged from $7\text{-}8 \times 10^{-6}$ at 2.5 thousand years ago (ka) to 3.6×10^{-6} at 1.5 ka, with intermediate values of $4.8\text{-}4.9 \times 10^{-6}$ during the past 0.7 ka. An endmember plot of ^{230}Th/^{232}Th$_0$ vs. $1/^{232}$Th displays a mixing trend with an intercept of $4.4 \pm 2.3 \times 10^{-6}$ at the 95% confidence level (Fig. 7b), which agrees with the crust-derived mean value of $4.5\text{-}4.7 \times 10^{-6}$ (Richards and Dorale, 2003). Terrestrial sources with low ^{230}Th/^{232}Th dominated in the 4 coral heads since 1.5 ka. High ^{230}Th/^{232}Th$_0$ ratios recorded in the other 4 fossil corals, older than 1.5 ka, can be attributed to different sources such as open ocean water masses and carbonate-enriched substrate with high ^{230}Th (Robinson et al., 2004).

3.5.3.5. $^{230}Th/^{232}Th_0$ ***variation between sites.*** The natural dynamics of $^{230}Th/^{232}Th_0$ values in the different intertidal zones at sites of the Sumatran islands are more complicated and governed by variability of 4 sources: (1) terrestrial influx, (2) in-situ ingrowth ^{230}Th, (3) open-ocean surface seawater, and (4) upwelled cold seawater. Terrestrial sources appear to be dominant at the Lewak and North Pagai sites with low $^{230}Th/^{232}Th$ values of 3-4 $\times$ 10^{-6}. The Lago site, away from sources of significant terrestrial input and with a carbonate sandy substrate, shows high $^{230}Th/^{232}Th_0$ values of 7-9 $\times$ 10^{-6}. The regional seawater $^{230}Th/^{232}Th$ value is affected by the upwelling of cold seawater, which has been observed in the eastern Indian Ocean. For example, three strong positive Indian Ocean Dipole events, in 1877, 1994 and 1997, caused a 4 ^{o}C drop of sea surface temperature in the Mentawai reefs (Abram et al., 2003). The offshore islands have also experienced frequent rupture of active faults (Zachariasen et al., 1999; Sieh et al., 1999; Natawidjaja et al., 2004; Briggs et al., 2006). The proportion of the different thorium sources could plausibly be altered by uplift or subsidence associated with seismic displacements. A wide isochron-inferred initial range of 3.0-9.4 $\times$ 10^{-6} is shown in Fig. 5. The observed spatiotemporal variation of the means of the isochron-inferred $^{230}Th/^{232}Th_0$ ratios is 6.2 $\pm$ 3.2 $\times$ 10^{-6} at the sites, which sprawl across 800 km of the Sumatran islands (Figs. 5c-5h). The range, consistent with the previous value of 6.5 $\pm$ 6.5 $\times$ 10^{-6} published by Zachariasen et al. (1999), can be applied to ^{230}Th dating of intertidal corals in the Sumatran islands. However, high precision and accurate ^{230}Th dates will require well-constrained $^{230}Th/^{232}Th_0$ values specific to each site.

3.6. Limitations of high precision coral ^{230}Th dating

3.6.1. Prerequisites

High precision coral ^{230}Th dating with a precision of ± 1 yr requires an appropriate quality control with a careful sampling procedures, cross-contamination-free sample storage, and proper subsampling methods. The level and uncertainty of ^{230}Th blank in the chemical procedure and instrumental analysis should be effectively reduced. In this study, the procedural ^{230}Th blank of 0.0008 ± 0.0008 fmol ^{230}Th corresponds to an uncertainty of only ± 0.1-0.2 yr for 1-g coral samples.

3.6.2. Usage of appropriate ^{230}Th/^{232}Th$_0$ values

A bulk Earth crustal Th/U atomic ratio of 3.6-3.8 (Taylor and McLennan, 1985; 1995) and an assumed secular equilibrium between ^{230}Th and ^{238}U in the terrestrial upper or bulk continental crust is generally used to determine a ^{230}Th/^{232}Th value of 4-5 × 10^{-6} (Richards and Dorale, 2003); this, coupled with an arbitrary uncertainty of 50% or 100%, is often considered as the initial value for the age calculation in the ^{230}Th dating equation (Eq. 1). This study of modern and fossil coral and seawater samples indicates that the dissolved fraction of seawater is the primary thorium source for the coral skeleton. Variability of seawater ^{230}Th/^{232}Th values may result from mixing of sources with different thorium isotopic compositions. The use of the continental crust-derived values is valid where the dominant source of thorium is terrestrial. However, a positive age bias can be generated if the crustal value is used for sites with high ^{230}Th/^{232}Th sources. Isochron techniques can offer an accurate ^{230}Th/^{232}Th$_0$ value and a date with precision as good as ± 1 yr for corals younger than

100 years (Fig. 5).   A high precision date can also be achievable using site-specific ^{230}Th/^{232}Th$_0$ ratios (Fig. 6, Tables A3-A12).

3.6.3. Duplicate measurements

Three discordant ages in 141 U-Th points indicate additional ^{230}Th and ^{232}Th sources, presumably high-^{230}Th/^{232}Th carbonate sands and low-^{230}Th/^{232}Th terrestrial materials, which cause occasional biases of 4-23 years from the true age (Fig. 6; Tables A4, A6 and A10).   Coeval subsamples with different dating results indicate that heterogeneous coprecipitation places limits on high precision ^{230}Th dating.   In our study, these discordant ages were relatively rare (2.1% of the 141 total subsamples).   Duplicate measurement of coeval subsamples is suggested to verify the accuracy of high precision dates if ^{230}Th/^{232}Th$_0$ can be well estimated.

This issue is not critical for Quaternary samples, for which a ^{230}Th dating precision of $\pm$ 30-50 yrs is satisfactory.   For young corals, the occasional problem of detrital material with ^{230}Th/^{232}Th ratios significantly different from the dissolved value in seawater can be resolved by duplicate measurements of ^{230}Th ages at 2-3 coeval loci on a single growth band as a concordance test to rule out the influence of detrital material contributing an anomalous ^{230}Th/^{232}Th value.

4. Conclusions

Measurements of U-Th isotopic compositions in suspended particulate material and the dissolved fraction of seawater and in modern and fossil corals from sites in the western Pacific Ocean, South China Sea, and eastern Indian Ocean were performed to

understand the natural variation of $^{230}Th/^{232}Th_0$ in shallow seawater corals and its effects on coral ^{230}Th dating.   To approach this objective, sample preparation procedures and chemical procedures have been refined, resulting in an equivalent age uncertainty of only $\pm$ 0.2-0.3 yr.

Coral isochron-derived $^{230}Th/^{232}Th_0$ ratios, comparison of ^{230}Th ages and absolute ages, and thorium analyses in seawater samples demonstrate that coral skeletal thorium originates mainly from the dissolved fraction of seawater.   The $^{230}Th/^{232}Th_0$ value is strongly influenced by local hydrological setting. $^{230}Th/^{232}Th_0$ values are low (3-5 $\times$ 10^{-6}) at Son Tra and Nanwan, two continental shelf sites with intense terrestrial material input.   The variation of initial values at each site is ~ 10%.   The crustal U-Th composition-inferred $^{230}Th/^{232}Th$ values can be used for age calculation. At the open-ocean site of Santo in the western Pacific, a value of 5.6 $\times$ 10^{-6} is observed at 4 horizons, and one high value of 24 $\times$ 10^{-6} is documented during a La Niña event in AD 1973-1976.   The initial values recorded in the intertidal corals at the sites in the Sumatran islands vary significantly from 3.0-9.4 $\times$ 10^{-6}. Accordingly, isochron techniques should be applied for high precision dating at these sites with a variety of $^{230}Th/^{232}Th_0$ ratios.   Detrital material with anomalously low or high $^{230}Th/^{232}Th$ ratios causes biases ranging from -23 to +4 years in this study. Duplicate measurement of coeval subsamples is recommended to verify the age accuracy.

Band-counted and tracer-tuned chronologies are usually characterized by compounded age errors of at least $\pm$ 2-3 yrs by AD 1800-1900.   Using site-specific $^{230}Th/^{232}Th_0$ values or isochron techniques, our study demonstrates that a coral ^{230}Th dating method with a precision as high as $\pm$ 1 yr is achievable for corals with ages less than 100 years.   The ability to obtain high precision and accuracy ages on young coral will be useful in diverse fields within the broad area of global change, including

oceanography, tectonic evolution, anthropogenic pollution history, paleoclimate and paleo-environment.   This ^{230}Th dating methodology can also be applied to different carbonate samples, such as speleothem and tufa.

Acknowledgements

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Captions

Fig. 1. Map of sample collection sites. Modern and fossil corals were collected from Nanwan, Taiwan, Son Tra Island, Vietnam, southern Espiritu Santo Island, Vanuatu, and the Sumatran islands. Surface seawater samples were collected from Nanwan, Son Tra, and North Pagai of the Sumatran islands.

Fig. 2. Time series of (**a**) tide height, (**b**) temperature, (**c**) ^{238}U concentration, (**d**) $\delta^{234}U$, (**e**) ^{232}Th concentration, and (**f**) $^{230}Th/^{232}Th$ ratio for filtered Nanwan seawater collected from July 31, 06:00 AM to August 1, 10:00 PM, 2004. Means and 2-sigma errors of U-Th data are given.

Fig. 3. Time series of (**a**) tide height, (**b**) temperature, (**c**) ^{238}U concentration, (**d**) $\delta^{234}U$, (**e**) ^{232}Th concentration, and (**f**) $^{230}Th/^{232}Th$ ratio for the suspended particulate matter of Nanwan seawater collected from October 28, 7:00 AM to October 30, 1:00 AM, 2004. Average values and 2-sigma errors of U-Th data are listed by record.

Fig. 4. A plot of $^{230}Th/^{232}Th$ vs. $1/^{232}Th$ for the dissolved fraction (solid cubes) and suspended particulate matter (gray cubes) of Nanwan seawater, dissolved fraction of Son Tra seawater (hollow circles), and dissolved fraction of intertidal seawater of North Pagai (hollow triangles).

Fig. 5. Isochron plots of $^{230}Th/^{232}Th$ vs. $^{234}U/^{232}Th$ (atomic ratios) for 23 coeval sets of subsamples from (**a**) three growth bands of two Nanwan coral slabs, NW0310 and NW0402, (**b**) two growth bands of the Santo 92MC coral, and (**c-h**) 18 horizons of corals from the Sumatran islands. Isochron-inferred $^{230}Th/^{232}Th_0$ ratios (*y*-intercepts with 2σ errors) are enlarged in insets.

Fig. 6. ^{230}Th age vs. growth band age plots of over 50 data points for coral subsamples of Nanwan, Son Tra, Santo, and the Sumatran islands. Eight of the ^{230}Th

ages are inferred from isochron techniques and the other ages are calculated with different ^{230}Th/^{232}Th$_0$ ratios (see text and Appendices).   Note that subsample NP00A1-1#5, which yielded a ^{230}Th age 23 ± 6 yrs younger than the isochron age of 8.1 ± 1.9 yrs, is not shown on this plot.   Data for subsamples that may have acquired high ^{230}Th in the storage room or during the subsampling process are given in black.

Fig. 7.   (**a**) Temporal variation of isochron-inferred ^{230}Th/^{232}Th$_0$ ratios for eight corals collected from Bulasat of the Sumatran islands over 2.5 ka.   (**b**) An endmember plot of ^{230}Th/^{232}Th$_0$ vs. 1/^{232}Th shows a mixing trend (gray line) with an intercept of $4.4 \pm 2.3 \times 10^{-6}$ at the 95% confidence (dashed lines).

Appendix captions

Fig. A1. Map of collection sites of corals (stars) and seawater samples (square) at Nanwan, Taiwan.

Fig. A2. The collection site of a living *Porites* coral (star) and seawater (square) at Son Tra Island, Vietnam.

Fig. A3. The coral coring site of sample 92MC (star) in southern Espiritu Santo Island of Vanuatu.

Fig. A4. The collection sites of modern and fossil corals (stars) and seawater (square) in the Sumatran islands.

Table A1. U-Th data for dissolved fractions of seawater samples from Nanwan, North Pagai and Son Tra.

Table A2. U-Th data in the suspended particulate matter of 5-L Nanwan seawater samples.

Table A3. U-Th data and ^{230}Th ages for subsamples of Nanwan NW030525, NW940101, and NW940617 corals.

Table A4. U-Th data and ^{230}Th ages for subsamples of Nanwan NW0310 and NW0402 corals.

Table A5. U-Th data and ^{230}Th ages for subsamples of the ST0506 coral slab from Son Tra Island, Vietnam.

Table A6. U-Th data and ^{230}Th ages for subsamples of the Santo 92MC coral.

Table A7. U-Th data and ^{230}Th ages for subsamples of the LWK05 coral collected from Lewak, northern Simeulue, Sumatran islands.

Table A8.   U-Th data and ^{230}Th ages for subsamples of coral slabs GSG05YNG and GSG05OLD, collected from Gusong Bay, southwestern Simeulue, Sumatran islands.

Table A9.   U-Th data and ^{230}Th ages for subsamples of two coral slabs, LG00A1 and LG99A1, from Lago of the Sumatran islands.

Table A10.   U-Th data and ^{230}Th ages for subsamples of two coral slabs, NP00A1 and NP00A2, from the east coast of North Pagai, Sumatran islands.

Table A11.   U-Th data and ^{230}Th ages for subsamples of 8 coral slabs from Bulasat of South Pagai, Sumatran islands.

Table A12.   U-Th data and ^{230}Th ages for subsamples of 3 coral slabs from Saumang of South Pagai, Sumatran islands.