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A HIGH-RESOLUTION RADIOCARBON CALIBRATION BETWEEN 11,700 AND 12,400 CALENDAR YEARS BP DERIVED FROM ^{230}TH AGES OF CORALS FROM ESPIRITU SANTO ISLAND, VANUATU

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ABSTRACT. This paper presents radiocarbon results from a single *Diploastrea heliopora* coral from Vanuatu that lived during the Younger Dryas climatic episode, between *ca.* 11,700 and 12,400 calendar yr BP. The specimen has been independently dated with multiple ^{230}Th measurements to permit calibration of the ^{14}C time scale. Growth bands in the coral were used to identify individual years of growth. ^{14}C measurements were made on each year. These values were averaged to achieve decadal resolution for the ^{14}C calibration. The relative uncertainty of the decadal ^{14}C data was below 1% (2σ). The data are in good agreement with the existing dendrochronology and allow for high-resolution calibration for most years. Variations in the fine structure of the ^{14}C time series preserved in this specimen demonstrate sporadic rapid increases in the Δ^{14}C content of the surface ocean and atmosphere. Certain sharp rises in Δ^{14}C are coincident with gaps in coral growth evidenced by several hiatuses. These may be related to rapid climatic changes that occurred during the Younger Dryas. This is the first coral calibration with decadal resolution and the only such data set to extend beyond the dendrochronology-based ^{14}C calibration.

INTRODUCTION

The ^{14}C calibration curve commonly used today (Stuiver, Long and Kra 1993) evolved over the past 30 years and represents the efforts of a number of laboratories. The 1993 calibration covers the last 11.4 ka and is based on thousands of ^{14}C measurements of tree rings. The dendrocalibration has recently been extended to *ca.* 11.9 ka (Kromer and Spurk 1998), but beyond this point the tree-ring record is uncalibrated. The existence of relatively old floating Tasmanian tree-ring series should eventually provide a means for extending the existing dendrocalibration in the future (Barbetti *et al.* 1992; Tuniz *et al.* 1997).

Researchers in the field have sought to extend the dendrochronological limit using several alternative methods, including calibration based on counting varved sediments (Wohlfarth 1996; Hughen *et al.* 1998) and calibration using carbonates dated with ^{230}Th, such as corals (Bard *et al.* 1990, 1993; Edwards *et al.* 1993) and speleothems (Vogel and Kronfeld 1997; Goslar *et al.* 1997; Richards *et al.* 1997). Although these studies have produced useful information about past variations in the ^{14}C content of the atmosphere, none of them has approached the resolution and precision of dendrochronology. The purpose of this paper is to extract a high-resolution (decadal) ^{14}C calibration record from coral with precision comparable to the dendrocalibration. This is possible because certain corals are annually banded (see, *e.g.*, Knutson, Buddemeier and Smith 1972) and can be dated very accurately using the ^{230}Th age dating technique (Edwards, Chen and Wasserburg 1987). This is the first study to adopt such a strategy for the purpose of calibration.

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METHODS

Site Selection and AMS ^{14}C Sample Preparation

The samples analyzed in this study were collected from a core drilled on the shore of a tectonically active coast on Espiritu Santo Island, Vanuatu (Fig. 1). The Late Quaternary tectonics and environmental history of this site were described by Taylor *et al.* (1987) and Cabioch *et al.* (1997). The Santo Island site has uplifted at a rapid and variable rate (up to *ca.* 5mm yr^{-1}) over the past 20 ka, which allowed for repeated coral colonization of the emerging reef. A portion of one of our cores intersected a single *Diploastrea heliopora* coral that lived between *ca.* 11.7 and 12.4 ka BP according to our ^{230}Th measurements (see below).

Four features of this specimen of *Diploastrea heliopora* are significant to this study: 1) this species produces annual growth bands that are visible in X-radiographs; 2) our core penetrated the coral nearly perpendicular to the plane of growth, permitting annual subsampling along its entire length; 3) the coral lived for *ca.* 400 yr over a 720-yr interval; and 4) the specimen lived during the Younger Dryas climatic episode, which is known to have been a time of rapid change in atmospheric $\Delta^{14}C$ (Edwards *et al.* 1993). The record is continuous over four intervals punctuated by three hiatuses. Individual years were identified from pairs of light and dark growth bands as seen in the X-radiographs of the sample. In order to reduce the variability introduced by subannual ^{14}C variations in the coral skeleton (Brown *et al.* 1993), combined light and dark couplets representing full years were sampled and analyzed together. To avoid contamination with modern carbon, the core was analyzed with the X-ray powder diffraction technique to check for calcite recrystallization. All of the samples analyzed in this study were pretreated using the selective dissolution technique described by Burr *et al.* (1992).

Definition of Terms and Analytical Procedures

^{14}C Dating

To calculate ^{14}C ages, $^{14}C/^{13}C$ ratios in the samples and standards were compared to determine the fraction of modern carbon (F) values, defined as

$$F \equiv (^{14}C/^{13}C)_S / (^{14}C/^{13}C)_{STD} \tag{1}$$

where $(^{14}C/^{13}C)_S$ is the measured ratio in the sample, normalized to $\delta^{13}C = -25‰$, and $(^{14}C/^{13}C)_{STD}$ is the calculated modern standard ratio (1950 AD), determined from measurements of NBS oxalic acid standards, also normalized to −25‰ (Donahue, Jull and Toolin 1990). The age of the sample is computed with the equation

$$^{14}C\ \text{age} = -\tau \ln F \tag{2}$$

where τ is the Libby mean life (5568/ln 2 = 8033 yr).

In order to compare ^{14}C dates with atmospheric ^{14}C values, the ^{14}C ages were reservoir-corrected using the relationship

$$^{14}C\ \text{age}_{RC} = {}^{14}C\ \text{age} - RC \tag{3}$$

where ^{14}C age_{RC} is the reservoir-corrected age and RC is the reservoir correction in years. To estimate the reservoir correction at the site, we measured 35 prebomb samples of known age and calculated the average. The reservoir age calculated in this manner is 494 ± 10 yr. We assume in our calculations that the reservoir correction is constant. This is consistent with the dendrocalibration for the past 11.9 ka, but it should be emphasized that the reservoir age is affected by the source of surface ocean water and could vary with changing paleo-ocean circulation patterns.

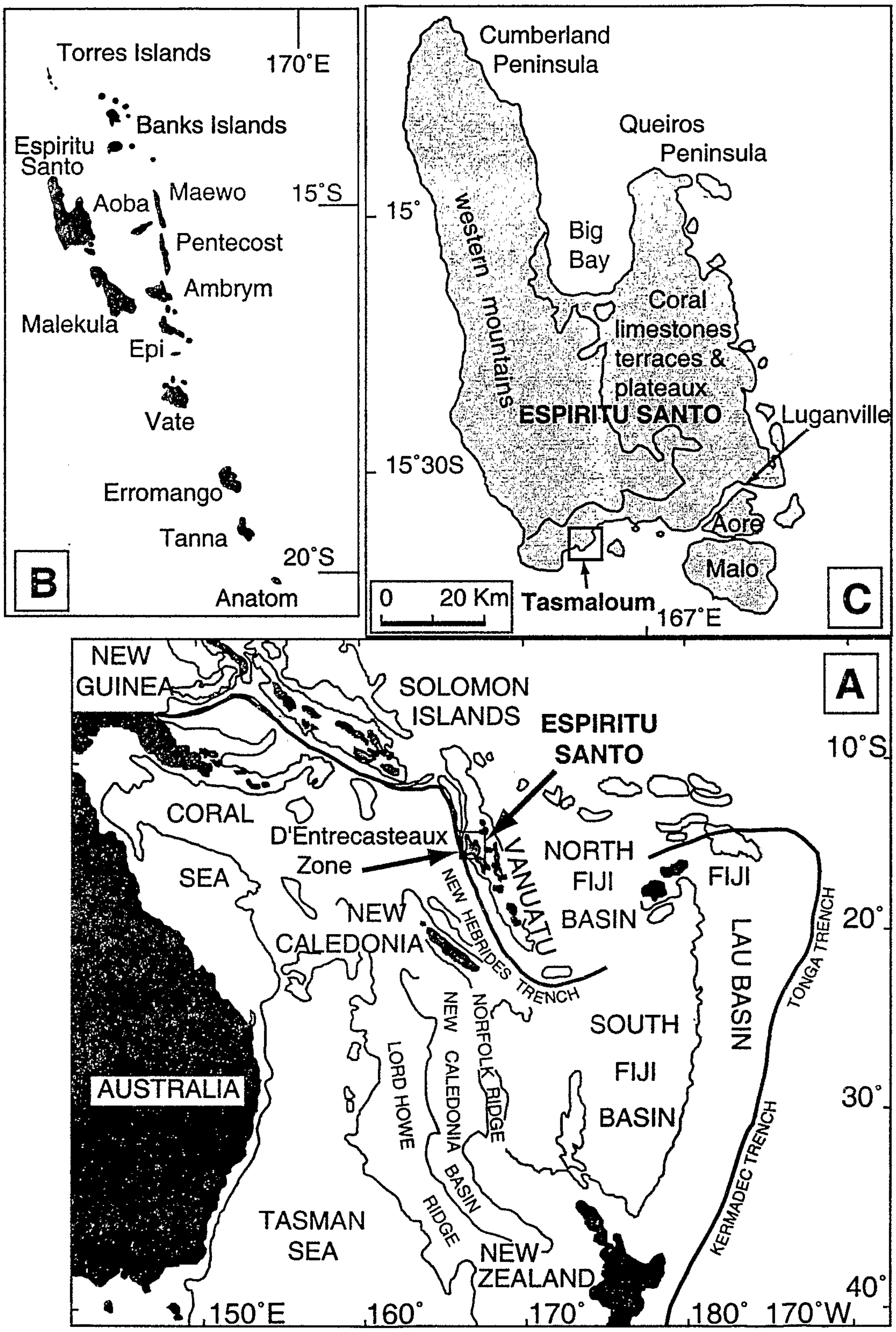

Fig. 1. Map showing the location of Vanuatu. A. Regional view; B. major islands of Vanuatu; C. Espiritu Santo Island with Tasmaloum peninsula (drill site).

To calculate reservoir-corrected fraction of modern values (F_{RC}), we define the relationship

$$^{14}\mathrm{C\ age}_{\mathrm{RC}} \equiv -\tau \ln F_{\mathrm{RC}} \ . \quad (4)$$

Combining equations 2, 3, and 4 yields the expression

$$F_{\mathrm{RC}} = F\, e^{\mathrm{RC}/\tau} \ . \quad (5)$$

The uncertainty in F_{RC} depends on the uncertainty in F and on the uncertainty in RC. Propagating these two sources of error yields the expression

$$\sigma_{F_{\mathrm{RC}}} = \left\{ \left(e^{\mathrm{RC}/\tau}\right)^2 (\sigma_F)^2 + \left[\left(\frac{F}{\tau}\right)\left(e^{\mathrm{RC}/\tau}\right)\right]^2 (\sigma_{\mathrm{RC}})^2 \right\}^{1/2} \quad (6)$$

where the σ's represent the uncertainties in F_{RC}, F, and RC.

Δ^{14}C values were computed from F_{RC} values. Δ^{14}C is a relative measure of the ^{14}C/^{12}C (or ^{14}C/^{13}C) content of the atmosphere, as compared with the assumed value for 1950. Positive values indicate an excess relative to 1950 and negative values indicate a relative ^{14}C deficit. Δ^{14}C values were computed with the expression

$$\Delta^{14}\mathrm{C} = (F_{\mathrm{RC}}\, e^{\lambda t} - 1)\ 1000‰ \quad (7)$$

where λ is the decay constant for the 5730-yr half-life, and t is the calendar age of the sample in years BP (before 1950), determined with the ^{230}Th technique. This value for Δ^{14}C is equivalent to the age-corrected value for Δ given in Stuiver and Polach (1977).

The total uncertainty of Δ^{14}C includes uncertainties in F_{RC} and ^{230}Th ages. Propagating these yields the expression

$$\sigma_{\Delta} = 1000 e^{\lambda t}\left[(F_{\mathrm{RC}}\lambda)^2 \sigma_t^2 + \sigma_{F_{\mathrm{RC}}}{}^2\right]^{1/2} \quad (8)$$

where σ_{Δ} is the total uncertainty in Δ^{14}C, t is the calendar age of sample in years BP, σ_t is the standard deviation reflecting the uncertainty in the age (uncertainty in the ^{230}Th date) and $\sigma_{F_{\mathrm{RC}}}$ is the uncertainty in F_{RC}.

^{230}Th Technique

^{230}Th dating of corals relies on the decay of ^{234}U (half-life 244.5×10^3 yr) to ^{230}Th (half-life 75.4×10^3 yr). Both isotopes accumulate in the coral as relatively long-lived intermediate daughter products from ^{238}U decay. Initial ^{230}Th in modern corals is negligible due to the extreme low solubility of Th in seawater. The amount of ^{230}Th dissolved in seawater is approximately equivalent to the amount produced by one year of ^{234}U decay (Edwards, Chen and Wasserburg 1987). Uranium is more soluble than thorium, and dissolved uranium becomes incorporated into a coral's skeleton as it grows. Typical uranium concentrations for the coral samples analyzed here are *ca.* 3 ppm.

Assuming a closed system and assuming zero initial ^{230}Th, the ^{230}Th age of the coral can be calculated using the equation (Broecker 1963; Broecker and Thurber 1965):

$$\left[{}^{230}\mathrm{Th}/{}^{238}\mathrm{U}\right] - 1 = -e^{-\lambda_{230}T} + \left(\delta^{234}\mathrm{U}_m/1000\right)\left[\lambda_{230}/(\lambda_{230}-\lambda_{234})\right]\left(1-e^{(\lambda_{234}-\lambda_{230})T}\right) \quad (9)$$

where the value [^{230}Th/^{238}U] is the ^{230}Th/^{238}U activity ratio, λ_{230} and λ_{234} are the decay constants for ^{230}Th and ^{234}U, T is the sample age in years, and $\delta^{234}U_m$ is the measured δ^{234}U value, defined as

$$\delta^{234}U_m = \{[(^{234}U/^{238}U)_{measured} / (^{234}U/^{238}U)_{se}] - 1\} \times 1000 \tag{10}$$

where $(^{234}U/^{238}U)_{se}$ is the ^{234}U/^{238}U ratio at secular equilibrium (Edwards, Chen and Wasserburg 1987).

The initial ^{234}U/^{238}U ratio of the corals reflects that of seawater, which is presently in excess of the secular equilibrium value by *ca.* 150‰. This value has not varied by more than 2‰ over the last 13,000 yr (Edwards *et al.* 1993). This means that significant deviations from observed initial ^{234}U/^{238}U values can be used to identify samples that may have been altered.

The weighted average initial δ^{234}U value for these corals is 148.8 ± 0.5 (2σ). This mean value is within errors of that determined for deglacial New Guinea corals (Edwards *et al.* 1993). No ^{230}Th age reversals were seen in the 13 age determinations along the core, and we observed a 1:1 relationship between the ^{230}Th ages and growth bands determined by counting layers (see below).

Coral ^{230}Th ages reported in this study were measured using thermal ionization mass spectrometry (TIMS) following the method of Edwards, Chen and Wasserburg (1987) and Edwards *et al.* (1993). ^{230}Th concentrations were measured using a ^{229}Th spike; uranium concentrations were measured using a mixed ^{233}U/^{236}U spike. Uranium measurements were made with a double zone-refined Re filament. Th measurements were made on a single zone-refined Re filament with a graphite substrate.

RESULTS AND DISCUSSION

^{230}Th and ^{14}C Results

The chronology of the *Diploastrea heliopora* core is shown diagrammatically in Figure 2. It consists of four continuous sections, punctuated by three hiatuses. The hiatuses represent times when the coral died off for some period and later recolonized. The durations of the hiatuses were determined by combining the ^{230}Th dates with growth band counts. The thorium results are given in Table 1. Differences in the ages of specific growth bands along continuous sections of coral were determined by counting bands and by computing differences between ^{230}Th dates. These two methods agree perfectly within quoted uncertainties. To obtain the most precise age estimate possible for a given piece of coral, the ^{230}Th ages were averaged after adjusting the ages of measured growth bands to the first year of growth for each piece (Table 1B). These average ages of each section were combined with the growth band counts to complete the calendar chronology of the entire core (Fig. 2). The total number of years computed in this manner is 720. The duration of all of the growth hiatuses is *ca.* 100 yr for all three hiatuses and the total number of growth years is *ca.* 400.

The ^{14}C results from the *Diploastrea* core (Table 2) are plotted in Figure 3 along with the tree ring ^{14}C data of Kromer and Spurk (1998). The two data sets overlap and the overall trend between the two sets of measurements is in good agreement. Both sets of data are plotted with 2σ uncertainties. For the coral data these are less than 1%. This uncertainty is computed as the larger of the internal or external variance in the population of annual measurements that contribute to the decadal average. The uncertainty in each annual measurement includes contributions deduced from counting statistics, machine random error and the reservoir correction.

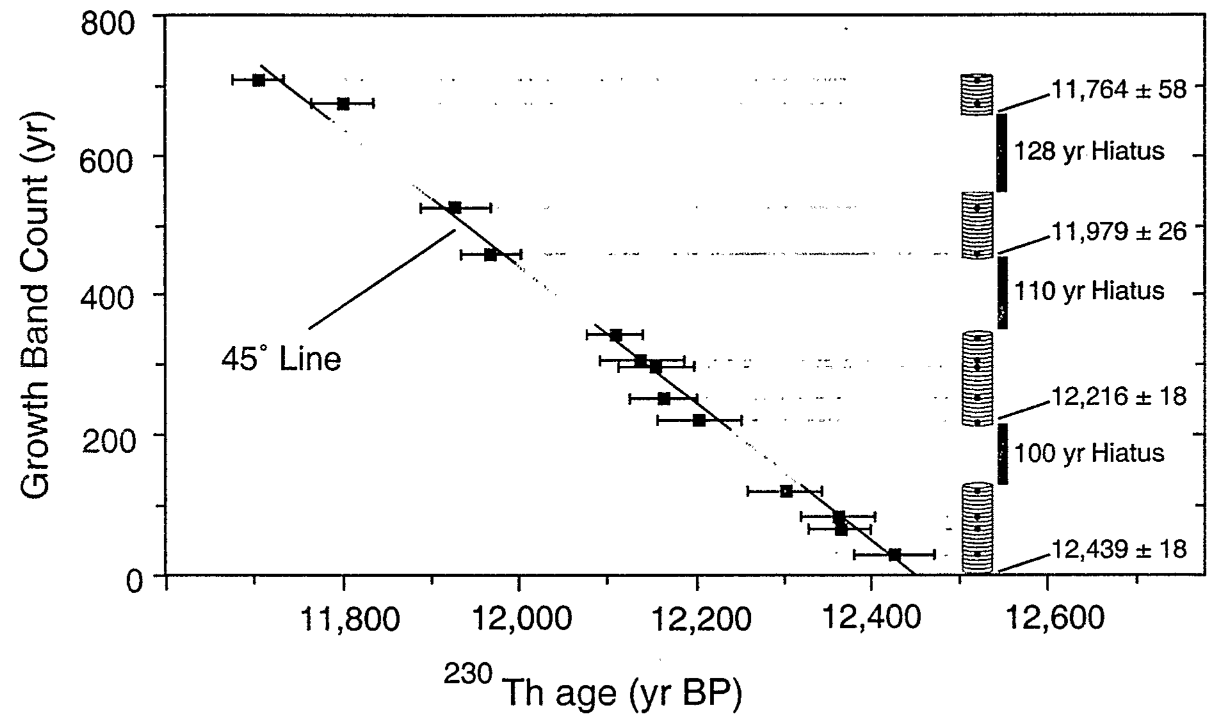

Fig. 2. Diagrammatic representation of the *Diploastrea heliopora* core showing the deduced chronology in yr BP

Fine Structure in the Record

The precision of these measurements permits our first look into the fine structure of the ^{14}C record during portions of the Younger Dryas climatic event. Significant variations (wiggles) are evident in the record. This is not surprising, as the Younger Dryas period is known to be a period of rapid change in atmospheric $\Delta^{14}C$ (Edwards *et al.* 1993). $\Delta^{14}C$ values are plotted in Figure 3B. The earliest part of this core shows a steady rise in $\Delta^{14}C$ followed by a hiatus. The second segment shows a distinct oscillation culminating in a sharp rise and a second hiatus. The third segment shows a modest rise followed by a sudden sharp rise and a hiatus; the final segment records a 50-yr period of large rapid $\Delta^{14}C$ variations. The initial sharp rise in $\Delta^{14}C$ is consistent with the global marker described by Hajdas *et al.* (Hajdas and Bonani 1997) for the onset of the Younger Dryas. The peak in the *Diploastrea* curve observed in the first section of core is also temporally coincident with the beginning of a distinct pause in sea level rise documented in the New Guinea sea level reconstruction (Edwards *et al.* 1993).

Of particular interest are the two sharp increases in $\Delta^{14}C$ prior to the death of the organism at the second and third hiatuses. Possible causes of death include 1) burial by volcanic debris or flooding, 2) sudden emergence resulting from tectonic activity or sea level variations, and 3) a rapid change in water temperature that exceeded the tolerance limits of the coral. The observed changes in $\Delta^{14}C$ prior to the second and third hiatuses cannot be explained by burial. Emergence accompanied by some recrystallization could raise the $\Delta^{14}C$ value of the coral, but the X-ray powder diffraction data and $\delta^{234}U$ results do not show any evidence of recrystallization. A rapid change in water temperature could result from changes in paleocirculation. In this case the water would be expected to have an elevated $\Delta^{14}C$ value and be either too cold or too hot for the coral to survive. This possibility can be tested using coral paleothermometry.

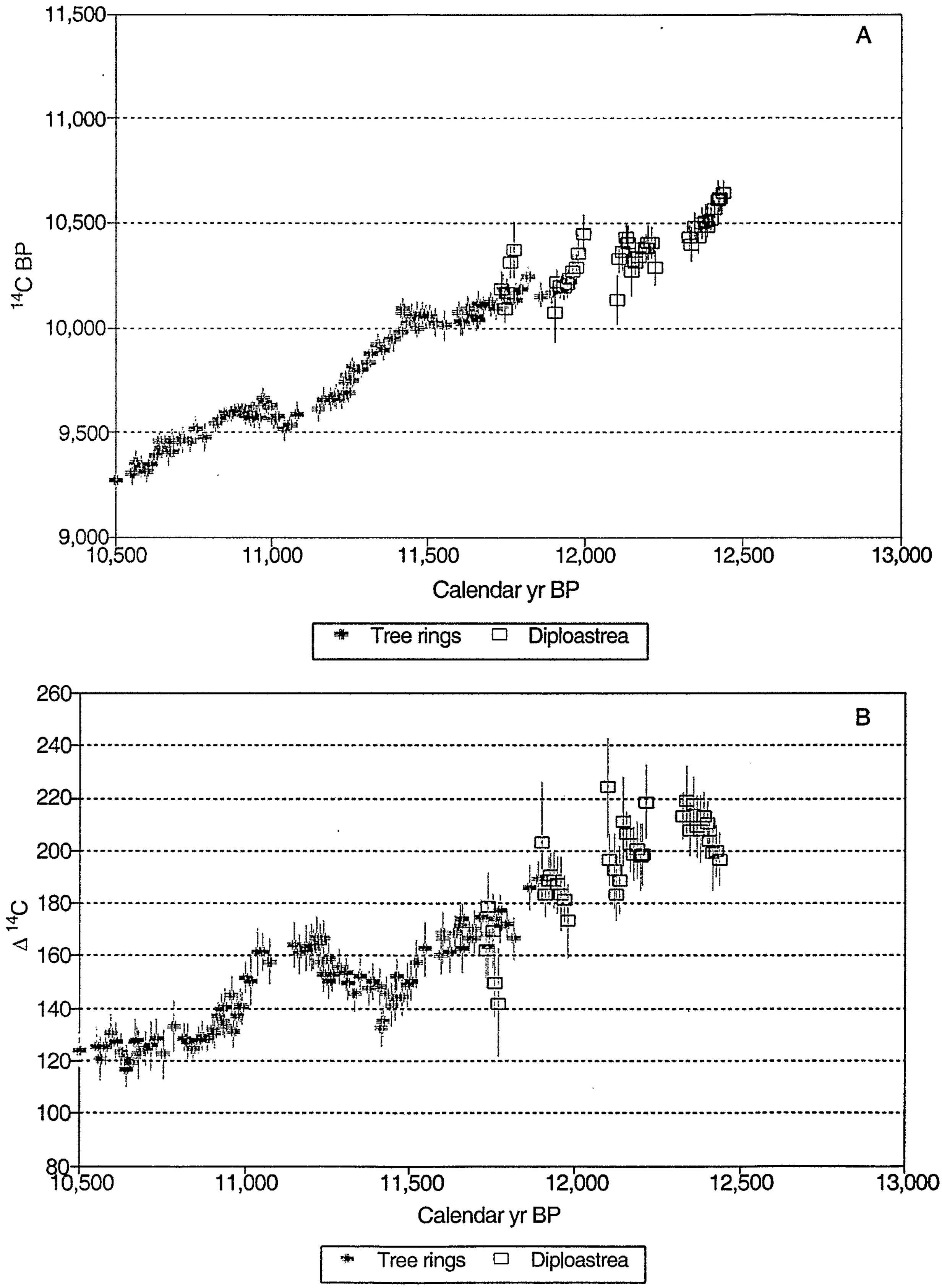

Fig. 3. Comparison of the coral data from this study with the tree ring data of Kromer and Spurk (1998, revised from Kromer and Becker 1993). All uncertainties are 2σ. A. ^{14}C ages; B. $\Delta^{14}C$ values.

TABLE 1A. ^{230}Th Results for Individual Years

Sample	^{230}Th age ± 2σ (yr BP)	$\delta^{234}U_{initial}$ ± 2σ	^{238}U (ppm)	^{232}Th (ppt)	Growth band (year number)
Top growth band	Section 4				720
9-11-12.5-b	11,705 ± 29	149.3 ± 1.0	2.357	597.0	710
9-11-12.5	11,800 ± 35	148.8 ± 1.1	2.792	800.0	675
Growth hiatus					**128 ± 64 (2σ)**
Top growth band	Section 3				549
9-12.5-14.0-1	11,928 ± 39	149.3 ± 1.0	2.970	131.0	526
9-12.5-14.0-2	11,968 ± 34	150.7 ± 1.5	2.598	28.2	460
Growth hiatus					**110 ± 32 (2σ)**
Top growth band	Section 2				350
9-12.5-14.0-3a	12,108 ± 32	146.6 ± 1.2	2.542	55.0	345
9-12.5-14.0-3b	12,138 ± 47	148.0 ± 1.1	2.590	1925	306
9-12.5-14.0-3c	12,154 ± 42	152.3 ± 1.5	2.543	49.6	297
9-12.5-14.0-4	12,163 ± 37	148.9 ± 1.3	2.691	104.0	251
9-12.5-14.0-5	12,204 ± 47	146.1 ± 1.2	2.570	1015	223
Growth hiatus					**100 ± 26 (2σ)**
Top growth band	Section 1				123
9-12.5-14.0-6a	12,301 ± 43	150.5 ± 1.0	2.520	83.0	121
9-12.5-14.0-6b	12,362 ± 30	150.5 ± 1.1	2.460	165.0	84
9-14.0-15.5-1	12,364 ± 34	149.9 ± 1.6	2.733	674.0	67
9-14.0-15.5-2	12,425 ± 46	147.4 ± 1.3	2.613	1386	31
Basal growth band					1

TABLE 1B. ^{230}Th ages of Continuous Sections of Coral Based on Weighted Mean Values

Section number	Number of years	Nominal age of oldest year in section (±2σ)
Section 4	45	11,764 ± 58
Growth hiatus	**128**	
Section 3	87	11,979 ± 26
Growth hiatus	**110**	
Section 2	127	12,216 ± 18
Growth hiatus	**100**	
Section 1	123	12,439 ± 18

Comparisons with Other Calibration Data

Other sources of calibration information with which to compare the *Diploastrea heliopora* record include: 1) other published coral results from Barbados and Mururoa (Bard *et al.* 1990; Bard *et al.* 1993), Papua New Guinea (Edwards *et al.* 1993), and Tahiti (Bard *et al.* 1996); 2) European varved lakes (Wohlfarth 1996; Wohlfarth, Björck and Possnert 1995; Hajdas *et al.* 1993, 1995; Goslar *et al.* 1992, 1995); and 3) marine varved sediments from the Cariaco basin (Hughen *et al.* 1998).

TABLE 2. Radiocarbon results. Decadal averages; weighted means from multiple measurements. The number of annual bands averaged for each result is given as n. Uncertainties are 2σ (see text).

^{230}Th age (yr BP)	^{14}C age (yr BP)	Δ^{14}C (‰)	n
11,730	10,189 ± 82	162 ± 14	4
11,740	10,086 ± 69	179 ± 13	10
11,750	10,161 ± 76	169 ± 14	10
11,760	10,308 ± 77	149 ± 13	10
11,770	10,370 ± 138	142 ± 21	2
11,900	10,077 ± 151	203 ± 23	8
11,910	10,219 ± 59	184 ± 9	10
11,920	10,195 ± 58	189 ± 9	10
11,930	10,192 ± 58	190 ± 9	10
11,940	10,213 ± 70	189 ± 11	9
11,950	10,234 ± 70	187 ± 11	10
11,960	10,268 ± 88	184 ± 14	10
11,970	10,290 ± 59	182 ± 9	10
11,980	10,357 ± 89	173 ± 14	4
12,100	10,134 ± 122	224 ± 19	6
12,110	10,327 ± 65	196 ± 10	10
12,120	10,364 ± 95	192 ± 14	10
12,130	10,435 ± 66	183 ± 10	10
12,140	10,407 ± 84	189 ± 13	10
12,150	10,268 ± 112	211 ± 17	10
12,160	10,311 ± 59	206 ± 9	10
12,170	10,342 ± 71	203 ± 11	10
12,180	10,379 ± 66	199 ± 10	10
12,190	10,379 ± 71	200 ± 11	9
12,200	10,407 ± 84	197 ± 13	10
12,210	10,410 ± 78	198 ± 12	10
12,220	10,286 ± 89	219 ± 14	6
12,330	10,429 ± 60	213 ± 9	8
12,340	10,398 ± 84	219 ± 13	10
12,350	10,483 ± 66	208 ± 10	10
12,360	10,429 ± 72	218 ± 11	10
12,370	10,495 ± 78	209 ± 12	10
12,380	10,511 ± 85	208 ± 13	10
12,390	10,486 ± 54	213 ± 9	10
12,400	10,517 ± 55	210 ± 9	10
12,410	10,568 ± 43	204 ± 7	10
12,420	10,610 ± 92	199 ± 14	10
12,430	10,616 ± 61	200 ± 10	10
12,440	10,645 ± 61	197 ± 10	9

Comparisons with previously published coral data are given in Figure 4. The agreement is within errors for all years of overlap. A comparison with European varved lakes (Fig. 5) is less definitive than the comparison with other corals, but the revised Swedish record (Wohlfarth 1996) and Lake Gościąż record (Goslar *et al.* 1995) both agree with our results, within errors. Other varved lake records from Soppensee (Hajdas *et al.* 1993) and Lake Holzmaar (Hajdas *et al.* 1995) do not agree with our coral chronology, but these European records are currently being revised (I. Hajdas, personal communication). A comparison with the marine varve record from the Cariaco basin (Hughen *et al.* 1998) is shown in Figure 6. The agreement is within errors for all points, but the two records appear to begin to diverge *ca.* 12.5 ka BP. This apparent trend could reflect a real shift in the relative reservoir ages of the two sites. More data from the two records should clarify the extent of the deviation.

Conclusion

This study extends the high-resolution ^{14}C calibration beyond the current dendrochronological limit and covers most of the period between *ca.* 11.7 and 12.4 calendar ka BP with decadal resolution. It is the first coral study to achieve routine 1% (2σ) age uncertainties with coral and the first study to exploit growth bands in coral for the purpose of ^{14}C calibration. The relatively high precision and resolution of this calibration permits a look into the fine structure of the curve during the Younger Dryas climatic episode. The data show that this was a period of large, rapid variations in $\Delta^{14}C$ superimposed on the drop in $\Delta^{14}C$ identified by Edwards *et al.* (1993). The data agree with the existing dendrocalibration, European varved records, the marine varved record and existing coral data over this time period.

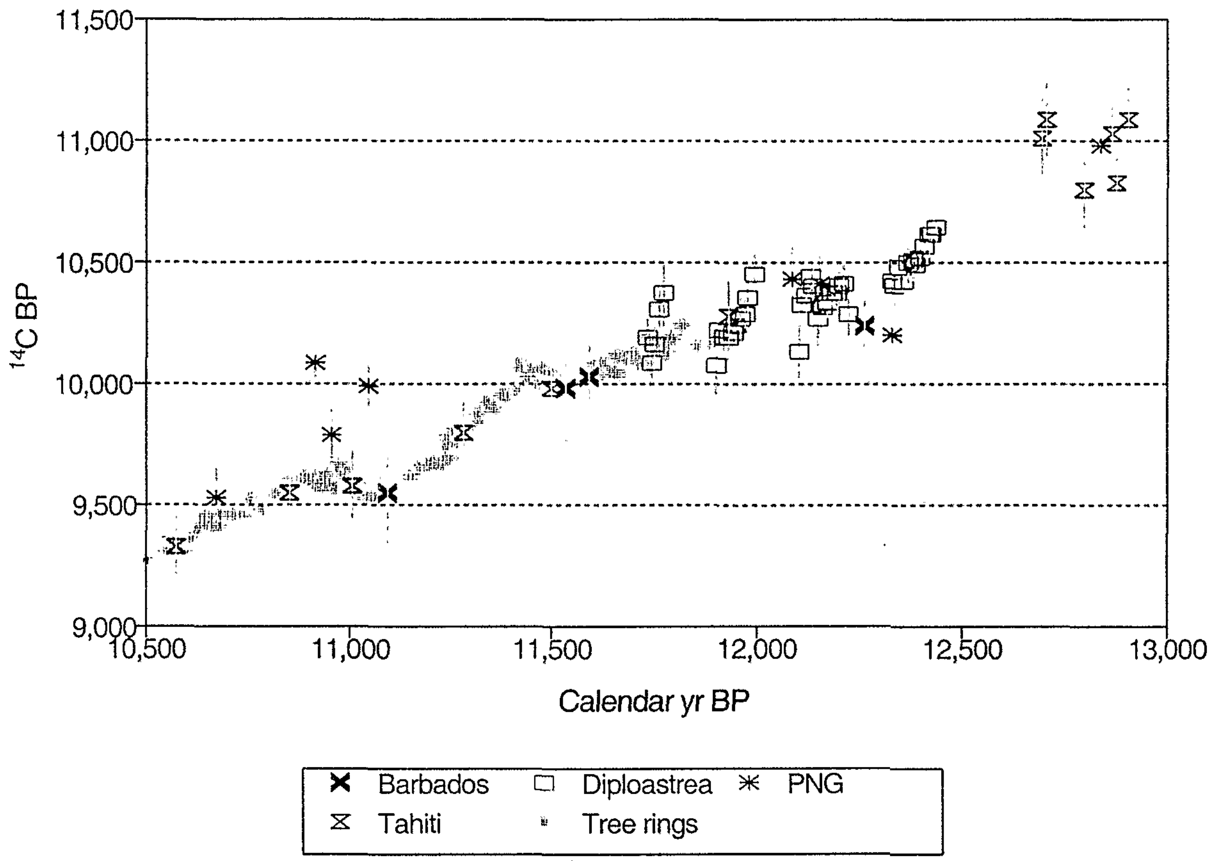

Fig. 4. Comparison of the coral data from this study with the coral data from Barbados and Mururoa (Bard *et al.* 1993), Papua New Guinea (Edwards *et al.* 1993), and Tahiti (Bard *et al.* 1996). All uncertainties are 2σ.

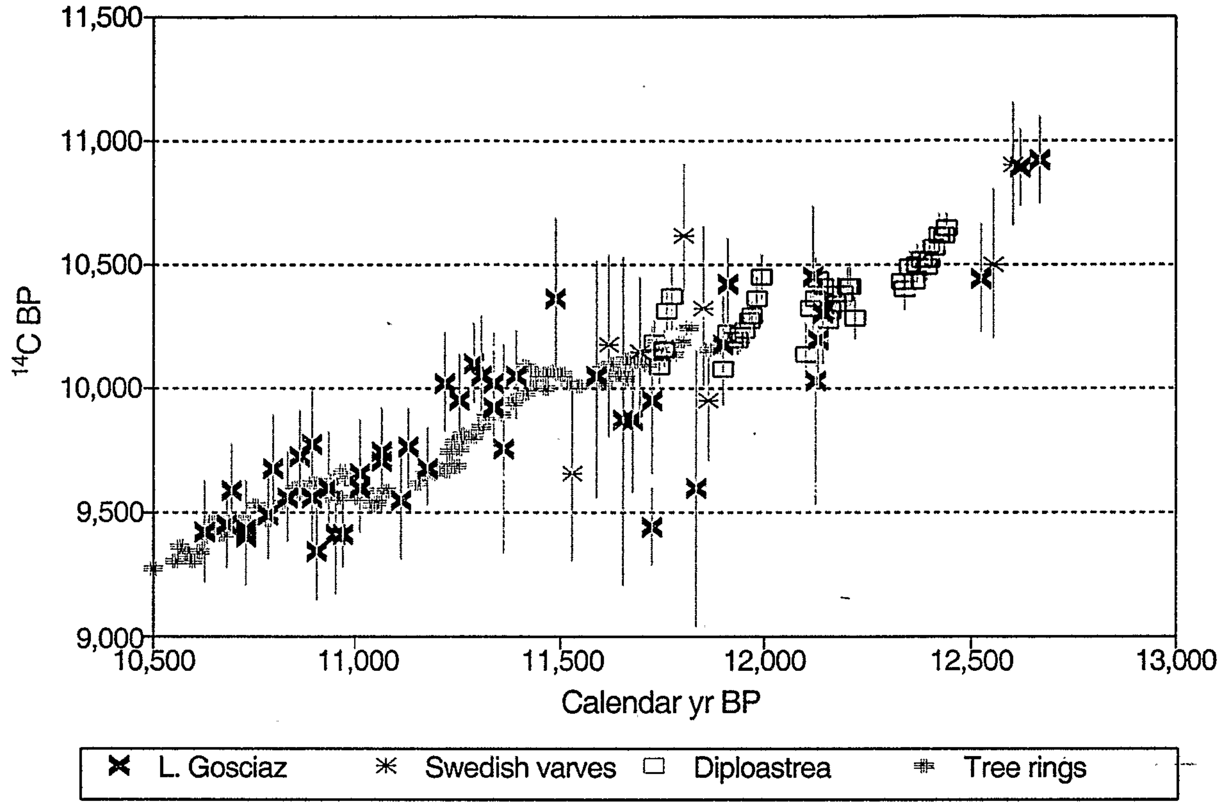

Fig. 5. Comparison of the coral data from this study with European varve data from Sweden (Wohlfarth 1996), and Lake Gościąż, Poland (Goslar *et al.* 1995). All uncertainties are 2σ.

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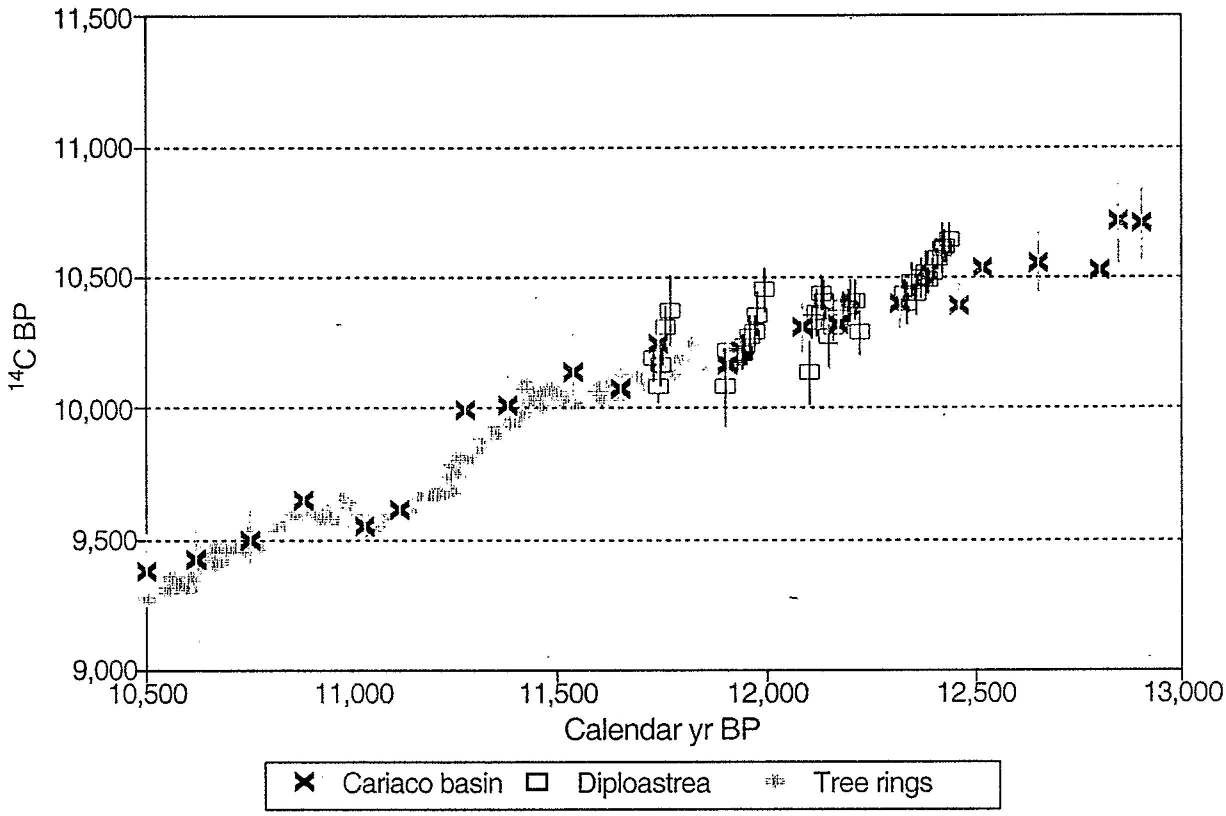

Fig. 6. Comparison of the coral data from this study with Cariaco basin marine varve data (Hughen *et al.* 1998). All uncertainties are 2σ.

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