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Title

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Permalink

Journal

ISSN

Authors

Druffel, Ellen R. M
Griffin, Sheila

Publication Date

1993

DOI

Copyright Information

Peer reviewed

Large Variations of Surface Ocean Radiocarbon: Evidence of Circulation Changes in the Southwestern Pacific

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Radiocarbon ($\Delta^{14}C$) and stable isotope ($\delta^{18}O$ and $\delta^{13}C$) records are presented for biannual samples from a 323-year banded coral series collected from the southern Great Barrier Reef, Australia. The high-precision $\Delta^{14}C$ record contains variations on an interannual timescale, that are particularly large between A.D. 1680 and 1730. By comparison with tree ring $\Delta^{14}C$ records [*Stuiver and Quay*, 1980; M. Stuiver, personal communication, 1992), it is clear that these shifts were not caused by changes in the $\Delta^{14}C$ of atmospheric CO_2. Changes in vertical mixing and large scale advective changes involving source waters to the western Coral Sea region are likely processes that could account for these large $\Delta^{14}C$ variations. Most low $\Delta^{14}C$ values for the period A.D. 1635–1875 coincide with El Niño/Southern Oscillation (ENSO) events as reported by *Quinn et al.* [1987] for the eastern tropical Pacific. However, ENSO does not explain all of the variations, especially during 1875–1920 when $\Delta^{14}C$ values remained high. Cross-spectral analysis of the early half of the $\Delta^{14}C$ and $\delta^{18}O$ records (A.D. 1635–1795) reveals that the 6-year period is coherent; this coherency is not present in the latter half (A.D. 1797–1957) of the isotope records. These data support the concept of century timescale changes in the nature of ENSO, as it is manifest in the southwestern Pacific. Our coral record shows no evidence of a Suess effect, the lowering of $\Delta^{14}C$ from late 1800s through 1955 due mainly to CO_2 input from fossil fuel burning. This is coincident with the change we observe in the nature of ENSO and is further evidence that a long-term change in mixing of upper waters occurred in this region.

1. INTRODUCTION

Prior to nuclear weapons testing in the 1950s and early 1960s, radiocarbon was produced in the stratosphere solely by the reaction of cosmic ray secondaries (neutrons) and atomic nitrogen. Prebomb $\Delta^{14}C$ of atmospheric CO_2, as reconstructed from dendrochronologically dated tree rings, varied by 10–20‰ on century timescales [*Suess*, 1958, 1968; *Stuiver*, 1961]. *Stuiver et al.*, [1991] assert that the similarity (in timing and magnitude) of the records of tree ring $\Delta^{14}C$ and ice core [10]Be [*Beer et al.*, 1988] indicates that solar forcing appears to have been the primary cause of century timescale variations.

A question that is yet unanswered is the role that the oceans have played in controlling the atmospheric $\Delta^{14}C$ record. Changes in average wind speed or rates of ocean mixing, if sufficiently large, could cause changes in the $\Delta^{14}C$ of atmospheric CO_2. Time histories of high-precision $\Delta^{14}C$ values for the surface waters of the ocean are the missing information needed to quantify the oceanic contribution toward shaping the observed variations in prebomb atmospheric $\Delta^{14}C$ records.

Most hermatypic, or reef-building, corals that grow in the surface ocean accrete aragonitic skeletons at rapid rates (0.2–2 cm/yr). Many of the coral species studied to date contain annual variations in skeletal density which are discernible by X-ray of a 1-cm-thick slab [*Knutson et al.*, 1972]. The source of carbonate to the skeleton is primarily from dissolved inorganic carbon (DIC) in the surrounding seawa-

ter. Thus, within the coral skeletons lie annual records of past seawater $\Delta^{14}C$ values [e.g., *Nozaki et al.*, 1978; *Druffel and Linick*, 1978; *Druffel*, 1987]. The coral skeleton also holds a storehouse of other past chemical and isotopic information, including trace element content [*Shen and Boyle*, 1988; *Linn et al.*, 1990], stable carbon and oxygen isotope values [*Weber et al.*, 1975; *Dunbar et al.*, 1991] and fulvic acid concentrations [*Isdale*, 1984] in seawater.

Most coral $\Delta^{14}C$ records are from the postbomb (after 1955) period, and with the exception of postbomb results from Bermuda [*Druffel*, 1989], all reported $\Delta^{14}C$ measurements in banded corals are of average precision (± 4-8‰). Thus, high-precision $\Delta^{14}C$ measurements from the prebomb period must be obtained if comparison with high-precision tree ring $\Delta^{14}C$ records, which display variations of the order of only 20‰ [*Stuiver and Quay*, 1980], are to be meaningful. This is especially important given that a $\Delta^{14}C$ perturbation in the atmosphere will appear only as half of that in the surface ocean [*Oeschger et al.*, 1975] as a result of slow equilibration time, and mixing between surface and subsurface waters.

2. SAMPLING LOCATION

A single colony of *Porites australiensis*, standing 7.5 m high, was sampled for this study. The coral base was at 10 m water depth on the leeward, outer side of Abraham Reef (22°06'S, 153°00'E). Abraham Reef is located in the Swains Reef complex in the southwestern tip of the outer Great Barrier Reef, approximately 200 km offshore from Gladstone in Queensland, Australia. The coral site is well flushed by open ocean waters that are derived from southwestward transport.

A 9.5-cm diameter core was taken by P. Isdale and colleagues (Australian Institute of Marine Science) with a pneumatic underwater drill from the vertical axis of the coral

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in December, 1985. Nine sections of core were taken, with a total core length of 4.94 m. We recovered a longer core of this colony in May 1991, and results from this study will be forthcoming.

3. METHODS

The sections of core were air dried, then cut with a water-cooled rock saw along the axis of corallite growth. Slabs 9 mm thick were X-rayed to detect periodic variations in the density of the accreted aragonite. Confirmation of the annual periodicity of the high- and low-density band couplets is evident from the distribution of bomb ^{14}C in the coral (E.R.M. Druffel, manuscript in preparation, 1993) and δ^{18}O results from 7–10 subsamples per band in the earlier part of the core. The growing surface of this coral species is irregular, which made it difficult to observe the annual density bands in some sections of the core. Thus, a larger than normal error (±5 years) has been assigned to the oldest bands from this core (A.D. 1635). A band saw was used [Griffin and Druffel, 1985] to cut the coral slabs into biannual samples for the early growth (before A.D. 1948) and annual samples for later growth (A.D. 1948–1957). In this paper, dates for samples prior to 1948 will be referred to as the latter year of the 2-year pair, i.e., 1946–1947 is referred to as 1947. The cut was made directly underneath the high-density band, which makes the midpoint of each 2-year band about December of the first year. Radiocarbon measurements were performed on coral samples (~25 g) that had been acidified and converted to acetylene gas via a lithium carbide intermediate [Griffin and Druffel, 1985]. Samples were counted in six quartz gas proportional beta counters (all volumes 1.5 L) for six to seven 2-day periods at 900 mm of Hg and 22°C.

All Δ^{14}C measurements were corrected for isotope fractionation to a δ^{13}C (relative to PDB-1) of -25.0‰ and for decay since the time of formation [Stuiver and Polach, 1977]. The δ^{13}C values for this correction were measured on CO_2 produced from reburned acetylene gas. Errors on the Δ^{14}C measurements based on counting statistics were ±2.0‰. Laboratory or system random errors are 25% of this value for modern samples; thus the reported error is ±2.5‰. All of the biannual and annual coral samples were measured for δ^{13}C and δ^{18}O using previously described methods [Druffel, 1989]. In addition, selected 4 to 6 year portions of the core were ground off using a dremel tool into 0.10 to 0.15-year wide layers and measured for stable isotopes. Isotope ratios were measured on a V.G. Micromass 602E mass spectrometer. The error determined from the standard deviation of replicate analyses of numerous coral samples was ±0.09‰ for both δ^{18}O and δ^{13}C.

4. RESULTS

4.1. High-Precision Radiocarbon in Australian Corals

The high-precision Δ^{14}C time history for the Abraham Reef coral is presented in Figure 1a and the measurements are listed in Table 1. The values range from -58‰ in 1865 to -30‰ in 1691. The mean of the first half (pre-1800) of the Δ^{14}C values (-45.0 ± 5.6 (SD)‰, N=75) is higher than that for post-1800 values (-49.6 ± 3.6 (SD)‰, N=74). Overall, the standard deviation in the pre-1800 data is nearly twice that in the later data.

Δ^{14}C shifts from very high to very low values from 1677 to 1687, 1691 to 1703, and 1727 to 1729. These 20–22‰ shifts in Δ^{14}C are much larger than those expected from analytical scatter, as the 1σ error of the measurements (±2.5‰) is a factor of 8–9 times smaller than the range of values. In fact, these large shifts in Δ^{14}C are similar to the entire spatial range of prebomb (A.D. 1955) Δ^{14}C values in Pacific surface waters between the central ocean gyres (-35 to -45‰) and the nutrient-rich tongue off the coast of Peru in the eastern tropical Pacific (-65‰) [Druffel, 1981]. Subsequent to 1729, the interannual variability of the Δ^{14}C values is significantly less than that in the preceding period.

The temporal variations in the post-1800 results are smaller and generally occur at a slower rate. The magnitudes of these variations are 4 to 7 times greater than the 1σ error of the individual measurements, less than those observed for the late seventeenth and early eighteenth century data.

4.2. Comparison of Coral and Tree Ring Δ^{14}C

In order for atmospheric forcing to have directly caused the biannual Δ^{14}C variations (20–22‰) recorded in the coral, atmospheric values would need to have varied by roughly twice this, or 40–45‰. A range of only $\leq$5‰ is observed in the annual northern hemisphere tree ring record from the 1600s and 1700s (M. Stuiver, personal communication, 1992). One would not expect the southern hemisphere Δ^{14}C record in the atmosphere to have been much different, owing to the short mixing time of CO_2 between hemispheres. Thus atmospheric control of the short-term changes in surface ocean Δ^{14}C is not plausible.

Conversely, some of the long-term variations in the coral Δ^{14}C record do appear to have been caused by atmospheric forcing. In general, the tree ring data show a broad Δ^{14}C maximum during the Maunder minimum in sunspot numbers (A.D. 1654–1714) [Eddy, 1976] and a decrease in Δ^{14}C after A.D. 1900 (Figure 1a) due to fossil fuel dilution and solar variation, known as the Suess effect [Suess, 1953]. Similarly, the coral record shows on average higher Δ^{14}C values during the Maunder minimum. However, there is little suggestion of a Suess effect in the coral record after 1900. Overall, the decadal-averaged tree ring and biannual coral Δ^{14}C records are similar in shape prior to 1900, with larger trends, as expected, in the tree ring data.

To determine the deviation of the long term coral trend from that expected from the atmospheric tree ring record, we compare our results (boxcar averaged over 20-year increments) with the model-calculated Δ^{14}C record of the mixed layer (using bidecadal atmospheric Δ^{14}C values and a box-diffusion model of the oceans) [Stuiver et al., 1986] (see Figure 1b). The two records follow the same general trends until 1900. After 1900, the coral values do not decrease as would have been expected by the Suess effect observed in the model-calculated Δ^{14}C record.

The absence of a Suess effect in the coral data is perplexing. A Suess effect of about 12‰ was observed in southern hemisphere tree rings, approximately two-thirds that observed in the northern hemisphere (M. Stuiver, personal communication, 1992). Thus, we would expect to observe a 6 ± 2‰ decrease in the coral Δ^{14}C from 1900 to 1955. Is it possible that a several-decade timescale change in circulation masked the Suess effect at this southwestern Pacific
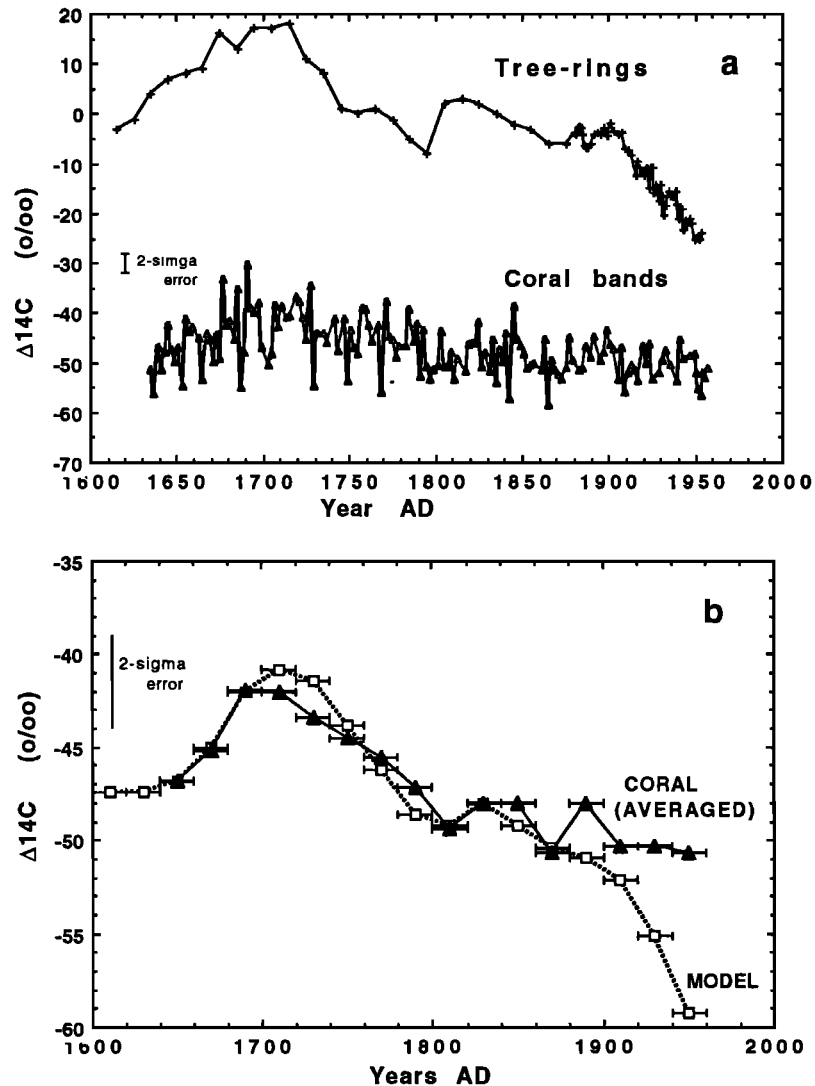
Fig. 1. (a) High-precision $\Delta^{14}C$ measurements in biannual (for A.D. 1635–1947) and annual (for A.D. 1948–1957) coral bands from the core described in the text from Abraham Reef ($22°06'S$, $153°00'E$) in the Swains, southern Great Barrier Reef region. The pluses show the high-precision $\Delta^{14}C$ measurements of *Stuiver and Quay* [1980] for decadal tree ring samples (representative of atmospheric CO_2) from the northwestern U.S. region. Errors based on counting statistics and laboratory errors are $\pm 2.5\%_o$ for the coral results and $\pm 2.0\%_o$ for the tree ring results. (b) Coral $\Delta^{14}C$ results, boxcar averaged over 20-year increments (where missing values were interpolated and included in the average), and model-calculated $\Delta^{14}C$ of the mixed layer, using bidecadal atmosphere $\Delta^{14}C$ values and a box-diffusion model of the oceans, assuming constant atmospheric CO_2 concentration, constant biospheric reservoir sizes, and constant gas exchange rates (values reported by *Stuiver et al.* [1986, Figure 10a]).

location? Or, as *Toggweiler et al.* [1991] suggest, was it the combination of the large-scale overturning of the South Pacific and the short time that this water has to equilibrate its $^{14}C/^{12}C$ ratio with atmospheric CO_2 that explains the low Suess effect in the South Pacific? These possibilities are discussed later in the paper.

4.3. Correlation Between Coral $\Delta^{14}C$ and ENSO Events

El Niño/Southern Oscillation (ENSO) events are interannual fluctuations in atmospheric and oceanic circulation that occur every 2–10 years [*Enfield*, 1988]. They are associated with periods of cool sea surface temperature (SST) anomalies in the Indonesian region ($5°-15°S$, $120°-160°E$), which precede the El Niño warming in the east Pacific and the changes in the Southern Oscillation by about one season [*Nicholls*, 1984]. It appears that past ENSO events correlate

with the Abraham Reef $\Delta^{14}C$ record (Figure 2). To assist the comparison, we have detrended the $\Delta^{14}C$ record by subtracting the individual values from a third-order polynomial fit of the data. ENSO events, as reported by *Quinn et al.* [1987], coincide with most low detrended $\Delta^{14}C$ values in the Australian coral (solid peaks, Figure 2). This agreement indicates that our Abraham Reef site is influenced by large scale circulation patterns and not merely by local changes. Some correlations are offset by $\pm 1-2$ sampling increments ($\pm 2-4$ years), which is within the error of the coral chronology. The *Quinn et al.* [1987] index was established using data from the west coast of northern South America and its adjacent Pacific Ocean waters. However, it has been shown that ENSO events affect the western, central, and eastern tropical Pacific zones differently with respect to precipitation [*Schonher and Nicholson*, 1989] and SST anomaly pat-

TABLE 1. Radiocarbon and Stable Isotope Values for
Abraham Reef Coral, 1635–1957

TABLE 1. (continued)

WH#	Year	$\Delta^{14}C$, ‰	$\delta^{13}C$, ‰	$\delta^{18}O$, ‰	WH#	Year	$\Delta^{14}C$, ‰	$\delta^{13}C$, ‰	$\delta^{18}O$, ‰
1145	1635	-51.3	-1.58	-4.81	955	1763	-45.6	-0.80	-5.18
1199	1637	-56.1	-1.27	-4.59	968	1765		-0.32	-4.85
703	1639	-46.9	-1.62	-5.10	788	1767	-42.6	-1.04	-4.95
1141	1641	-51.5	-1.24	-4.76	1254	1769	-56.0	-1.26	-5.00
1197	1643	-48.0	-1.36	-5.03	999	1771	-37.7	-1.37	-5.12
702	1645	-42.4	-1.38	-4.93	956	1773	-44.6	-1.29	-4.90
1156	1647	-47.4	-1.48	-4.84	1233	1775	-44.1	-1.13	-5.07
1206	1649	-49.8	-1.50	-4.99	968	1777	-48.9	-1.81	-4.94
692	1651	-46.8	-1.41	-5.00	1078	1779	-46.7	-1.55	-5.03
1179	1653	-54.5	-1.74	-4.92		1781		-1.62	-4.99
1194	1655	-41.1	-1.34	-4.78	969	1783	-46.5	-0.52	-5.09
706	1657	-43.7	-2.00	-4.83	1253	1785	-40.4	-0.59	-5.11
1177	1659	-42.8	-1.18	-5.03	970	1787	-45.7	-1.94	-5.19
	1661		-1.38	-4.95	1142	1789	-42.2	-0.61	-4.96
727	1663	-44.9	-1.16	-4.79	1136	1791	-52.6	-0.36	-5.02
1143	1665	-53.2	-1.19	-4.68	1046	1793	-43.4	-0.81	-5.30
1207	1667	-44.1	-1.21	-4.94	1159	1795	-50.8	-0.62	-5.26
730	1669	-45.3	-1.07	-4.71	1174	1797	-53.4	-0.60	-5.08
1160	1671	-49.8	-0.96	-4.98	1052	1799	-51.4	-0.99	-5.12
1198	1673	-44.3	-1.02	-5.03	1146	1801	-50.6	-1.30	-5.14
699	1675	-49.3	-0.88	-4.80	1257	1803	-44.6	-0.67	-5.14
1070	1677	-33.2	-0.88	-5.06	1063	1805	-50.8	-0.84	-5.07
1196	1679	-42.4	-1.06	-5.11	1163	1807	-50.7	-1.08	-5.07
721	1681	-41.6	-1.20	-4.80	1213	1809	-47.9	-0.98	-5.08
1132	1683	-45.3	-1.33	-4.93	1065	1811	-53.5	-1.21	-5.06
1192	1685	-35.0	-1.16	-4.97	1168	1813	-49.3	-1.13	-4.91
695	1687	-55.0	-1.36	-5.03		1815		-1.09	-5.03
778	1689	-48.0	-1.57	-5.25	1066	1817	-51.8	-0.64	-5.06
683	1691	-30.3	-1.13	-4.89	1164	1819	-46.2	-0.51	-4.98
734	1693	-39.0	-0.79	-5.04	1217	1821	-45.9	-0.57	-5.04
862	1695	-40.0	-0.61	-5.21	1062	1823	-46.1	-0.34	-5.12
1191	1697	-38.1	-0.62	-5.22	1162	1825	-41.9	-0.69	-5.26
696	1699	-47.0	-1.11	-5.25	1211	1827	-50.7	-0.40	-5.08
	1701		-0.40	-5.08	1068	1829	-47.9	-0.37	-4.99
859	1703	-50.6	-1.22	-5.26	1170	1831	-51.6	-0.41	-5.02
691	1705	-48.3	-0.84	-4.97	1180	1833	-45.4	-0.84	-5.07
857	1707	-38.4	-1.10	-4.79	1127	1835	-54.0	-0.55	-5.12
1195	1709	-42.9	-1.07	-5.05	1167	1837	-47.1	-0.78	-5.12
750	1711	-38.5	-1.44	-5.16	1216	1839	-52.1	-0.68	-4.79
1242	1713	-40.7	-1.60	-5.03	1069	1841	-44.2	-0.70	-5.09
780	1715	-40.5	-1.07	-5.07	1152	1843	-57.2	-0.65	-4.99
	1717		-1.63	-5.15	1190	1845	-38.7	-0.63	-4.98
865	1719	-36.6	-0.94	-4.98	1051	1847	-45.3	-0.61	-4.91
1240	1721	-38.1	-0.46	-5.00	1169	1849	-46.7	-0.74	-5.15
737	1723	-40.8	-0.74	-4.83	1210	1851	-48.1	-0.66	-5.06
1071	1725	-45.3	-0.78	-5.10	1073	1853	-51.1	-0.70	-5.01
1239	1727	-34.5	-0.79	-5.17	1182	1855	-50.5	-0.26	-5.02
688	1729	-54.6	-0.91	-5.08	1147	1857	-50.3	-0.49	-4.81
864	1731	-44.1	-0.79	-4.94		1859		-0.46	-5.09
1256	1733		-0.32	-5.03	1137	1861	-51.3	-0.41	-4.97
747	1735	-44.1	-1.02	-5.02	1226	1863	-45.2	-0.46	-5.13
1130	1737	-46.1	-1.21	-4.81	1057	1865	-58.4	-0.76	-5.05
1255	1739		-1.27	-5.07	1172	1867	-49.6	-0.72	-4.82
760	1741	-41.1	-0.67	-5.05	1171	1869	-51.5	-0.72	-5.10
1128	1743	-47.5	-0.51	-5.21	1126	1871	-52.5	-0.62	-5.15
	1745		-0.69	-5.00	1155	1873	-53.5	-0.54	-5.40
754	1747	-41.3	-0.61	-4.96	1176	1875	-51.0	-0.51	-5.21
1072	1749	-53.7	-0.71	-5.01	1084	1877	-44.9	-0.64	-5.05
1237	1751	-43.4	-0.84	-5.26	1151	1879	-49.4	-0.43	-4.95
752	1753	-46.8	-1.05	-4.85		1881		-0.28	-4.97
1088	1755	-48.1	-0.84	-5.05	1087	1883	-51.3	-0.54	-5.10
1184	1757	-39.0	-0.76	-5.02	1173	1885	-51.0	-0.33	-5.16
1314	1759	-40.1	-0.53	-4.94	1223	1887	-49.0	-0.67	-5.09
1252	1761	-42.3	-0.66	-4.93	786	1889	-49.0	-0.87	-5.30

TABLE 1. (continued)

WH#	Year	$\Delta^{14}C$, ‰	$\delta^{13}C$, ‰	$\delta^{18}O$, ‰
1161	1891	-44.6	-0.48	-5.24
	1893		-0.46	-5.02
1054	1895	-49.6	-0.53	-5.37
1148	1897	-48.0	-0.99	-5.71
1212	1899	-43.5	-0.58	-5.09
1049	1901	-46.2	-0.78	-5.07
1175	1903	-47.1	-0.79	-5.24
	1905	-53.2	-0.77	-5.26
1228	1907	-46.8	-0.73	-5.21
1122	1909	-55.8	-0.76	-5.12
1140	1911	-52.0	-0.48	-5.24
1053	1913	-50.4	-0.69	-5.22
1241	1915	-51.5	-0.86	-4.98
1119	1917	-53.7	-0.89	-5.14
1243	1919	-46.6	-0.83	-5.01
1055	1921	-50.2	-0.68	-4.79
1234	1923	-46.4	-0.67	-5.25
797	1925	-53.3	-0.49	-5.15
	1927		-0.71	-5.30
1047	1929	-52.0	-0.92	-5.44
1181	1931	-49.5	-0.50	-5.16
813	1933	-47.5	-1.00	-5.42
1144	1935	-50.4	-0.74	-5.06
	1937		-0.67	-5.12
813	1939	-53.6	-0.61	-5.02
1189	1941	-44.1	-0.69	-4.89
1044	1943	-49.1	-0.77	-5.10
	1945		-0.92	-5.28
812	1947	-48.7	-1.11	-5.13
	1948		-0.72	-4.94
1093	1949	-48.2	-0.94	-5.05
811	1950	-52.2	-0.30	-5.97
1187	1951	-55.4	-0.91	-5.01
	1952		-1.29	-5.31
1083	1953	-56.5	-1.43	-5.08
1188	1954	-52.1	-1.13	-5.22
1064	1955	-53.0	-1.31	-5.27
	1956		-0.89	-5.13
1056	1957	-51.2	-0.80	-5.15

terns [*Fu et al.*, 1986; *Rasmusson and Wallis*, 1983]. Thus, we do not expect the western Pacific ENSO record to agree completely with Quinn et al.'s ENSO index of the eastern tropical Pacific.

Along these lines, there are a number of ENSO events that coincided with high $\Delta^{14}C$ values (stippled peaks, Figure 2). It is important to note that the majority of these occur within an anomolous period of 1875-1920. This anomolous period also encompasses the first half of the Suess effect in the atmosphere (see Figure 1a). Is it conceivable that the absence of a Suess effect in the Abraham coral could have been a result of long-term changes in circulation associated with the high-$\Delta^{14}C$ waters present during ENSOs in the 1875–1920 period? Intuitively, the lack of a Suess effect might imply the presence of deeper waters less affected by fossil fuel CO_2 from the atmosphere. However, deeper waters would bring in a low $\Delta^{14}C$ signal, and a source of high $\Delta^{14}C$ waters would have been needed to maintain the high coral $\Delta^{14}C$ values up until 1955.

A most striking feature of the coral $\Delta^{14}C$ record is the large range of 20-22‰ before 1730, and the smaller range in values thereafter. Annual $\Delta^{14}C$ measurements at Abraham Reef in the 1950s (Table 1) reveal decreases from the norm of 2-7‰ during the ENSO events of 1951, 1953, and 1957, whereas those decreases during the late 1600s and early 1700s were 10-12‰. Keeping in mind that annual bands were sampled for the 1950s, and biannual bands were sampled before 1947, the actual amplitude of $\Delta^{14}C$ changes 300 years ago could have been as much as 3 times the ENSO variations in the 1950s. This would imply that a large change in the intensity of ENSO events had occurred, as manifested by surface DIC $\Delta^{14}C$ in the southwest Pacific.

4.4. Stable Isotope Records

The $\delta^{18}O$ results for splits of the same samples used for the $\Delta^{14}C$ analyses are shown in Figure 3a and listed in Table 1. Most values lie between -4.8 and -5.4‰. Over the 323-year record, both the least squares fit and the third-order polynomial fit of the data reveal a decline of 0.3-0.4‰. Superimposed on this trend are variations that occur on several-year to decade timescales; the amplitudes of these
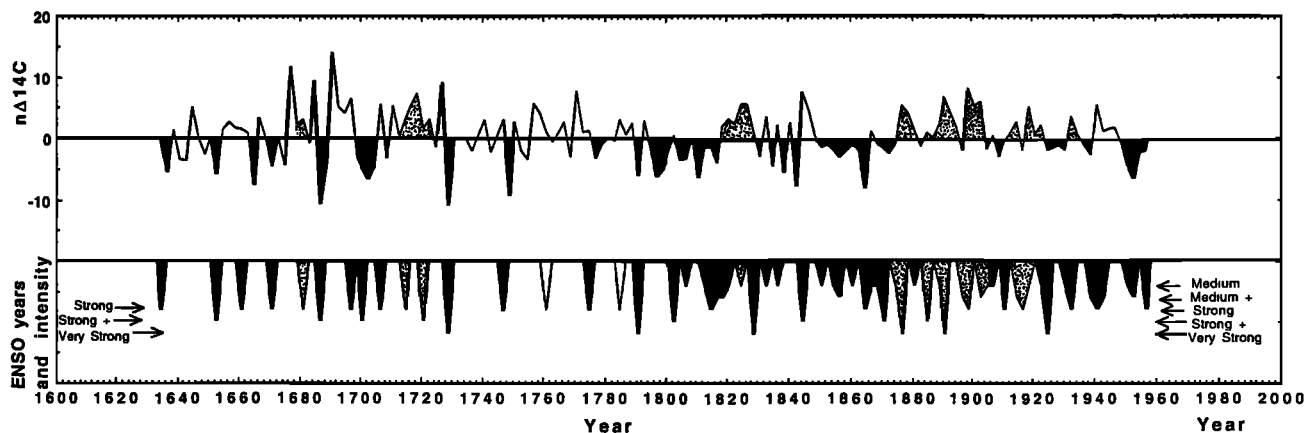

Fig. 2. Biannual Australian coral $\Delta^{14}C$ values (detrended with respect to a third-order polynomial fit) plotted with incidences of strong and very strong ENSO events before A.D. 1800 and including medium ENSO events after A.D. 1800 (plotted biannually) as reported by *Quinn et al.* [1987]. Solid peaks represent correlation between ENSO events and coincident (±2–4 years) $n\Delta^{14}C$ lows, stippled areas represent correlation between ENSO events and coincident $\Delta^{14}C$ highs, and open areas indicate no correlation.
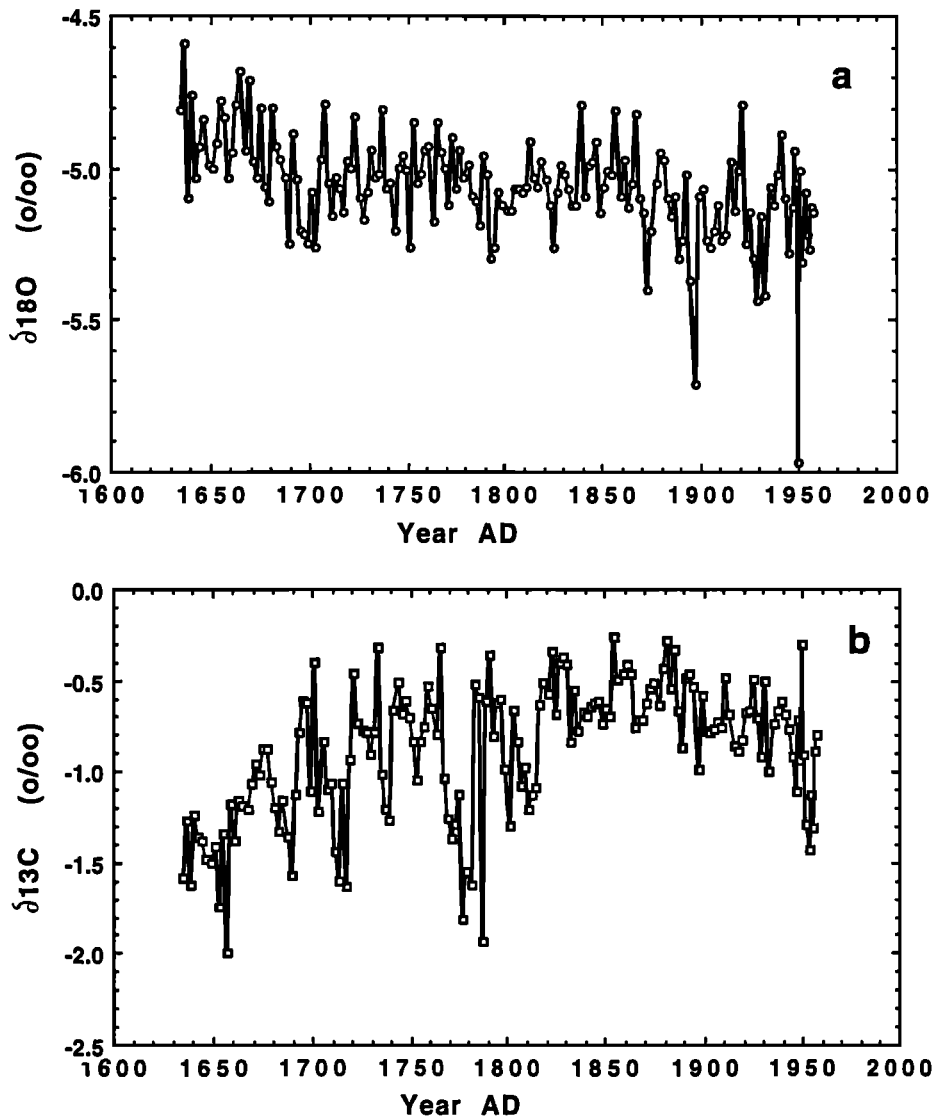
Fig. 3. (a) Records of $\delta^{18}O$ and (b) $\delta^{13}C$ for biannual coral samples in per mil (‰). Lines connect individual points. See text for details.

variations appear to be less during the pre-1870 period (0.1–0.2‰) than those after 1870 (0.3–0.5‰).

The $\delta^{13}C$ results for the biannual samples span a much larger range, from about -1.8 to -0.4‰ (Figure 3b). A least squares fit of these results reveals an increase in $\delta^{13}C$ of 0.5‰ over the 323-year record, though the increase is not monotonic. The $\delta^{13}C$ results show prominent shifts, with several-decade-long periods, and much higher amplitudes during the earlier part of the record (pre-1800).

The seasonal $\delta^{18}O$ measurements range from -5.8 to -4.0‰, with a mean value of -4.9‰(Figure 4a). Seasonal fluctuations are apparent, with a range from 0.6 to 1.2‰. About two thirds of the $\delta^{18}O$ results from high density bands (solid symbols in Figure 4a) appear at or near high $\delta^{18}O$ values, i.e., during low SST or southern winter.

The seasonal $\delta^{13}C$ measurements range from -2.2 to +0.2‰, with a mean value of -1.0‰ (Figure 4b). The seasonal regularity of the $\delta^{13}C$ record is not as apparent as that in the $\delta^{18}O$ data, and is not expected owing to the preponderance of vital effects that plague coral $\delta^{13}C$ records [Swart, 1983].

4.5. Parameters Controlling Seasonal $\delta^{18}O$ and $\delta^{13}C$

The two primary factors controlling the $\delta^{18}O$ in corals are SST and $\delta^{18}O$ of the seawater, which is directly correlated with salinity [Epstein and Mayeda, 1953; Craig and Gordon, 1965; Dunbar and Wellington, 1981]. However, observations of salinity on a seasonal basis varied by less than 0.3‰ in the 1960s [Rochford, 1968], and would thus translate to less than 0.1‰ change in coral $\delta^{18}O$ over an annual cycle. Although we do not have the salinity data to eliminate the possibility of significant seawater $\delta^{18}O$ variations over our observation period, the potential remains as our site is located near the rainiest sector on Earth.

Linear relationships between $\delta^{18}O$ in corals and SST have a slope of 0.22‰ per 1°C [Fairbanks and Dodge, 1979; Druffel, 1985]. Bimonthly color SST charts obtained from NOAA AVHRR data surrounding Abraham Reef from 1988–1992 show a seasonal temperature range of 6 ± 1°C (equivalent to a range in $\delta^{18}O$ values of 1.1–1.5‰). The range in $\delta^{18}O$ values observed in the coral during the late 1600s and early 1700s was 0.6–1.2‰ (Figure 4a), equal to or less than those expected from present-day SST observations. Assum-
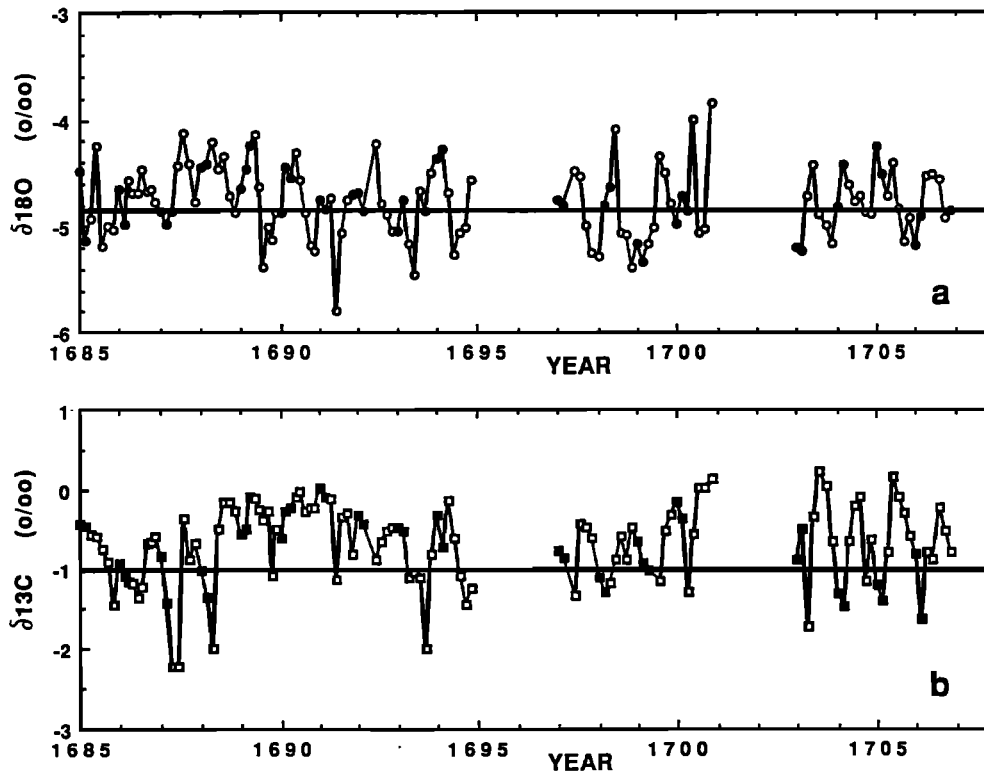
Fig. 4. (a) Records of $\delta^{18}O$ and (b) $\delta^{13}C$ for seasonal coral samples (7–9 per year) for selected periods during the late 1600s and early 1700s. Lines are drawn through the average δ values. The solid symbols represent high-density portions of the skeletal bands. Note that the very strong ENSO event of 1687–1688 is marked by high $\delta^{18}O$ and low $\delta^{13}C$ values.

ing that (1) SST and coral $\delta^{18}O$ correlate with the expected slope (0.22‰ per 1°C) and (2) salinity changes were minimal, these results suggest that the seasonal range in SST may have been significantly less during the turn of the eighteenth century than it is now.

Variability of $\delta^{13}C$ in corals is controlled by water composition ($\delta^{13}C$ of DIC) and by ambient light incident on the coral's growth surface [*McConnaughey*, 1989a,b]. Photosynthesis by the symbiotic alga zooxanthallae produces organic matter depleted in ^{13}C, leaving the DIC pool in the coral enriched in ^{13}C [*Fairbanks and Dodge*, 1979]. Indeed, $\delta^{13}C$ of coral aragonite has been observed to decrease linearly with depth (and reduced light) in the upper 15–20 m of the water column, at a rate of 0.09 $\pm$ 0.03‰ per m [*Fairbanks and Dodge*, 1979; *Weber et al.*, 1975; *Land et al.*, 1975]. As a result of upward growth over its 323-year lifetime, the growing surface of our coral changed from an average water depth of 8 m to 3 m. This would correspond to an increase in $\delta^{13}C$ of about 0.45‰, equal to the observed increase of 0.5‰ in the biannual $\delta^{13}C$ record (Figure 3b).

4.6. Correlations Among the Isotope Records

There is an apparent anomaly in the seasonal isotope data during 1687–1689, when $\delta^{18}O$ values remain higher than -4.9‰ and $\delta^{13}C$ values are lowest (Figure 4). This is coincident with the very strong ENSO event of 1687–1688 reported by *Quinn et al.* [1987]. The $\delta^{18}O$ data during 1687–1689 are high, as would be expected during periods of cooler SST in this region during ENSO. Also, the $\Delta^{14}C$ result for the 1686–1687 and 1688–1689 periods were low (-55.0 and -48.0‰, respectively) relative to surrounding values (Ta-

ble 1), as would be expected from a deeper and/or cooler water mass [*Burling and Garner*, 1959]. The remainder of the seasonal record examined for stable isotopes (Figure 4) had no concurrent ENSO events and showed no significant deviations from the norm. There was no visible correlation between the ENSO record of *Quinn et al.* [1987] and each of the biannual stable isotope records (Figure 3).

4.7. Spectral and Cross-Spectral Analyses

Periodicities within the individual isotope records were investigated using spectral and cross spectral analyses. Fourier transform of the correlation functions of each isotope record were done based on the Blackman-Tukey method [*Jenkins and Watts*, 1968]. We used the detrended $\delta^{13}C$, $\delta^{18}O$, and $\Delta^{14}C$ data from the biannual coral samples for the period A.D. 1635–1957. Negative values of detrended $\delta^{18}O$ were used, as there is an inverse correlation between $\delta^{18}O$ and $\Delta^{14}C$. Interpolation of the missing $\Delta^{14}C$ values was done by averaging the results from adjacent samples. The data were windowed in the time domain using a Hanning window with a width of 3 data points. The overlap of points was $n/3$, though changes from $n/4$ to $2n/3$ still revealed stable spectra. Because the isotope records changed over the 323-year timescale, we performed spectral analyses for the first (A.D. 1635–1795) and second (A.D. 1797–1957) halves of each of the records separately. Bandwidth was 0.0225 year^{-1}, that is, about 1 and 9 years at periods of 6 and 20 years, respectively.

The spectral density for the first half of the detrended $\Delta^{14}C$ (solid line in Figure 5a) exhibits variance at the 5-, 6-, 7-, 16-, and 27-year cycles that are significant to the 95%

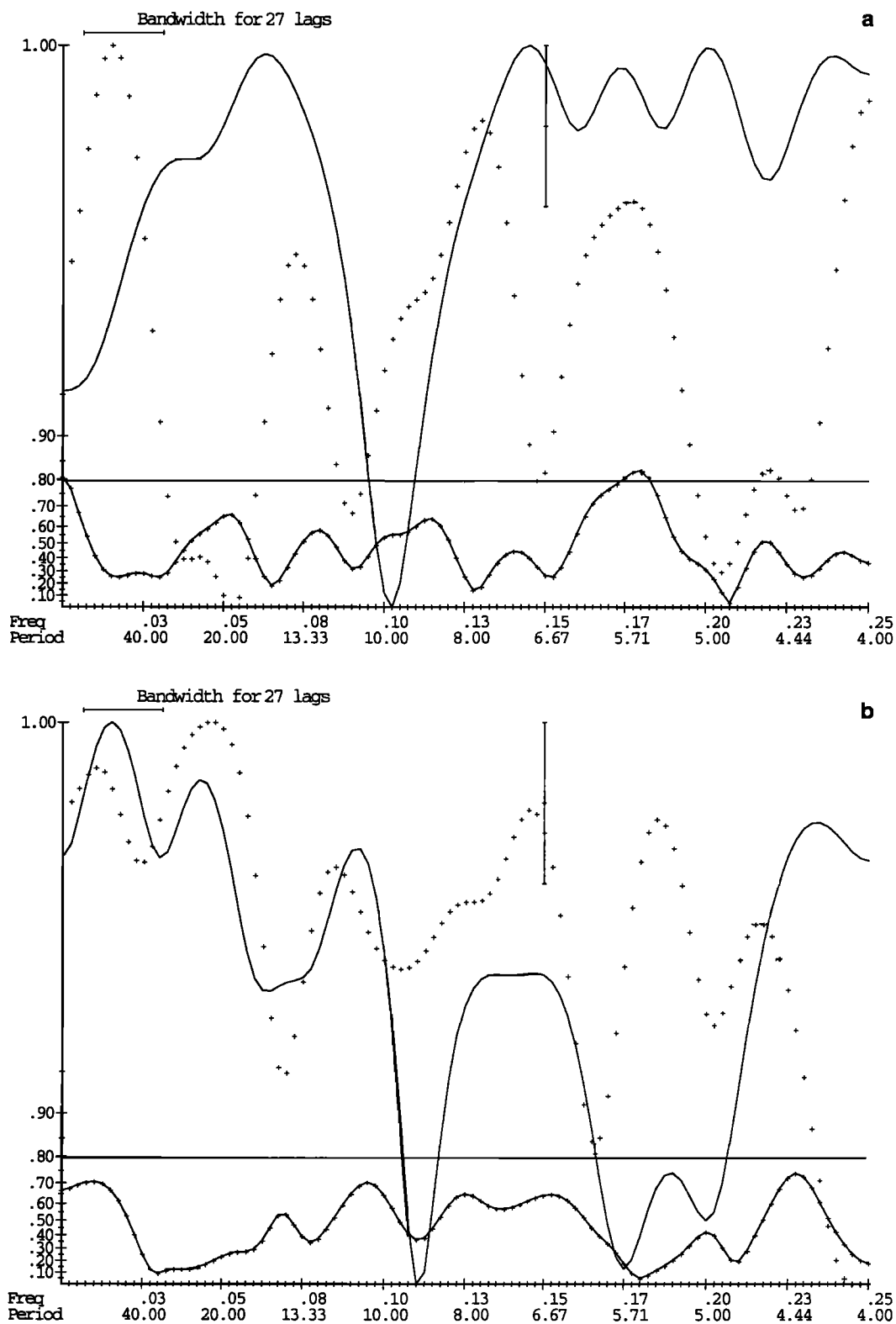Fig. 5. Cross spectral analyses for the (a) first (A.D. 1635–1795) and (b) second (A.D. 1797–1955) halves of the detrended Δ^{14}C and δ^{18}O records. The x axes are frequency (upper numbers in reciprocal years) and periods (lower numbers in years), and y axes are the power spectral density values (note log scale). Bottom of the vertical bar indicates the 95% confidence limit for variance of each isotopic record. Cycles less than 4.5 years are not significant, given the Nyquist frequency of 4 years. Cycles greater than about 40 years are also not significant, given that each record is only 162 years long. Coherency values are plotted in the lower third of each figure, with the 95% confidence level indicated by a horizontal line. See text for details.

confidence limit. Alternatively, spectral density for the second half of the detrended $\Delta^{14}C$ exhibits variance at longer cycles only, 11 and 24 years (Figure 5b, solid line). The spectral density for the first half of the detrended $\delta^{18}O$ record (pluses in Figure 5a) displays variance at the 6- and 8-year cycles that are significant to the 95% confidence limit. In the second half of the $\delta^{18}O$ record, spectral density displays significant variance at short and long cycles (5, 7, 12, 22 years). Thus, both the $\Delta^{14}C$ and $\delta^{18}O$ records display a higher amount of variance for the shorter cycles during the A.D. 1635–1795 period, than for the longer cycles. The opposite is true for the A.D. 1797–1957 period. Short cycles are observed in the ENSO record (E.R.M. Druffel, unpublished data, 1992), and it may be this phenomenon that dominates the variance in the isotope records early on.

In order to demonstrate similarities in the records, cross-spectral analyses were performed separately for early and late sections of the detrended $\Delta^{14}C$ and $\delta^{18}O$ data. For the first half of the records, the 6-year period was coherent to the 95% confidence level (Figure 5a, line with pluses); $\delta^{18}O$ lags $\Delta^{14}C$ by 1.0 ± 0.4 years. In the nineteenth and twentieth centuries, no periods displayed coherence to the 95% confidence level (Figure 5b). A 6-year cycle is also found in the spectral density of ENSO occurrences. The coherent 6-year cycle in the early part of the records suggests that a similar process, perhaps ENSO, controlled both isotopes, with $\Delta^{14}C$ changes preceding $\delta^{18}O$ changes. However, this coherency breaks down during the subsequent 2 centuries.

5. DISCUSSION

In order to determine the reasons why interannual $\Delta^{14}C$ variability was so high during the late A.D. 1600s and early 1700s, we first need to examine the controls on surface ocean DIC $\Delta^{14}C$. First, gas exchange of $^{14}CO_2$ between atmosphere and surface ocean occurs with about a 10-year equilibration time [Broecker and Peng, 1974], much longer than for other gases. Second, and by far the most important, is mixing with waters that contain different $\Delta^{14}C$ signatures. For example, the apparent reservoir "age" of nonpolar surface waters is about 400 ± 100 years. This "age" is due to mixing of subsurface waters, which have been depleted with respect to ^{14}C by isolation from the atmosphere, into the mixed layer. Variability in surface ocean $\Delta^{14}C$ can be achieved by mixing in two ways: (1) change in the depths of the mixed layer or the thermocline, or change in the rate of vertical mixing between surface (high $\Delta^{14}C$) and subsurface (low $\Delta^{14}C$) waters, and (2) large-scale advective (horizontal) changes in source waters that contain different $\Delta^{14}C$ signatures.

To help us model the vertical $\Delta^{14}C$ gradient necessary to have caused the changes observed during the late 1600s and early 1700s, we construct the following conceptual representation of the mixed layer and thermocline. A typical temperature profile of the west Coral Sea during winter consists of a temperature of 24°C in the mixed layer (75-m deep) and a decrease to 8°C by 600 m [Scully-Power and France, 1969a,b,c]. In summer, the mixed layer is shallower (~50 m) with an average SST of 28°–29°C. The only prebomb $\Delta^{14}C$ profile even remotely close to Abraham Reef was reported by Rafter [1968] from 21°S, 173°W, in March 1958; values were -40‰ at the surface, -52‰ at 140 m, and -80‰ by 580 m. Thus, an approximately monotonic decrease of

$\Delta^{14}C$ with depth (~6‰ per 100 m) can be estimated. Using a purely vertical model, water would need to originate from 200-m depth to decrease $\Delta^{14}C$ in the surface ocean from an average of -45‰ to -55‰. The thermocline has been observed to rise in the north Coral Sea during ENSO, sometimes by more than 50 m (G. Meyers, personal communication, 1992). A rise of this magnitude in the south Coral Sea would presumably lower the $\Delta^{14}C$ of the surface DIC somewhat. But it is difficult to imagine that water would be drawn exclusively from 200 m, unless very different water column structure and $\Delta^{14}C$ signature existed 300 years ago. It is difficult to compare a site 20° to the east with our south Coral Sea site. Unfortunately, these are the only prebomb $\Delta^{14}C$ values that are available.

It is equally difficult to explain the four high $\Delta^{14}C$ values of -35 to -30‰ during the Little Ice Age using alterations in vertical mixing alone. High $\Delta^{14}C$ carbon would need to originate from the atmosphere, where values averaged 15‰ in A.D. 1700 (Figure 1a). To raise $\Delta^{14}C$ of surface DIC from -45‰ to -33‰, one would need to exchange a 50-m mixed layer with CO_2 in the atmosphere for a period of 1 year and keep it isolated from mixing with subsurface waters; this assumes a 20 mol/m^2/yr exchange rate, and a [DIC] of 2 mol/m^3. Such conditions were not likely, especially during the Little Ice Age, when wind strength may have been stronger and mixing between surface and subsurface waters greater than they are today.

We hypothesize that both vertical and horizontal mixing changes need to be invoked to have caused the observed large shifts in $\Delta^{14}C$ during the late A.D. 1600s and early 1700s. Horizontal mixing processes do dominate this region. Pickard et al. [1977] report that no water types are formed in the Coral Sea; rather water from external sources flow into the area. Hydrographic data from the western Coral Sea adjacent to the Great Barrier Reef (14°–24°) shows that a westward flow dominates, bifurcating at 18°S, with a current flowing to the south through the Abraham Reef area throughout the year [Church, 1987] (see Figure 6). From temperature and salinity characteristics, Rochford [1968] determined that the two main westward flows into the Coral Sea were from the South Equatorial Current (SEC), and from the East Australian Current (EAC), which originates in the west central South Pacific [Pickard et al., 1977]. Coral data from Fanning Island (4°N, 159°W) [Druffel, 1987] indicate that the prebomb $\Delta^{14}C$ signature for the SEC was low, -60 ± 5‰, due to divergence along the equator; seawater DIC data indicate a prebomb $\Delta^{14}C$ signature for the EAC was -38 ± 5‰ [Rafter, 1968]. The average of the recent coral results (-49‰) falls in the middle of this range, as one would expect for a mixture of these two sources.

A change in advective (horizontal) mixing that would alter the $\Delta^{14}C$ of the surface ocean DIC $\Delta^{14}C$ at Abraham Reef involves the variable transport of SEC waters into the Coral Sea [Church, 1987]. Recent observations of salinity, temperature, and sea level height during the 1982–1983 ENSO suggest that the center of the subtropical gyre in the South Pacific shifted southward [Meyers and Donoguy, 1984; Wyrtki, 1984]. The westward SEC flowed in the 10°–20°S latitude band during the 1982–1983 ENSO, much further south than during normal years (0°–10°S). This would bring low $\Delta^{14}C$ surface waters from the SEC into the Coral Sea during periods of ENSO and could provide for end-member mixing that would result in an overall lower $\Delta^{14}C$ value than
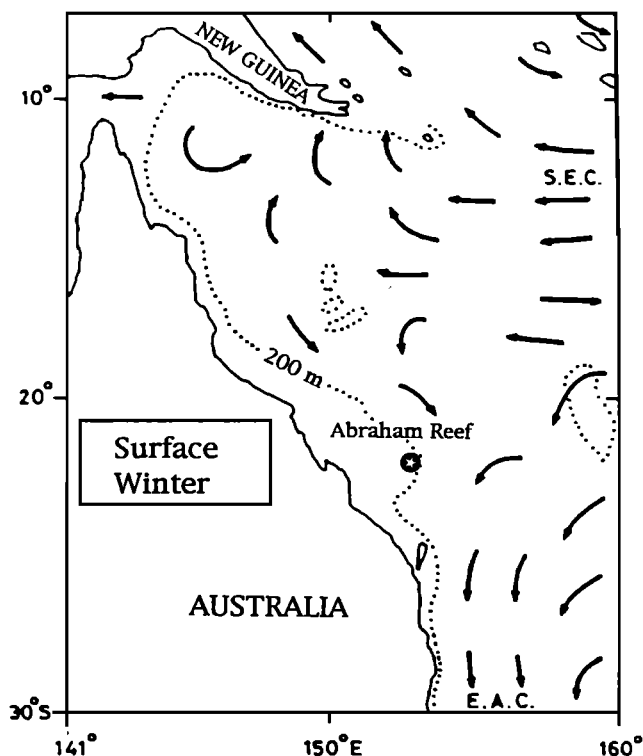
Fig. 6. Mean circulation of surface currents in the Coral Sea and area further south during winter (adapted from *Pickard et al.* [1977, Figure 63]).

during non-ENSO years. It is not known if these excursions occurred on longer timescales (decades to centuries) owing to the insufficient sampling time. However, isotopic data presented here, namely (1) the large variability of $\Delta^{14}C$ noticed only from A.D. 1680 to 1730, (2) the presumably warmer, more variable SST present in the last 100 years, and (3) the absent Suess effect, all universally point to decade to century-timescale variability of circulation and/or mixing in the southwestern Pacific.

Toggweiler et al. [1991] proposed that the large-scale overturning of South Pacific waters with subantarctic water might explain the imperceptible Suess effect in the south. They describe a single, steady state process of thermocline ventilation involving successively less dense features of the South Pacific, that evolve from each other via diapycnal alteration, but leaving the $\Delta^{14}C$ signature of this water mass unchanged. Our data neither substantiate nor repudiate this hypothesis. However, it is clear that a model of South Pacific thermocline circulation would need to resolve the decade timescale changes in $\delta^{18}O$ and $\Delta^{14}C$ that have been presented here.

We hypothesize that there were long-term changes in the circulation of the south Coral Sea and that this could have been achieved by one or a combination of the following: (1) change in the duration and/or intensity of the southerly excursions of the southern subtropical gyre, thereby directing low $\Delta^{14}C$, SEC waters into the south Coral Sea, (2) change in the depth of the mixed layer or the thermocline, and (3) change in the rate of mixing between surface and subsurface water masses within the Coral Sea. The isotopic records show that there has been a long-term change in the nature of ENSO over the past 4 centuries in the southwestern Pacific. Steady state conditions with respect to circulation appear not to have been maintained within the last century

or so, and we hypothesize that this is the main cause for an absent Suess effect in the Abraham Reef coral.

To specifically identify the relative magnitude of vertical and advective changes in circulation that occurred in the past, future studies need to include $\Delta^{14}C$ and stable isotope records from corals collected along the western boundary of the tropical Pacific. Seasonal variability of $\Delta^{14}C$ needs to be examined, so that changes in the water mass surrounding the coral on short timescales can be assessed. Prebomb $\Delta^{14}C$ measurements, or estimates based on bomb ^{14}C distributions, of thermocline waters from the surrounding regions are essential. Proxies of nutrient levels in these waters, for example, Cd/Ca ratios in corals from Abrahams Reef [*Delaney and Linn*, unpublished data], will also provide a method of deconvolving vertical and advective (horizontal) changes in circulation.

6. CONCLUSIONS

Large interannual variations of surface ocean $\Delta^{14}C$ were recorded in annually banded corals from the southern Great Barrier Reef. Low $\Delta^{14}C$ values coincide with recorded strong ENSO events, suggesting a shoaling of the thermocline or advection of ^{14}C-depleted source waters to this location during ENSO. Our results show that during the late 1600s and early 1700s, the amplitude of DIC $\Delta^{14}C$ excursions were about 3 times larger than those during the nineteenth and twentieth centuries. This suggests a long-term change in the nature and manifestation of ENSO events, as it is reflected in the ^{14}C cycle in the southwestern Pacific.

The isotope data also point to an anomolous period from 1875 to 1920. There was no correlation between low $\Delta^{14}C$ and ENSO events. This suggests that warmer, higher $\Delta^{14}C$ waters predominated this region for 5 decades. This could have provided the necessary mask to disguise the Suess effect that was anticipated, had these waters been in steady state with surrounding water bodies.

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