

TROPICAL SOUTH CHINA SEA SURFACE ^{14}C RECORD IN AN ANNUALLY-BANDED CORAL

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ABSTRACT. A surface-water $\Delta^{14}C$ record of AD 1948–1999 in the tropical South China Sea (SCS) has been reconstructed from accelerator mass spectrometric radiocarbon measurements of annual bands of a *Porites* coral collected from Con Dao Island, Vietnam. Results gave the following $\Delta^{14}C$ time series: a steady state of –47.8 ± 2.8‰ (mean ± SD, $n = 8$) during 1948–1955 (i.e. in the pre-bomb period); a sharp increase during 1956–1966; a gradual increase during 1967–1973; a relatively high maximum value of ~174‰ in 1973; and a gradual decrease for the following period to ~86‰ in 1999. This $\Delta^{14}C$ record having a sharp increase and a relatively high peak is similar to the records of subtropical corals (latitudes 21–27°) and is distinctly different from the records of equatorial/tropical corals (latitudes <10°), although our coral sample was collected from an equatorial/tropical region (8°39′N, 106°33′E). This can be explained by the geographic, oceanographic, and climatic setting of our study site. The SCS is a semi-enclosed marginal sea in the far western tropical Pacific and is little influenced by equatorial upwelling or related ocean currents. Our study site is located in the southwestern SCS, where an enormous submerged plain (the Sunda Shelf) spreads out with very shallow waters (mean depth <100 m). Furthermore, in the SCS, the East Asian monsoon (a strong, seasonally reversing wind system) enhances air-sea gas exchange especially in the mainland coastal waters, including our study site. Such semi-enclosed shallow waters with enhanced ventilation were probably very sensitive to the atmospheric nuclear explosions in the late 1950s and early 1960s and caused the sharp increase and high peak in the coral $\Delta^{14}C$ record. Our coral $\Delta^{14}C$ values in the southwestern SCS are significantly higher than the values in the northwestern SCS (Xisha Islands), which seems to suggest that meridional mixing of surface waters is not active in the SCS and that the open-ocean water intruding into the northern SCS (i.e. the Kuroshio intrusion) has only a limited influence on the southern SCS.

INTRODUCTION

Massive coral species living in the tropical/subtropical shallow waters form annual growth bands in their calcareous skeletons (aragonite). To visualize the annual bands, a massive coral sample is sliced to a thickness of several millimeters and photographed under long-wavelength UV light (e.g. ~350 nm) or X-rayed (e.g. Knutson et al. 1972; Buddemeier et al. 1974; Isdale 1984; Barnes and Taylor 2001). Massive corals occasionally grow up to several meters in height with annual bands of hundreds of years. Core samples drilled from such long-lived massive corals are useful for reconstructing the history of sea-surface environment/climate.

Corals secrete their skeletons using dissolved inorganic carbon (DIC), calcium, and many other elements of the ambient seawater. Therefore, radiocarbon measurements of coral annual bands provide time series of the ^{14}C concentration ($\Delta^{14}C$) of DIC in sea-surface water (Druffel 1997). ^{14}C is constantly produced naturally in the stratosphere and was also produced artificially as a result of atmospheric nuclear explosions conducted mainly in the late 1950s and early 1960s. The produced ^{14}C is quickly combined with oxygen to form $^{14}CO_2$ and incorporated into the ocean, soil, vegetation, and so on. The atmospheric nuclear explosions almost doubled the natural atmospheric ^{14}C concentration (the $\Delta^{14}C$ peaks of ~1000‰ in the Northern Hemisphere in 1964 and ~700‰ in the Southern Hemisphere in 1965; Nydal and Lövseth 1983; Levin et al. 1985, 1994; Manning and Melhuish 1994), breaking the quasi-equilibrium of global ^{14}C distribution and leading to a great excess of ^{14}C

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in the surface ocean relative to the deeper ocean. The bomb-induced ocean-surface ^{14}C excess provided a good opportunity to investigate the world's ocean circulations and ventilations. Recent 50-yr coral $\Delta^{14}C$ records show responses to the nuclear explosions, which vary with regional oceanographic conditions such as water mass movement and oceanic ventilation. Such $\Delta^{14}C$ response curves (i.e. post-bomb coral $\Delta^{14}C$ data) contribute to our understanding of ocean circulations and also can be used for testing and improvements of ocean circulation models. Post-bomb coral $\Delta^{14}C$ data have been accumulated in the open seas of the Pacific, Atlantic, and Indian oceans but scarcely been done in semi-enclosed marginal seas such as the South China Sea, Caribbean Sea, and Gulf of Mexico.

In this study, we present a 52-yr-long $\Delta^{14}C$ record (AD 1948–1999) of an annually-banded coral collected from the southwestern South China Sea and discuss the oceanographic environments and surface-water circulations around that region.

SAMPLE COLLECTION AND GEOGRAPHIC/OCEANOGRAPHIC SETTING

In May 2000, a 68-cm-long core (5.5 cm in diameter) was vertically drilled from the top of a coral colony (*Porites* sp.: ~80 cm in height) living at 5–6 m water depth on the western side of Con Dao Island (8°39′36″N, 106°33′07″E), Vietnam. This island is located in the southwestern part of the South China Sea (SCS), about 90 km from the mouth of the Mekong River (Figure 1). The SCS located in the far western tropical Pacific is a semi-enclosed marginal sea with an area of ~3.6 million km^2 surrounded by the Southeast Asian mainland and the Malay Archipelago. The southwestern SCS where Con Dao Island is located is very shallow (mean depth <100 m), as a part of the Sunda Shelf, an enormous submerged plain stretching from the Java Sea. In contrast, the northeastern SCS is a deeper basin with a maximum depth of >4500 m.

The SCS is known to be a region of strong seasonal reversals in surface circulation driven by the East Asian monsoon: southwesterly winds (warm and humid) in boreal summer and northeasterly winds (cool and dry) in boreal winter. In general, the winter northeasterly winds cause southward and southwestward currents along the mainland coast, and these currents reverse under the summer southwesterly winds. The SCS has 7 straits leading into the East China Sea (the Taiwan Strait), the Philippine Sea (the Luzon Strait), the Sulu Sea (the Mindoro Strait and the Balabac Strait), the Java Sea (the Karimata Strait and the Gaspar Strait), and the Andaman Sea (the Strait of Malacca) (Figure 1). The Luzon Strait, having a width of ~350 km and a sill depth of ~2500 m, is much larger than the other 6 straits and connects the SCS directly to the open ocean. Through the Luzon Strait, the Kuroshio Current, which is the western boundary current originating from the North Equatorial Current, partly intrudes into the northeastern SCS (Figure 1) (e.g. Nitani 1972; Farris and Wimbush 1996; Metzger and Hurlburt 1996). The Kuroshio intrusion is considered the main inflow of open-ocean water to the SCS. The Mindanao Current, also originating from the North Equatorial Current, flows southward on the eastern side of the Philippines and partly passes through the archipelago and finally into the Indian Ocean, which is called the Indonesian Throughflow (Figure 1). The Indonesian Throughflow scarcely intrudes into the SCS via the Karimata/Gaspar straits.

METHODS

The coral core was sectioned, along its longitudinal axis, into 6-mm-thick slabs. The slabs were cleaned with distilled water in an ultrasonic bath, dried for a week, photographed under long-wavelength UV light (~352 nm), and X-rayed. The UV photo and X-ray photo revealed luminescent banding and density banding in the coral, respectively. The total number of the banding patterns in the UV photo is 52, which is the same as that in the X-ray photo. Those banding patterns are 10–12

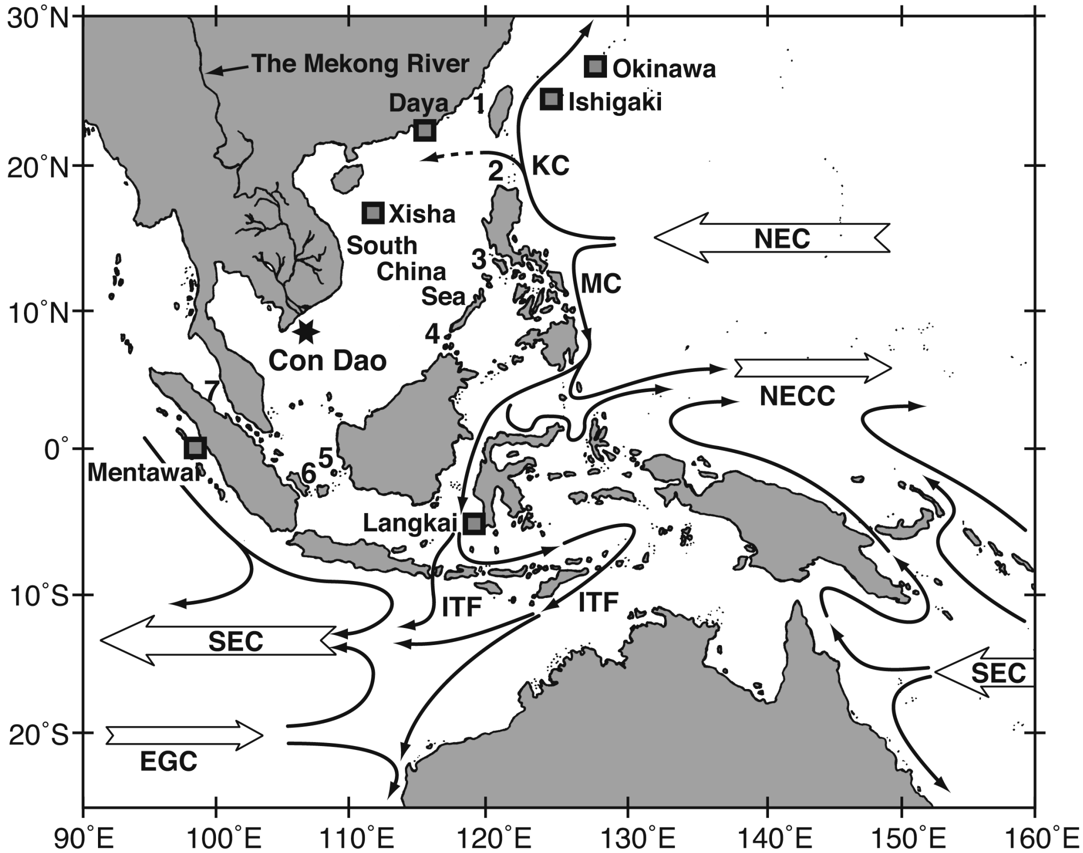

Figure 1 Map of the Southeast Asian archipelago showing the major near-surface currents around the region: NEC: North Equatorial Current; NECC: North Equatorial Countercurrent; KC: Kuroshio Current; MC: Mindanao Current; ITF: Indonesian Throughflow; SEC: South Equatorial Current; EGC: Eastern Gyral Current. The surface-water circulations in the South China Sea vary seasonally according to the East Asian monsoon characterized by southwesterly winds in boreal summer and northeasterly winds in boreal winter. In this study, a Δ^{14}C record of AD 1948–1999 has been obtained from an annually-banded coral sample collected from Con Dao Island in the southwestern South China Sea (indicated as a star). Con Dao Island is ~90 km from the mouth of the Mekong River. Seven straits of the South China Sea are indicated as numbers (1: Taiwan Strait, 2: Luzon Strait, 3: Mindoro Strait, 4: Balabac Strait, 5: Karimata Strait, 6: Gaspar Strait, 7: Strait of Malacca). Shaded boxes indicate the locations of previously published coral Δ^{14}C records in Southeast Asia. The records from Okinawa Island, Ishigaki Island, Xisha Islands, Langkai Island, and the Mentawai Islands are shown in Figure 3 or 4 for comparison with the Con Dao coral record.

mm in width, which is a typical annual growth rate of *Porites* coral skeletons. Thus, it was assumed that both the luminescent and density banding patterns represent annual growth bands of AD 1948–1999. This assumption has been confirmed by Sr/Ca measurements (i.e. a proxy for sea-surface temperature) along the coral's growth direction at 1-mm increments (T Mitsuguchi, PX Dang, H Kitagawa, T Uchida, Y Shibata, unpublished data). The luminescent banding was used as a guideline for taking subsamples from the coral slabs, because it was more clear-cut than the density banding. Comparison of the luminescent banding and the Sr/Ca fluctuation indicates that a bright luminescent line is formed around October every year. Since Con Dao Island is close to the mouth of the Mekong River, organic content of the coral sample may be relatively high and may become a contaminant in our ^{14}C analyses. In order to remove the possible organic contamination, the coral slabs were subjected to a sequence of chemical treatments with 5% H_2O_2 aqueous solution, 0.5M NaOH aqueous solution, and Milli-Q water (see Dang et al. [2004] for details).

The 52 annual growth increments of 1948–1999 were subsampled one by one from a bright luminescent line to the next by scraping the slab's surface (~2 mm in depth and ~5 mm in width) with an engraving tool. The 52 subsamples were crushed and homogenized one by one in an agate mortar. Dang et al. (2004) reported $\Delta^{14}C$ measurements for the 12 subsamples of 1949–1960, where each subsample was dissolved in 85% phosphoric acid to extract CO_2. Here, we accomplish $\Delta^{14}C$ measurements of all the 52 subsamples using another CO_2 extraction technique that decomposes coral carbonate by combustion. Prior to the combustion process, the 52 subsamples were treated with 4 mL of 15% H_2O_2 aqueous solution at 70 °C for 15 min under ultrasonic agitation, rinsed with 4 mL of Milli-Q water repeatedly, and then heated at 450 °C for 4 hr for further removal of possible organic contamination. About 9 mg of each subsample was put into a tin capsule and then burned at 950 °C in a combustion furnace that is free from atmospheric-CO_2 contamination. The extracted CO_2 was selectively trapped and purified using a chemical trap and cryogenic traps. In these CO_2 extraction/purification processes, we used a Vario EL III elemental analyzer (Elementar Analysensysteme GmbH, Germany) that had been modified for sample preparation for accelerator mass-spectrometric (AMS) ^{14}C measurement in the National Institute for Environmental Studies (NIES), Tsukuba, Japan (see Yoneda et al. [2004] for details). The purified CO_2 was converted to graphite on an iron powder catalyst (1.0 mg) at ~650 °C with H_2 gas as the reducing agent. The graphite targets were measured for $^{14}C/^{12}C$ and $^{13}C/^{12}C$ ratios using the AMS facilities in NIES named NIES-TERRA (Tandem accelerator for Environmental Research and Radiocarbon Analysis). Results are reported as $\Delta^{14}C$ (‰) for all the subsamples and also as a conventional ^{14}C age (BP) for the pre-bomb subsamples of 1948–1955, according to the definition by Stuiver and Polach (1977). The $\Delta^{14}C$ values were calculated using the absolute international standard radioactivity and the subsamples radioactivity corrected for decay between the years of measurement and coral growth and also for isotope fractionation to a $\delta^{13}C$ value of –25‰ (relative to VPDB).

RESULTS

As shown in Figure 2, we first compared $\Delta^{14}C$ results between the 2 different techniques in CO_2 extraction for the 12 subsamples of 1949–1960: by combustion at 950 °C (this study) and by dissolution in 85% phosphoric acid (Dang et al. 2004). In both cases, $\Delta^{14}C$ data were obtained from the AMS facilities NIES-TERRA. The comparison shows good agreement between the 2 techniques with 70% confidence intervals overlapping for 10 data points and 95% confidence intervals overlapping for all the 12 data points. As a result of an increase in atmospheric nuclear explosions, the $\Delta^{14}C$ value starts to increase from 1956. Thus, the boundary between the pre- and post-bomb periods can be placed between the data of 1955 and 1956.

Dang et al. (2004) calculated a regional correction of the globally averaged ^{14}C marine reservoir age (i.e. a ΔR value) for the southwestern SCS using their pre-bomb data of 1949–1955. In this study, a $\Delta^{14}C$ data of 1948 has been newly added to the pre-bomb data set (see Figure 2). We calculate the ΔR value using our pre-bomb data of 1948–1955 (Table 1). Our calculation gives $\Delta R = -81 \pm 23$ yr (mean ± SD, $n = 8$) for the southwestern SCS according to the calculation of Dang et al. (2004), who used the model-based, globally averaged marine ^{14}C age of 473 BP of 1950 (Stuiver et al. 1998a,b). As expected, our calculation is indistinguishable from the calculation of Dang et al. (2004): $\Delta R = -74 \pm 39$ yr (mean ± SD, $n = 7$).

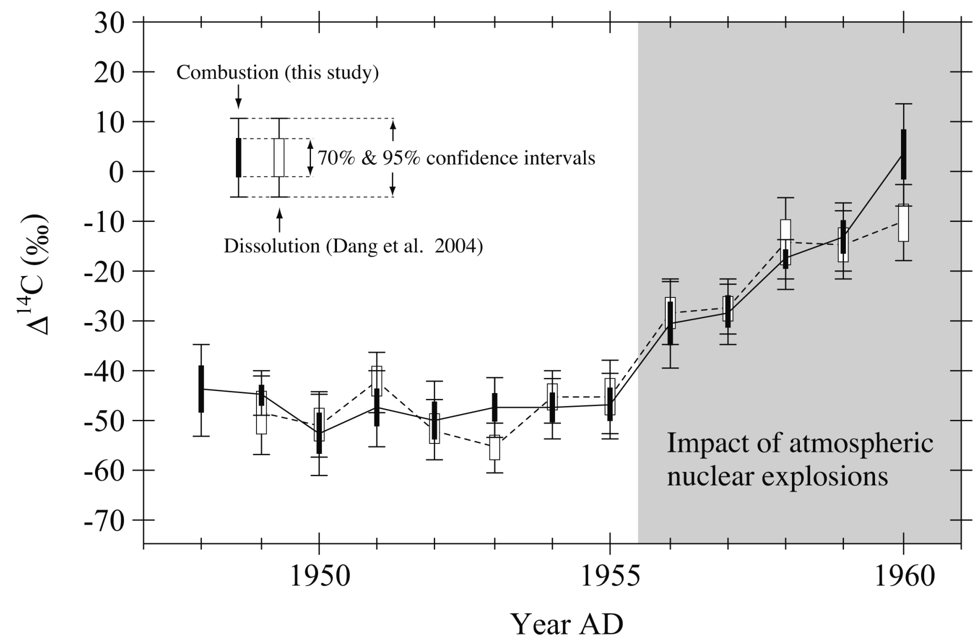

Figure 2 Comparison of $\Delta^{14}C$ results between 2 different techniques in CO_2 extraction for 12 annual growth increments (AD 1949–1960) in a coral sample from Con Dao Island, Vietnam. One is by combustion at 950 °C (this study) and the other is by dissolution in 85% phosphoric acid (Dang et al. 2004). In both cases, $\Delta^{14}C$ data were obtained by use of the AMS facilities NIES-TERRA. Results of the 2 techniques agree well with each other, with 70% confidence intervals overlapping for 10 data points and 95% confidence intervals overlapping for all the 12 data points. As a result of an increase in atmospheric nuclear explosions, the $\Delta^{14}C$ value starts to increase from 1956.

Table 1 Pre-bomb ^{14}C data (AD 1948–1955) of an annually-banded coral sample from Con Dao Island in the south of Vietnam, and calculation of ΔR values.

Year (AD)	$\Delta^{14}C$ (‰)	±1 σ (‰)	^{14}C age (BP)	±1 σ (yr)	Model age (BP)	ΔR (yr)
1948.3	–44.0	4.7	363	39	473	–110
1949.3	–45.1	3.0	372	25	473	–101
1950.3	–53.0	4.1	438	35	473	–35
1951.3	–47.6	3.8	391	32	473	–82
1952.3	–50.1	4.0	411	33	473	–62
1953.3	–47.5	2.7	388	23	473	–85
1954.3	–47.8	3.1	389	26	473	–84
1955.3	–47.2	3.3	384	28	473	–89
Average	*–47.8*	*3.6*	*392*	*30*	*473*	*–81*
S.D.	2.8	—	23	—	—	23

The post-bomb $\Delta^{14}C$ data (1956–1999) obtained in this study are listed in Table 2. Combined with the pre-bomb data, the overall description of the $\Delta^{14}C$ time series is as follows: a steady state of –47.8 ± 2.8‰ (mean ± SD, n = 8) during the pre-bomb period; a sharp increase during 1956–1966; a gradual increase during 1967–1973; a maximum value of ~174‰ in 1973; and a gradual decrease for the following period to ~86‰ in 1999.

Table 2 Post-bomb ^{14}C data (AD 1956–1999) of an annually-banded coral sample from Con Dao Island in the south of Vietnam.

Year (AD)	$\Delta^{14}C$ (‰)	±1 σ (‰)	Year (AD)	$\Delta^{14}C$ (‰)	±1 σ (‰)
1956.3	–30.7	4.4	1978.3	163.2	3.4
1957.3	–28.4	3.3	1979.3	158.4	4.2
1958.3	–17.6	3.0	1980.3	155.7	4.1
1959.3	–13.2	3.5	1981.3	154.9	4.7
1960.3	3.5	5.1	1982.3	151.1	3.4
1961.3	16.0	3.5	1983.3	151.6	5.0
1962.3	27.1	3.2	1984.3	141.4	5.3
1963.3	47.6	3.3	1985.3	136.8	4.7
1964.3	84.6	3.0	1986.3	132.7	5.7
1965.3	127.3	3.8	1987.3	123.6	6.5
1966.3	149.8	3.7	1988.3	118.1	3.7
1967.3	156.3	3.1	1989.3	110.7	4.5
1968.3	162.6	3.3	1990.3	116.1	3.2
1969.3	164.6	3.5	1991.3	109.0	4.0
1970.3	163.6	5.6	1992.3	105.9	4.9
1971.3	169.9	4.6	1993.3	100.1	5.1
1972.3	167.5	4.3	1994.3	99.7	4.9
1973.3	173.8	3.4	1995.3	93.1	3.4
1974.3	173.1	4.7	1996.3	93.8	4.9
1975.3	169.9	3.7	1997.3	87.6	3.1
1976.3	169.5	3.0	1998.3	89.7	3.5
1977.3	166.8	4.9	1999.3	86.0	3.8

DISCUSSION

Previously reported coral $\Delta^{14}C$ records in the SCS are very limited and give incomplete post-bomb data: 1955–1958 in Xisha Islands (Southon et al. 2002); 1956–1960 in Con Dao Island (Dang et al. 2004); and 1977–1998 in Daya Bay (Shen et al. 2004). The Xisha Islands and Daya Bay are located in the northern SCS, and Con Dao Island is in the southern SCS (Figure 1). For the period after 1984, the $\Delta^{14}C$ record in Daya Bay frequently exceeds the atmospheric $\Delta^{14}C$ value. This anomalous data may be partly related to the fact that the coral sample was collected only ~1.5 km from the thermal effluent outfall of the Daya Bay Nuclear Power Station, the construction of which started in 1987 and the commercial operation in 1994. In any case, the Daya Bay record seems to reflect some local effect and to be unusable for oceanographic discussions.

Our coral $\Delta^{14}C$ record in Con Dao Island is the first, in the SCS, to cover the whole post-bomb period within the 20th century. The Con Dao coral $\Delta^{14}C$ record has been compared with other coral $\Delta^{14}C$ records obtained in latitudes <10° (Figure 3) (Druffel 1981; Moore et al. 1997; Guilderson and Schrag 1998; Guilderson et al. 1998, 2004; Grumet et al. 2004; Schmidt et al. 2004) and in latitudes 16–27° (Figure 4) (Konishi et al. 1982; Druffel 1987; Toggweiler et al. 1991; Druffel and Griffin 1995; Guilderson et al. 2000; Southon et al. 2002; Mitsuguchi et al. 2004). Some of the $\Delta^{14}C$ records shown in Figures 3 and 4 are derived from Southeast Asia (i.e. Con Dao Island, Mentawai Islands, Langkai Island, Okinawa Island, Ishigaki Island, and Xisha Islands); their locations are indicated in Figure 1. Although from an equatorial/tropical region (8°39′N, 106°33′E), the Con Dao coral record is distinctly different from the records of other equatorial/tropical corals (Figure 3) and is similar to the records of subtropical corals (i.e. Okinawa, Ishigaki, French Frigate Shoals, and Rarotonga in latitudes 21–27°) (Figure 4). Owing to equatorial upwelling and related ocean currents, the post-bomb $\Delta^{14}C$ records of equatorial/tropical corals generally show gentler increases and lower, delayed

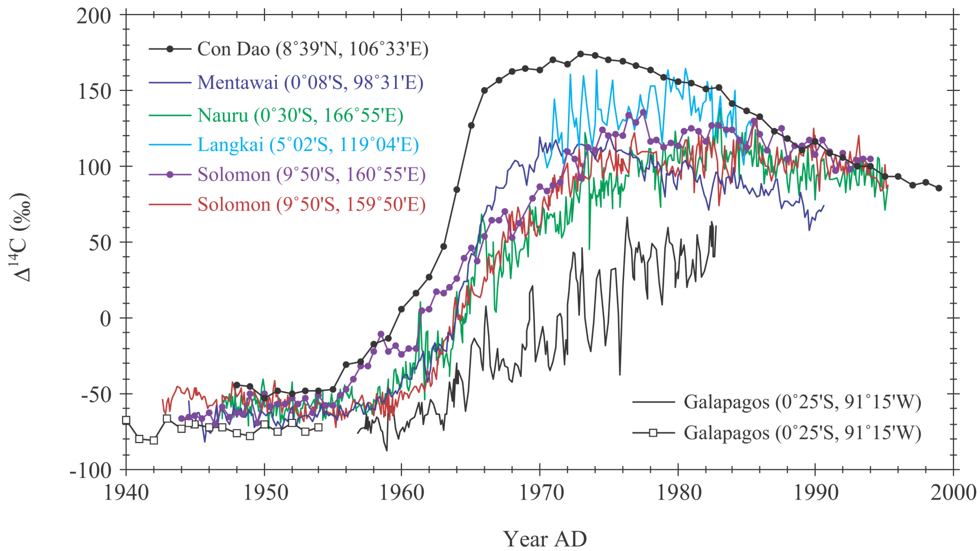

Figure 3 Comparison of a tropical South China Sea coral Δ^{14}C record from Con Dao Island (8°39′N, 106°33′E; this study) with coral Δ^{14}C records from a latitude range of <10°: Mentawai Islands (Grumet et al. 2004), Nauru Island (Guilderson et al. 1998), Langkai Island (Moore et al. 1997), Solomon Islands (Guilderson et al. 2004; Schmidt et al. 2004), and Galapagos Islands (Druffel 1981; Guilderson and Schrag 1998).

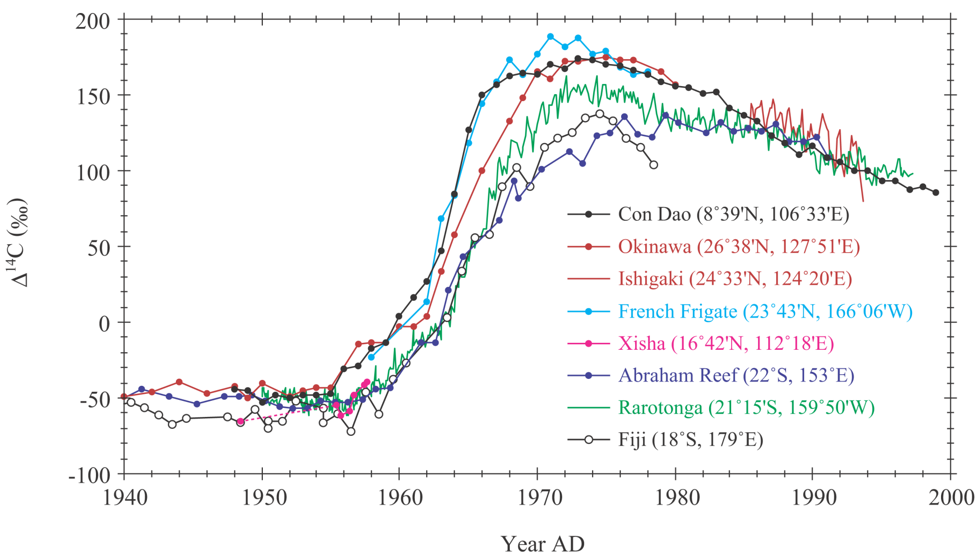

Figure 4 Comparison of a tropical South China Sea coral Δ^{14}C record from Con Dao Island (8°39′N, 106°33′E; this study) with coral Δ^{14}C records from a latitude range of 16–27°: Okinawa Island (Konishi et al. 1982), Ishigaki Island (Mitsuguchi et al. 2004), French Frigate Shoals (Druffel 1987), Xisha Islands (Southon et al. 2002), Abraham Reef (Druffel and Griffin 1995), Rarotonga Island (Guilderson et al. 2000), and Fiji Islands (Toggweiler et al. 1991).

peaks compared with those of subtropical corals. However, the Con Dao coral record shows one of the earliest, sharpest, and largest increases in response to the atmospheric nuclear explosions. This can be explained by the geographic, oceanographic, and climatic setting of our study site, which is itemized as follows:

- The SCS is a semi-enclosed marginal sea in the far western tropical Pacific and is generally little influenced by the Pacific equatorial upwelling system or related ocean currents.
- Con Dao Island lies on the Sunda Shelf, which is an enormous submerged plain stretching from the Java Sea to the southwestern SCS with very shallow waters (mean depth <100 m).
- In the SCS, the East Asian monsoon enhances air-sea gas exchange especially in the mainland coastal waters, including Con Dao Island.

Such semi-enclosed shallow waters with enhanced ventilation were probably very sensitive to the atmospheric nuclear explosions in the late 1950s and early 1960s and caused the sharp increase and high peak of $\Delta^{14}C$ in the Con Dao coral record.

For the equatorial/tropical $\Delta^{14}C$ records shown in Figure 3, the record in Langkai Island is the nearest to the record in Con Dao Island, although annual averages of the Langkai data are ~40‰ lower than the Con Dao data during the early to middle 1970s (Figure 4). The Langkai record has an annually averaged peak of ~155‰ in 1979–1980, while the peak of the Con Dao record (~174‰) occurs in 1973. These results are explained by the similarity and difference in geographic and oceanographic setting between the 2 sites. Although both Langkai Island and Con Dao Island are located inside the Southeast Asian archipelago, Langkai Island lies outside the SCS and just in the path of the Indonesian Throughflow that originates from the North Equatorial Current and scarcely flows to the Karimata/Gaspar straits (Figure 1).

The main flow of open-ocean water into the SCS probably occurs in the Luzon Strait, which is known as the Kuroshio intrusion (Figure 1). The Kuroshio Current is the western boundary current, which originates from the North Equatorial Current and passes the Luzon Strait, Ishigaki Island, and Okinawa Island. Since the Luzon Strait (~20°N, ~122°E) is ~1000 km upstream from Okinawa Island (~27°N, ~128°E), the surface water intruding through the Luzon Strait presumably has a lower $\Delta^{14}C$ value than the surface water around Okinawa Island. The coral $\Delta^{14}C$ data in the Xisha Islands, northwestern SCS (available for the years 1948, 1955–1958) are 11–27‰ lower than those in Okinawa Island (Figure 4), which may suggest that the intruded Kuroshio water reaches the Xisha Islands. On the other hand, the coral $\Delta^{14}C$ data in Con Dao Island are 8–28‰ higher than those in the Xisha Islands and similar to the data in Okinawa Island before 1960. After 1960, the Con Dao record shows a steeper increase with higher $\Delta^{14}C$ values compared with the Okinawa record. These results seem to suggest that meridional mixing of surface waters is not active in the SCS and that the influence of the Kuroshio intrusion on the southern SCS is significantly limited. Thus, we conclude that the distinctive $\Delta^{14}C$ record in Con Dao Island is primarily due to monsoon-induced active air-sea gas exchange in the semi-enclosed shallow waters.

Qu et al. (2005) inferred a surface-water circulation between the Pacific and the SCS on the basis of wind data and a high-resolution general circulation model. The inferred circulation starts as the Kuroshio intrusion through the Luzon Strait, runs through the SCS as a southward flow, enters the Java Sea through the Karimata/Gaspar straits, and finally returns to the Pacific through the Makassar Strait. Numerical models using the coral $\Delta^{14}C$ records in Con Dao Island, Xisha Islands, Langkai Island, and Okinawa/Ishigaki Islands will contribute to understanding the ocean circulation of the SCS as well as examining the inference of Qu et al. (2005), although additional long coral records in this region would be a great help.

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