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SEASONAL RADIOCARBON VARIATION OF SURFACE SEAWATER RECORDED IN A CORAL FROM KIKAI ISLAND, SUBTROPICAL NORTHWESTERN PACIFIC

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ABSTRACT. A coral radiocarbon ($\Delta^{14}C$) investigation with a high time-resolution is crucial for reconstructing secular and seasonal $\Delta^{14}C$ changes in the surface seawater which potentially reflect ocean circulations and dynamic ocean-atmosphere interactions. The $\Delta^{14}C$ values of a modern coral (*Porites* sp.) from Kikai Island, southern Japan, in the subtropical northwestern Pacific, were determined for the period of 1991–1998 at a monthly resolution. A coral $\Delta^{14}C$ time series for the 8 yr indicated seasonal cycles superimposed on a secular decreasing trend of 3.8‰ per yr. The seasonal amplitude of the coral $\Delta^{14}C$ was about 18‰ on the average, and the minimum $\Delta^{14}C$ was observed in late spring and summer. The $\Delta^{14}C$ changes were tentatively explained by horizontal oceanic advections around Kikai Island or over the wide range of the equatorial and subequatorial Pacific.

INTRODUCTION

The radiocarbon concentration (represented by $\Delta^{14}C$, which includes a normalization for mass dependent fractionation and decay) of the dissolved inorganic carbon (DIC) in seawater is influenced by several oceanographical factors; these factors include air-sea CO_2 exchange and vertical and horizontal water-body mixing. Investigating seasonal and secular $\Delta^{14}C$ changes of the DIC in the surface water provides valuable information on ocean circumstances.

The coral annual bands can record the past $\Delta^{14}C$ changes of DIC of the surrounding surface seawater during the skeletal accretion. Some massive-type corals living in shallow seawater grow for tens to hundreds of years at rates of more than 1 cm per yr while making annual skeletal density bands. Thus, corals can provide long-term histories with an exact time axis, as opposed to the snapshot views of $\Delta^{14}C$ distributions that accrue from occasional observations made by big projects such as the Geochemical Oceans Sections Study (GEOSECS) and the World Ocean Circulation Experiment (WOCE). ^{14}C measurements across coral density bands have revealed the lowering of $\Delta^{14}C$ at the ocean surface from the late 1800s through to 1955, which was mostly due to the uptake of anthropogenic ^{14}C-free CO_2 emission to the atmosphere by burning of fossil fuels, as well as a sudden increase in the atmospheric testing of nuclear weapons in the 1950s and 1960s (Druffel and Linick 1978; Nozaki et al. 1978). Recent investigations on the coral $\Delta^{14}C$ variations with sub-annual resolution give a new insight of intra-annual decadal variability of the ocean circulations in surface and subsurface water and air-sea CO_2 exchange (Druffel 1989; Guilderson et al. 1998). Based on high, dense ^{14}C measurements of coral bands from Kikai Island, we report here on seasonal and secular $\Delta^{14}C$ variations during 1991–1998 in the subequatorial northwestern Pacific.

SAMPLES AND ANALYTICAL METHODS

Kikai Island (28°16′ to 28°22′N, 129°55′ to 130°02′E) is located in the central part of the Ryukyu Islands of Japan in the northwestern Pacific (Figure 1). The Central and Northern Ryukyu Islands are situated near the northern boundary of a coral reef formation in the subtropical climate zone. Kikai Island consists of several uplifted terraces of Quaternary coral reefs that have experienced a

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high uplift rate of 1.8 m/kyr over the past 120 kyr BP (Ota and Omura 1992). The modern coral reef of Kikai Island is not well developed and has been growing without a reef crest or moat on the outer side of Holocene Terrace IV, which is estimated to have uplifted most recently after 2600 cal BP (U/Th age) (Webster et al. 1998).

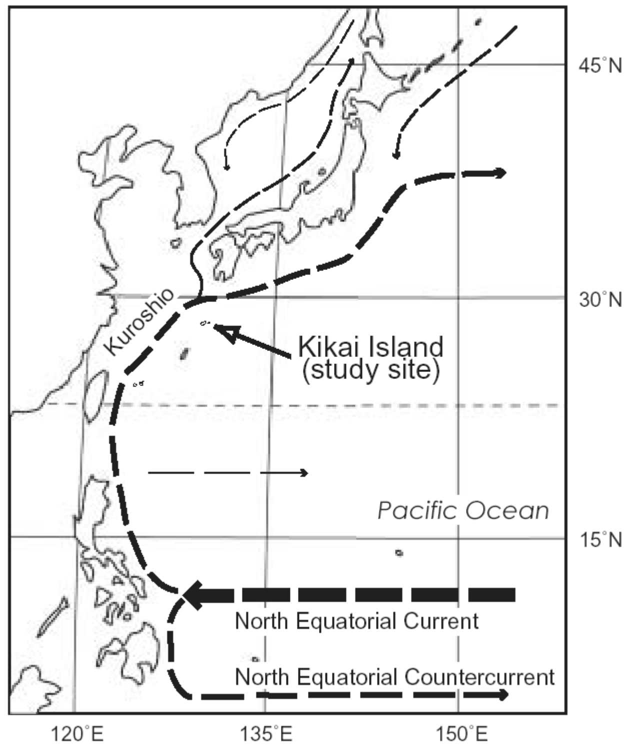

Figure 1 Location of Kikai Island and major surface currents in the northwestern Pacific. A modern coral colony of *Porites* sp. was collected off the southwestern coast of Kikai Island in the central part of the Ryukyu Islands, southern Japan.

A 25-cm-diameter modern, massive colony of *Porites* sp. was collected at the water depth of 2.9 m at Araki on the southwestern coast of Kikai Island in July 1998. The coral colony was cut to a 5-mm-thick skeletal slab parallel to the growth axis. Based on the observation of the X-radiograph image, a sampling line along the growth axis was determined. After cleaning the slab in an ultrasonic bath, samples were drilled out from the slab at 1.2-mm intervals for $\delta^{18}O$ measurements and at 1.5-mm intervals for $\Delta^{14}C$ measurements.

$\delta^{18}O$ analysis of 40- to 70-μg aliquots was performed on a Finnigan MAT252 isotope ratio mass spectrometer equipped with an automated carbonate reaction device (Kiel III) at the University of Tokyo. All measurements are reported relative to V-PDB; external precision for samples of the laboratory carbonate standard was 0.03‰ for $\delta^{18}O$ in general.

Approximately 7 mg of coral powder was dissolved under a vacuum with 85% phosphoric acid, and the carbon dioxide was converted to graphite by means of hydrogen reduction with an iron catalyst. ^{14}C measurements were conducted at the National Institute for Environmental Studies (NIES-TERRA), Japan. $\Delta^{14}C$ (‰) with decay collection are calculated according to standards defined by Stuiver and Polach (1977).

RESULTS AND DISCUSSION

$\delta^{18}O$ and $\Delta^{14}C$ of Kikai Island's Coral

Figure 2 shows $\delta^{18}O$ and $\Delta^{14}C$ variations as a function of depth from the coral surface and the X-radiograph image of a modern coral (Lab nr: KK-Ar-m1) from Kikai Island. The $\delta^{18}O$ values depict seasonal changes of about 1.8‰ on amplitude. The coral $\Delta^{14}C$ values demonstrate seasonal fluctuations superimposed on a secular decreasing toward the present.

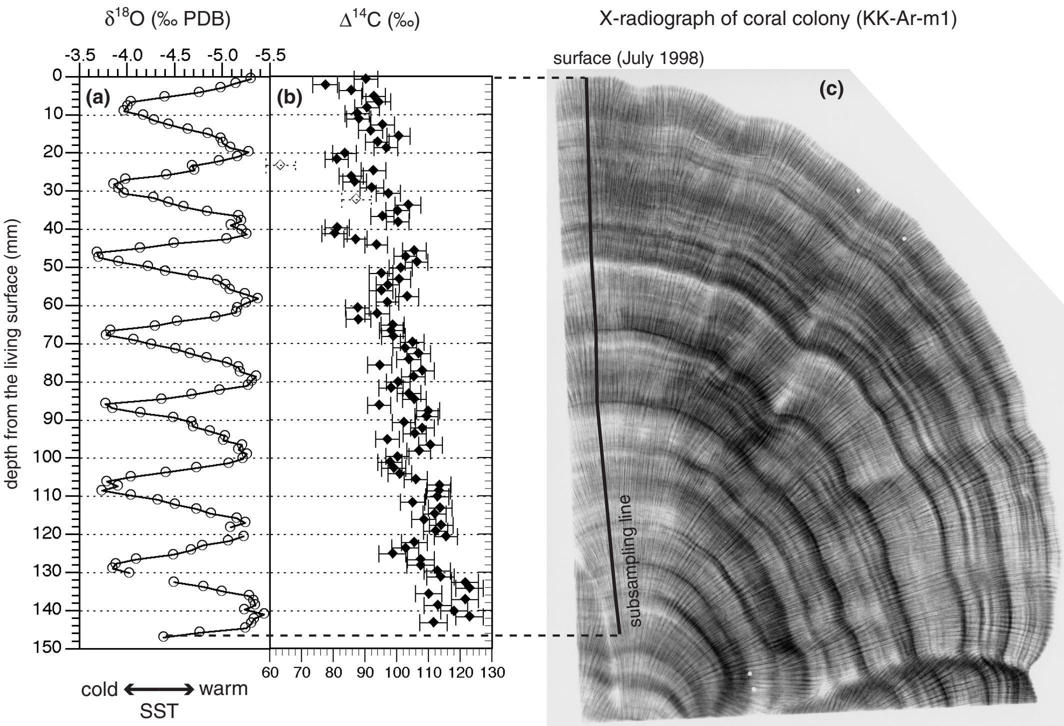

Figure 2 (a) $\delta^{18}O$ and (b) $\Delta^{14}C$ results from a modern coral colony (KK-Ar-m1) from Kikai Island as a function of depth from the living surface. (c) An X-radiograph of the coral is also shown. The subsampling line is indicated as a thick line along the growth axis. Error bars (1 σ) of $\Delta^{14}C$ are also indicated. Two $\Delta^{14}C$ data (shown as open squares) were rejected because total ^{14}C counts for these samples during AMS measurements were less than half of those for other samples.

Determination of Seasonal $\delta^{18}O$ and $\Delta^{14}C$ Fluctuation

Coral $\delta^{18}O$ is controlled by both sea surface temperature (SST) and seawater oxygen isotope ($\delta^{18}O_{sw}$) (e.g. Weber and Woodhead 1972). At Kikai Island, the influence of SST on coral $\delta^{18}O$ changes would be much larger than that of $\delta^{18}O_{sw}$. Observed $\delta^{18}O_{sw}$ at the Kikai Island's coast was relatively constant in a range of less than 0.2‰ (SMOW), without an apparent seasonality, during the 2 yr from 1999 to 2000 (Morimoto 2002). The amplitude of the seasonal SST variations is 8 °C on average at least for the last 20 yr from IGOSS NMC data (http://ingrid.ldeo.columbia.edu/). Maximum and minimum SST is 28–29 °C in July or August and 20.0–21.5 °C in February or March, respectively. Annual SST amplitude of 8 °C is transformed to a $\delta^{18}O$ change of about 1.6‰. The $\delta^{18}O$-based estimation of SST agreed well with the observed SST. The regression between estimated and observed SST showed a high correlation, $r^2 = 0.84$, suggesting that coral $\delta^{18}O$ at Kikai Island is likely controlled by SST.

The SST-$\delta^{18}O$ relationship was applied for determining the chronology with a monthly resolution. Coral $\delta^{18}O$ peaks, both in summer and winter, were fitted to the dates of 3-week-averaged SST maximums and minimums from IGOSS NMC data. For the sections between summer and winter SST peaks, we interpolated assuming linear growth rates between $\delta^{18}O$ peaks to determine the time axis. The coral $\delta^{18}O$ and $\Delta^{14}C$ changes on the time axis as well as monthly SSTs are shown in Figure 3. The $\Delta^{14}C$ time series has a time resolution of 12–15 subsamples per yr during 1991–1998.

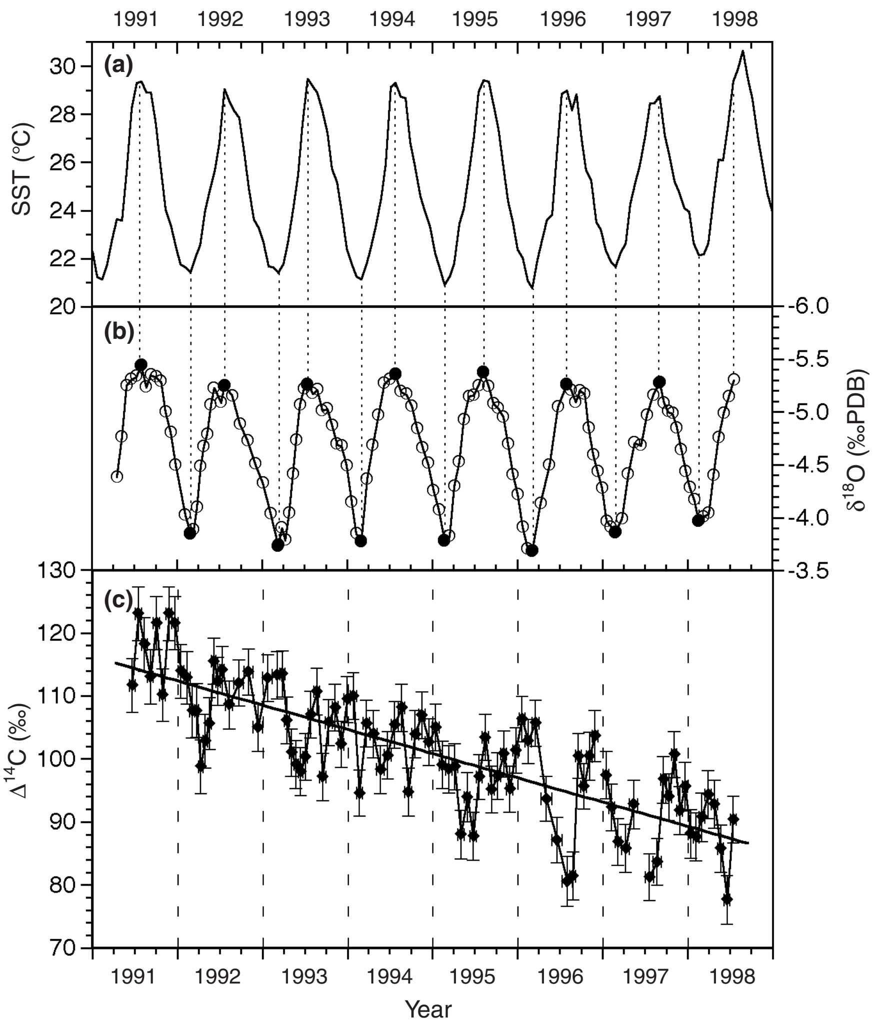

Figure 3 (a) Three-week mean SST (from IGOSS NMC data), (b) coral $\delta^{18}O$, and (c) $\Delta^{14}C$ time series. The timescale for the coral was determined from the SST-$\delta^{18}O$ relationship using summer and winter peak data (fittings are shown as dotted lines). Coral $\delta^{18}O$ peaks are indicated as solid circles. The $\Delta^{14}C$ time series for 1991–1998 shows seasonal and interannual variability. A linear $\Delta^{14}C$ decreasing trend of 3.8 ‰/yr is drawn as a thick line. Vertical error bars are standard errors ($\pm 1\ \sigma$) of the weighted mean $\Delta^{14}C$, and horizontal bars indicate periods contained in each coral subsample.

Secular and Seasonal Changes of $\Delta^{14}C$ for 1991–1998

The coral $\Delta^{14}C$ values at Kikai Island decreased from 123 to 78‰ during the 8 yr of 1991–1998 (Figure 3). A $\Delta^{14}C$ decreasing trend of 3.8 ‰/yr was obtained from a linear fitting for all data. Measurements from Europe over the last 2 decades provide evidence for a strong continental Suess effect, with fossil fuels driving down $\Delta^{14}C$ in atmospheric CO_2 (Levin et al. 1989; Meijer et al. 1995; Levin and Kromer 1997). The $\Delta^{14}C$ secular decreasing from coral is similar to the atmospheric ^{14}C changes in the 1990s observed at wide range of the Northern Hemispheric stations.

The seasonal amplitude of coral $\Delta^{14}C$ at Kikai Island exceeded 18‰ for the period of 1991–1998, with a minimum in late spring or summer and a maximum in winter (Figure 3). The minimum month of residual $\Delta^{14}C$ differs from year to year after subtracting the linear decreasing trend; February in 1994, April in 1992, May in 1995, June in 1993, July in 1997, and August in 1996. Among these seasonal changes, a drop of 25‰ in 1996 from March to August is the largest decrease in the period.

For the equatorial Pacific, a few previous studies have shown sub-annual variability of coral $\Delta^{14}C$ in the term after atmospheric nuclear bomb testing. Coral $\Delta^{14}C$ at Nauru Island (0.5°S, 166.9°E) is characterized mainly by large interannual variability, in concert with ENSO-associated trade wind changes as well as atmospheric $\Delta^{14}C$ changes. After 1960, only some years have shown a clear seasonal cycle of 20‰ (Guilderson et al. 1998). At the Galapagos Islands in the eastern Pacific, coral $\Delta^{14}C$ reveals large seasonal cycles ranging from 20–100‰ during the period 1957–1983. This phenomenon can be explained by an upwelling signal related to ENSO and atmospheric $^{14}CO_2$ changes (Guilderson and Schrag 1998). In the equatorial western Pacific, large interannual and seasonal coral $\Delta^{14}C$ variability of 15–60‰ in the Indonesian Seaway is observed during the period 1970–1985. This variability has been mostly related to contribution changes of source waters to the strait throughflow (Moore et al. 1997). From $\Delta^{14}C$ annual variations of Kikai Island and these previous studies, there are apparent seasonal $\Delta^{14}C$ cycles of DIC in the surface seawater over the wide range of the equatorial and subequatorial Pacific.

Moore et al. (1997) summarized potential sources of seasonal $\Delta^{14}C$ variability of the surface ocean, such as an invasion of CO_2 from the atmosphere, regional seawater mixing due to an upwelling, and horizontal advection. The phase of seasonal $\Delta^{14}C$ cycles in atmospheric CO_2 with minimum values in winter somewhat differs from that of coral $\Delta^{14}C$ cycles from Kikai Island. The amplitudes of atmospheric $\Delta^{14}C$ changes in this term are 5–30‰ in Europe, not markedly larger than those of coral $\Delta^{14}C$ (Meijer et al. 1995; Levin and Kromer 1997). Therefore, seasonal $\Delta^{14}C$ changes of DIC in the surface ocean cannot be explained only by direct effect of atmospheric ^{14}C. Furthermore, Kikai Island is not located in a region that is influenced by a local upwelling. The seasonality of $\Delta^{14}C$ may indicate a larger-scale oceanic phenomenon.

The spatial distribution of the sea-surface DIC-$\Delta^{14}C$ in the Pacific was revealed by the GEOSECS and WOCE programs. In the equatorial region, modeling studies have estimated that seasonally varying lateral advections are the dominant mechanism for $\Delta^{14}C$ variability of the surface waters (Rodgers et al. 1997). The seasonal $\Delta^{14}C$ cycle observed in this study is potentially explained by horizontal advections, for example, of the North Equatorial Current and the Kuroshio Current.

The present and future investigation of coral-based $\Delta^{14}C$ reconstructions of surface water DIC make it possible to understand seasonal cycles of horizontal advections of the ocean, together with CO_2 exchanges between atmosphere and ocean over the last several decades to centuries.

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REFERENCES

Druffel ERM. 1989. Decade timescale variability of ventilation in the North Atlantic: high-precision measurements of bomb radiocarbon in banded corals. *Journal of Geophysical Research* 94(C3):3271–85.

Druffel EM, Linick TW. 1978. Radiocarbon in annual coral rings from Florida. *Geophysical Research Letters* 5(11):913–6.

Guilderson TP, Schrag DP. 1998. Abrupt shift in subsurface temperature in the tropical Pacific associated with changes in El Niño. *Science* 281:240–3.

Guilderson TP, Schrag DP, Kashgarian M, Southon J. 1998. Radiocarbon variability in the western equatorial Pacific inferred from a high-resolution coral record from Nauru Island. *Journal of Geophysical Research* 103(C11):24,641–50.

Levin I, Kromer B. 1997. Twenty years of atmospheric $^{14}CO_2$ observations at Schauinsland station, Germany. *Radiocarbon* 39(2):205–18.

Levin I, Schuchard J, Kromer B, Münnich KO. 1989. The continental European Suess effect. *Radiocarbon* 31(3):431–40.

Meijer HA, van der Plicht J, Gislefoss JS, Nydal R. 1995. Comparing long-term atmospheric ^{14}C and ^{3}H records near Groningen, the Netherlands with Fruholmen, Norway and Izaña, Canary Islands ^{14}C stations. *Radiocarbon* 37(1):39–50.

Moore MD, Schrag DP, Kashgarian M. 1997. Coral radiocarbon constraints on the source of the Indonesian throughflow. *Journal of Geophysical Research* 102(C6):12,359–65.

Morimoto M. 2002. A high time-resolution calibration of coral oxygen isotope records and mid-Holocene climate in the northwestern Pacific from corals [PhD dissertation]. Tokyo: University of Tokyo. 209 p.

Nozaki Y, Rye DM, Turekian KK, Dodge RE. 1978. A 200-year record of carbon-13 and carbon-14 variations in a Bermuda coral. *Geophysical Research Letters* 5(10):825–8.

Ota Y, Omura A. 1992. Constructing styles and rates of tectonic uplift of coral reef terraces in the Ryukyu and Daito Islands, southwestern Japan. *Quaternary International* 15/16:17–29.

Rodgers KB, Cane MA, Schrag DP. 1997. Seasonal variability of sea surface $\Delta^{14}C$ in the equatorial Pacific in an ocean circulation model. *Journal of Geophysical Research* 102(C8):18,627–39.

Stuiver M, Polach HA. 1977. Discussion: reporting of ^{14}C data. *Radiocarbon* 19(3):355–63.

Weber JN, Woodhead PMJ. 1972. Temperature dependence of oxygen-18 concentration in reef coral carbonates. *Journal of Geophysical Research* 77:463–73.

Webster JM, Davies PJ, Konishi K. 1998. Model of fringing reef development in response to progressive sea level fall over the last 7000 years—(Kikai-jima, Ryukyu Islands, Japan). *Coral Reefs* 17:289–308.