DECADAL CHANGES OF RADIOCARBON IN THE SURFACE BAY OF BENGAL: THREE DECADES AFTER GEOSECS AND ONE DECADE AFTER WOCE

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ABSTRACT. Radiocarbon was measured in the surface seawater dissolved inorganic carbon (DIC) of the Bay of Bengal during November 2006. A meridional transect of the $\Delta^{14}C$ in DIC was obtained from measurements in closely spaced samples collected roughly along 88°E. The $\Delta^{14}C$ of these samples ranged from 44‰ to 57.7‰ (mean 51.8 ± 1.1‰, $n = 12$), and 38‰ at one station in the northern Bay of Bengal. The overall pattern of ^{14}C distribution in DIC of surface Bay of Bengal during 2006 was roughly similar to that during the WOCE expedition of 1995. These results indicate a $\Delta^{14}C$ decline rate of ~4‰ per decade since WOCE in the surface Bay of Bengal, which is much smaller compared to a decline rate of ~25‰ per decade observed in the 2 decades between the GEOSECS and WOCE expeditions, due to the smaller atmosphere-ocean $\Delta^{14}C$ gradient.

INTRODUCTION

Temporal changes of radiocarbon in oceanic dissolved inorganic carbon (DIC) provide valuable clues for ocean circulation and air-sea CO_2 exchange. Monitoring decadal changes of ^{14}C in oceanic DIC offers a way for validating ocean circulation models, which predict the spatial and temporal changes in the distribution of radiotracers (Guilderson et al. 2000). The earliest measurements of ^{14}C in the northern Indian Ocean were carried out during the GEOSECS expeditions of 1977–1978 (Stuiver and Östlund 1983), and in further detail 2 decades later during the WOCE expedition of 1994–1995 (Key and Quay 2002). Bhushan et al. (2000, 2003), Dutta et al. (2000, 2007), and Dutta (2001) reported DIC ^{14}C measurements at several other important locations in the northern Indian Ocean, from oceanographic cruises of the Physical Research Laboratory (PRL), India (Figure 1). Many of these stations were reoccupations of GEOSECS expeditions. Among other goals, these expeditions aimed to determine a "snapshot" scenario of oceanic ^{14}C, ocean circulation tracers, and decadal changes in bomb-^{14}C penetration.

The Bay of Bengal in the northern Indian Ocean experiences high rainfall and seasonally reversing surface ocean circulation driven by monsoonal winds (Schott and McCreary 2001; Shankar et al. 2002). The basin receives large freshwater input through several major rivers, as well as the Indonesian Throughflow (Sengupta et al. 2006). The northern Bay of Bengal also receives a large amount of freshwater through submarine groundwater discharge (Moore 1997). Owing to this unique oceanographic setting, with mixing of various water masses with contrasting ^{14}C contents and seasonally reversing circulation, the Bay of Bengal region is an area of considerable interest for studies of ^{14}C as a tracer. The WOCE surface $\Delta^{14}C$ results show large meridional variation (51–78‰ along 90°E) and much smaller zonal variation (55–65‰ along 10°N). These measurements revealed the highest surface $\Delta^{14}C$ values, around 78‰, observed in the SE Bay of Bengal near 8°30′N close to the Indonesian Throughflow, and 64‰ at a station in the NE Bay of Bengal. The lowest surface $\Delta^{14}C$ values of ~52‰ were observed in the SW Bay of Bengal due to advection of ^{14}C-depleted waters from the Arabian Sea. Low $\Delta^{14}C$ values are also expected in the northern Bay of Bengal close to the Ganges-Brahmaputra estuary, due to submarine discharge of old groundwater. ^{14}C ages of coastal groundwater samples near the northern Bay of Bengal range from ~3 kyr BP to as old as ~9 kyr BP (Ravi

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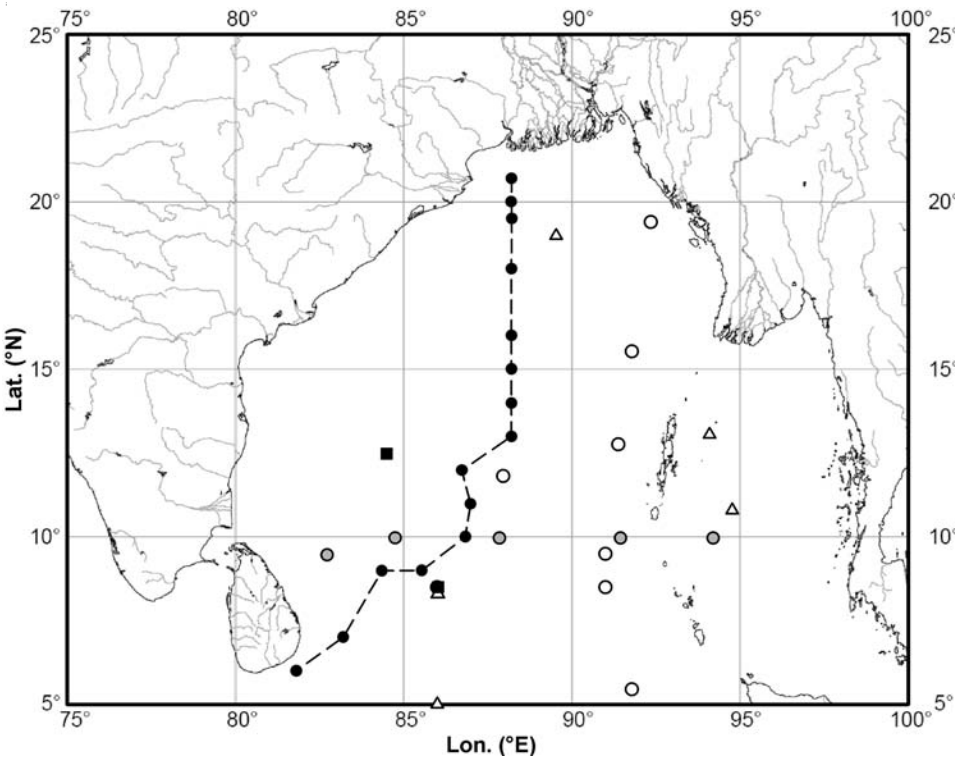

Figure 1 ^{14}C sampling stations in the Bay of Bengal: GEOSECS (stations #445 and #446, squares); WOCE (I09N, open circles; I10E, shaded circles); PRL (triangles); SM-TT-06 cruise (filled circles).

Prasad et al. 2008). Mixing of old groundwater via submarine groundwater discharge can significantly influence marine DIC ^{14}C ages. Measurements of samples from a PRL expedition in 1999 recorded the lowest surface $\Delta^{14}C$ of 18 ± 6‰ near 20°N (Dutta et al. 2000; Dutta 2001).

To understand the spatial variation and decadal change of ^{14}C in the surface water DIC of Bay of Bengal, we planned to measure ^{14}C in surface seawater samples at close by locations. Here, we report $\Delta^{14}C$ measured in the DIC of surface Bay of Bengal, measured in samples collected in 2006 along a meridional transect.

MATERIALS AND METHODS

Surface seawater samples were collected onboard the R/V *Samudra Manthan* during November 2006, approximately following a meridional transect along 88°E in the central Bay of Bengal from 21°N to about 5°N (Figure 1). Samples were collected from a depth of 5 m using 5-L PVC Niskin water samplers. Soon after collection, the samples were poisoned with saturated aqueous $HgCl_2$ solution and stored in refrigerated airtight bottles until further analysis. Salinity (conductivity) and temperature were measured onboard using a Multiline P4^{TM} water analysis kit. The samples were processed at the ^{14}C laboratory of the Institute of Physics, India. CO_2 was extracted from these samples by acidifying with H_3PO_4 under vacuum. The liberated CO_2 was cryogenically purified, converted to graphite by H_2 reduction over Fe catalyst (Vogel et al. 1984), and pressed into targets for accelerator mass spectrometry (AMS) ^{14}C analysis. ^{14}C measurements in the graphite samples were carried out at the AMS facility of the Center for Applied Isotope Studies, University of Georgia,

Athens, USA, using a NEC 1.5 SDH-1 0.5MV compact AMS system. An aliquot of CO_2 was used for $\delta^{13}C$ analysis at the National Institute of Oceanography, Goa, India, using a Finnigan Delta V™ stable isotope mass spectrometer. The average precision of $\delta^{13}C$ measurements was 0.04‰. The measured $\delta^{13}C$ values in these samples ranged from –0.4 to –1.2‰ (Table 1). These values possibly indicate a fractionation of about –2‰, since $\delta^{13}C$ of surface ocean DIC typically range from ~1 to 2‰. During the CO_2 extraction by acid-hydrolysis, even though the samples were agitated under vacuum using an ultrasonic bath, they were not actively purged using a stream of inert gas. The observed fractionation is likely due to incomplete CO_2 extraction during the hydrolysis stage. However, in all cases the CO_2 yields were >95% of the expected amount. The $\delta^{13}C$ results were only used for isotopic fractionation correction to determine $\Delta^{14}C$ (Stuiver and Polach 1977). Typical internal precisions for AMS $\Delta^{14}C$ measurements were better than ±3%, while the repeat measurements were within ±6%. ^{14}C measurements in the modern standards VIRI-A and VIRI-C agreed very well with their consensus values. The precision of $\Delta^{14}C$ measurements in the modern standards was ±5%.

Table 1 Results of ^{14}C analysis in surface samples from the Bay of Bengal.

Sample	Date	Lat. (°N); Lon. (°E)	Temp. (°C)	Salinity (mg/L)	$\delta^{13}C$ (‰)	$\Delta^{14}C$ (‰)	Lab code
SM-TT-06 / W-9	12-Nov-06	20.69; 88.21	28.4	30.0	–1.23	—	—
SM-TT-06 / W-10	13-Nov-06	20.00; 88.20	28.4	32.0	–0.59	38.5 ± 2.4	IP-869
SM-TT-06 / W-11	13-Nov-06	19.50; 88.22	28.2	30.8	–0.68	52.8 ± 2.9	IP-870
SM-TT-06 / W-12	14-Nov-06	18.00; 88.21	28.1	30.2	–0.52	55.3 ± 2.4	IP-871
SM-TT-06 / W-13	14-Nov-06	17.00; 88.21	28.5	29.7	–0.71	—	—
SM-TT-06 / W-14	14-Nov-06	16.01; 88.21	29.1	32.4	–0.39	51.4 ± 2.5	IP-872
SM-TT-06 / W-15	15-Nov-06	15.00; 88.21	29.3	32.2	–0.49	49.5 ± 2.4	IP-873
SM-TT-06 / W-16	15-Nov-06	14.00; 88.21	29.3	31.8	–0.61	57.7 ± 2.7	IP-874
SM-TT-06 / W-17	16-Nov-06	13.01; 88.21	29.3	32.3	–0.41	44.0 ± 2.4	IP-875
SM-TT-06 / W-18	16-Nov-06	12.00; 86.74	29.2	32.0	–0.88	—	—
SM-TT-06 / W-19	17-Nov-06	10.99; 86.99	29.2	32.8	–0.43	50.2 ± 2.8	IP-878
SM-TT-06 / W-20	17-Nov-06	10.01; 86.84	29.7	33.2	–0.61	52.2 ± 2.7	IP-879
SM-TT-06 / W-21	18-Nov-06	9.00; 85.54	29.8	32.4	–0.86	55.9 ± 3.4	IP-880
SM-TT-06 / W-22	18-Nov-06	8.99; 84.36	29.1	32.0	–0.83	54.4 ± 3.0	IP-882
SM-TT-06 / W-23	19-Nov-06	7.01; 83.21	29.2	32.3	–0.62	52.7 ± 2.8	IP-884
SM-TT-06 / W-24	19-Nov-06	6.00; 81.82	29.3	29.3	–0.99	49.2 ± 2.7	IP-885

RESULTS AND DISCUSSION

Spatial Variation of Surface $\Delta^{14}C$

The DIC $\Delta^{14}C$ of these surface seawater samples are presented in Table 1 and Figure 2. For the stations SM-TT-06/W-11 to W-24, the $\Delta^{14}C$ values mostly range from 44‰ to 57.7‰ (error-weighted mean 51.8 ± 1.1‰, n = 12). As evident in Figure 2, the overall pattern of $\Delta^{14}C$ distribution was roughly similar as it was during the WOCE expedition a decade ago. Higher $\Delta^{14}C$ values of 56‰ were recorded in the southern Bay of Bengal near 9°N, and also around 19°N. The lowest $\Delta^{14}C$ of 38.5‰ was observed at the station SM-TT-06/W-10 in the northern Bay of Bengal at 20°N. A low $\Delta^{14}C$ of 44‰ was also measured at SM-TT-06/W-17 in the central Bay of Bengal around 13°N.

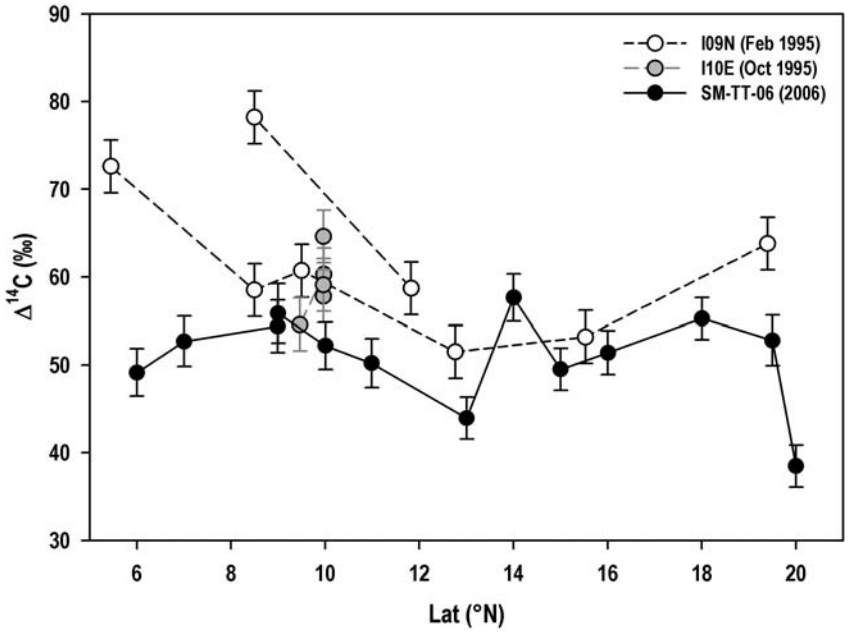

Figure 2 Spatial variation of $\Delta^{14}C$ in the DIC of the surface Bay of Bengal during the WOCE expedition and this study.

Decadal Changes of Surface $\Delta^{14}C$

To determine decadal changes in surface $\Delta^{14}C$, we focus on the ^{14}C measurements in the southwestern Bay of Bengal (within 7–13°N and 82–88°E), where ^{14}C data are available from all 3 earlier expeditions. Two GEOSECS stations, 4 WOCE stations (2 each of I09N and I10E), 1 PRL station, and 4 stations from this study (IOP/GSI) were chosen to determine their error-weighted average $\Delta^{14}C$ values. The average surface water $\Delta^{14}C$ during early 1978 was 110 ± 10‰ for GEOSECS stations #445 and #446. The average $\Delta^{14}C$ for WOCE I09N station (February 1995) was 67 ± 10‰, and 58 ± 3‰ for WOCE I10N station (October 1995). PRL ^{14}C measurements at the station SS#152-3829 (reoccupation of GEOSECS #446) were 38 ± 7‰ at 5 m and 64 ± 4‰ at 45 m. The average $\Delta^{14}C$ for the 4 stations of this study (SM-TT-06/W17 to W21) was 53.2 ± 2.5‰. These decadal changes of surface $\Delta^{14}C$ in the southwestern Bay of Bengal are shown in Figure 3. This analysis shows that $\Delta^{14}C$ in the surface Bay of Bengal DIC has decreased at ~25‰ per decade between 1978 and 1995, but only 4‰ in the decade between 1995 and 2006. A lower decline rate of surface ocean $\Delta^{14}C$ during the last decade is expected given the significantly lower atmosphere-ocean ^{14}C gradient. Tropospheric $\Delta^{14}C$ in the Northern Hemisphere has changed from ~326‰ in 1978 (Hua and Barbetti 2004) to ~105‰ in 1995 (Dutta et al. 2006) and ~52‰ during February 2007 (Xu et al. 2007). The atmosphere-ocean ^{14}C gradient at the Bay of Bengal was thus negligible during the period of this study.

Seasonal Changes of Surface $\Delta^{14}C$

In the above analysis of decadal ^{14}C changes, we have ignored any effect of seasonal ^{14}C variations in the surface Bay of Bengal. A large seasonal ^{14}C variation is not unexpected given the strong seasonality of monsoon-driven surface currents. To date, there is no detailed study of seasonal $\Delta^{14}C$ changes in the Bay of Bengal. From one-time ^{14}C measurements, it is not possible to study such sea-

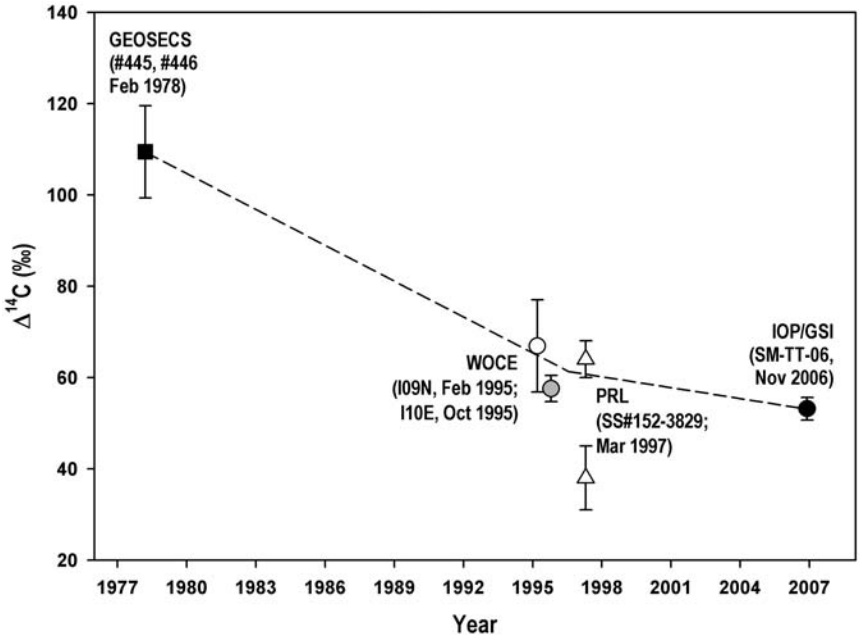

Figure 3 Decadal $\Delta^{14}C$ changes in the DIC of the surface Bay of Bengal (near 10°N, 86°E)

sonal changes. The results from WOCE stations I09N (February 1995) and I10E (October 1995) indicate a seasonal $\Delta^{14}C$ shift in the southwestern Bay of Bengal by 10‰, which is not statistically significant. Average surface $\Delta^{14}C$ values measured during the PRL expeditions in February/March of 1997 and 1999 was 47‰. From these results, it is apparent that the spatiotemporal variation of surface $\Delta^{14}C$ in the Bay of Bengal is complex. A large variability in $\Delta^{14}C$ of surface ocean DIC is not uncommon. Druffel and Griffin (2008) have demonstrated daily variability between 11‰ and 30‰ in surface DIC $\Delta^{14}C$ at multiple occupations of single sites in the central North Pacific, the Sargasso Sea, and the Southern Ocean. Monthly seasonal $\Delta^{14}C$ amplitudes were ~10–20‰ observed in the subtropical Pacific (Druffel 1987). Surprisingly, the amplitudes of such variations were not necessarily lower during the late 1990s than they were in the 1980s.

Further measurements are needed to better understand the spatial and temporal changes in $\Delta^{14}C$ of the surface Bay of Bengal. Measurements of $\Delta^{14}C$ along multiple transects are essential. Monthly measurements of $\Delta^{14}C$ at selected coastal locations are needed to decipher seasonal changes of ^{14}C in DIC in response to seasonally reversing surface circulation. A detailed study of $\Delta^{14}C$ close to the Ganges-Brahmaputra estuary in the northern Bay of Bengal may give insights to the effects of submarine groundwater discharge on marine ^{14}C.

CONCLUSIONS

$\Delta^{14}C$ in the DIC of the surface Bay of Bengal during 2006 mostly ranged from 44‰ to 58‰ (mean 52 ± 1‰). The lowest $\Delta^{14}C$ value of 38‰ was observed at a station in the northern Bay of Bengal. The latitudinal $\Delta^{14}C$ pattern as observed during November 2006 roughly followed the overall trend observed during the WOCE expedition during February and October 1995. The change in surface $\Delta^{14}C$ as observed for the southwestern Bay of Bengal (near 10°N, 86°E) was much smaller during 1995–2006 than it was in the 2 decades previous. The atmosphere-ocean $\Delta^{14}C$ gradient for the Bay

of Bengal during late 2006 was negligible. Further measurements are needed to evaluate the effects of seasonal circulation changes and submarine groundwater discharge on the ^{14}C in DIC at the northern Bay of Bengal.

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