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BOMB ^{14}C IN THE INDIAN OCEAN MEASURED BY ACCELERATOR MASS SPECTROMETRY: OCEANOGRAPHIC IMPLICATIONS

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ABSTRACT. AMS ^{14}C measurements on samples collected in the tropical-equatorial Indian Ocean during the INDIGO program (leg II, 1986) are presented and compared with β-counting results obtained under both INDIGO program and GEOSECS expedition in the Indian Ocean (1978). The most significant observation is a doubling of the bomb-^{14}C inventory and mean penetration depth in the equatorial zone. Based on hydrologic considerations, two hypotheses can be proposed: 1) direct influx of Pacific mid-latitude waters through the Indonesian archipelago and 2) advection and/or mixing with Mode Water from the southern gyre of the Indian Ocean. Results obtained with a general circulation model of the ocean suggest that the influx from the Pacific is important in the upper 300m and that below 500m the bomb-^{14}C budget is dominated by Mode Water advection.

INTRODUCTION

One of the ways to study the CO_2 exchange between the atmosphere and ocean is to measure ^{14}C which is produced mainly by interaction between cosmic-ray-produced neutrons and nitrogen atoms in the atmosphere. Rapidly after formation, ^{14}C atoms are oxidized and mixed as $^{14}CO_2$ in the whole atmosphere. The natural production is roughly balanced by the disappearance of $^{14}CO_2$ in other carbon reservoirs and, thus, atmospheric ^{14}C activity was in near steady state until the late 1950s when large-scale testing of thermonuclear weapons began. This brought the ^{14}C activity of the northern hemispheric troposphere to ca 100% above its normal value (Nydal & Lovseth, 1983), and it has been decreasing gradually in response to atmospheric homogenization and penetration of $^{14}CO_2$ into other carbon reservoirs, such as oceans and terrestrial biosphere. The bomb-produced ^{14}C can, thus, be used as a transient tracer to test models of oceanic circulation and quantify the oceanic invasion of CO_2 excess produced by anthropogenic deforestation and fossil-fuel consumption.

Such measurements have been carried out by conventional counting of the beta radioactivity of ^{14}C. This method typically requires several grams of carbon, thus sampling of ca 250L of sea water per ^{14}C determination and chemical extraction of dissolved carbonate species aboard the oceanographic vessel. We demonstrate that ^{14}C measurements can also be performed by Accelerator Mass Spectrometry (AMS), using water samples of only 100ml, without shipboard chemical treatment.

The water-column inventories for bomb ^{14}C at stations occupied during the GEOSECS program (1972–1978) show pronounced minimal values in equatorial zones (Broecker *et al*, 1985). In the case of the Indian Ocean, values between 10°N and 10°S were typically on the order of 5×10^9 atoms/cm^2, whereas the zone between 20°S and 40°S was characterized by

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values that were higher by a factor of three. Various oceanic models have led to the suggestion that a sizable portion of the bomb ^{14}C that entered the tropical ocean has been transported to adjacent temperate zones, and that the low-latitude deficit enables an equatorial upwelling component to be calculated (*cf*, eg, Broecker *et al*, 1985). We present ^{14}C measurements performed on water samples collected between 20°S and 10°N during the INDIGO expedition of the French *R/V Marion Dufresne* in the Indian Ocean (leg II, April 1986). Our data allow precise comparison with ^{14}C profiles obtained at the same locations during the GEOSECS cruise in 1978 (Stuiver & Östlund, 1983) and estimate the bomb-^{14}C penetration during the last eight years. Indeed, as the prenuclear distribution of natural ^{14}C in the ocean thermocline is poorly known, it is particularly important to have two signatures of the transient inventory in order to quantify precisely the chronology of the invasion and to verify the flux parameters of oceanic models.

METHODS

Deep sea water was collected by a "rosette" system equipped with conductivity, temperature, pressure and pO_2 sensors (CTD, Guideline Instruments Inc, pO_2 Beckman Co) and 12L Niskin bottles. The water was rapidly siphoned into borosilicated 500ml bottles and poisoned with 5ml of saturated $HgCl_2$ solution. This operation is designed to suppress isotope fractionation due to respiration and photosynthesis of micro-organisms, which could affect the $\delta^{13}C$ and $\Delta^{14}C$ measurements. The bottles were then closed with greased ground stoppers and stored immediately at +4°C in specially constructed shipping boxes in which the stoppers were kept tightly in place during the oceanographic cruise.

In the laboratory, the CO_2 is extracted from the sea water in a special vacuum system shown in Figure 1. The whole system is first evacuated until a residual pressure $<10^{-4}$ mbar can be maintained (with an oil diffusion pump); it is then filled with pure helium and the traps are cooled (T_1 and T_2 at –75°C and T_3 at –190°C). Ca 100ml of sea water is directly pushed by helium gas into the "stripping vessel", phosphoric acid is added to diminish the CO_2-solubility and a circulation of helium is established. The CO_2 is separated from the water at –75°C and then trapped at –190°C in the coiled trap. Since the vector gas is allowed to flow out of the system, the trapping efficiency of T_3 is of great importance. Preliminary experiments were conducted using a CO_2-coulometer on a similar extraction system built at L-DGO. In the system depicted in Figure 1 (built at the CFR), the operating parameters were determined manometrically and correspond to a flow rate of ca 100ml/min (normalized to the air) and to a stripping time of roughly 1 hr. The CO_2 is then separated in two aliquots: one for AMS and the other for $\delta^{13}C$ measurement on a conventional mass spectrometer. The extraction efficiency can be estimated and corresponds to a mean of ca 98-99% of the ΣCO_2 concentration measured at sea by a potentiometric method.

For AMS measurements, the samples and standards are prepared by catalytic reduction of CO_2 on iron powder as described by Vogel *et al* (1984). After a reduction which typically lasts 6 to 8 hr, the carbon-iron

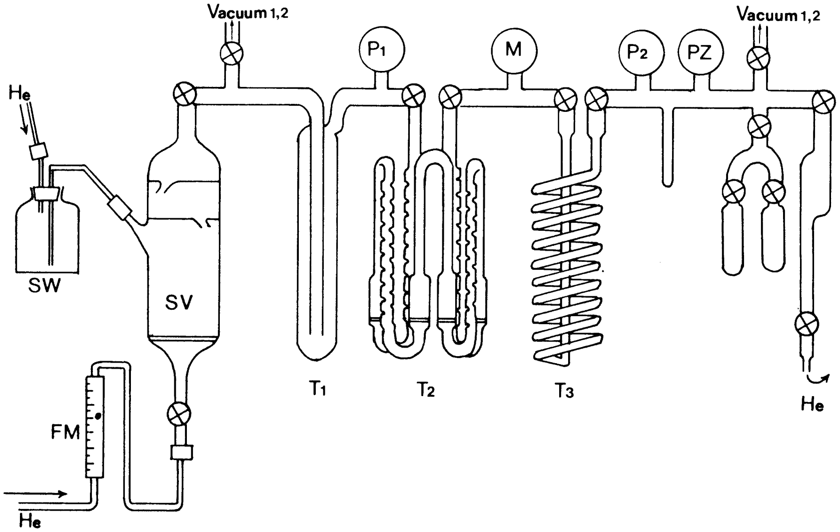


Fig 1. Schematic description of the CO_2 extraction system (CFR). T_1, T_2, T_3 = cold traps (respectively at -75°C, -75°C and -190°C); P_1, P_2 = Pirani gauges; M = manometer; PZ = piezoresistive pressure gauge; vacuum 1, 2 = vacuum generated by a primary rotary pump and an oil diffusion pump; SV = stripping vessel; FM = flow-meter, SW = seawater sample

mixture is then packed in a small aluminum cylinder to form a target ready for use in AMS. The procedure for AMS measurements has been described elsewhere (Arnold *et al*, 1987). For the data presented in the following development, we estimated the reproducibility of the oxalic acid standard which corresponds to a mean normalized ratio of 0.9947 and a standard deviation of $9.9^{o}/_{oo}$ (74 oxalic acid targets were measured in the seawater program). This value agrees well with the precision expected from the counting statistics generally obtained for this kind of measurement.

RADIOCARBON RESULTS

AMS results (CFR, Gif-sur-Yvette) were generated for water from seven oceanographic stations sampled during leg II of INDIGO (April 1986) (Table 1). Figure 2 presents the $\Delta^{14}C$ variations *vs* depth for seven locations: Station 27 (18°50′S, 54°50′E), Station 28 (17°20′S, 55°50′E), Station 32 (12°20′S, 53°40′E), Station 34 (8°50′S, 52°20′E), Station 45 (0°00′S, 50°60′E), Station 44 (0°00′S, 56°30′E), Station 65 (5°00′N, 52°00′E). Stations 27, 32 and 45 have the same locations as Stations 426, 424 and 420 of the GEOSECS expedition (Stuiver & Östlund, 1983) which allows us to compare the two sets of $\Delta^{14}C$ data. Also, several INDIGO stations were used for classical β-counting measurements of 200L samples (Tritium Laboratory, University of Miami). Figure 2 shows the results from Stations 27, 32 and 45, with a typical 1σ error ca $4^{o}/_{oo}$ (Östlund & Grall, 1988). The two INDIGO data sets agree well, taking into account the error bars.

TABLE 1

Δ^{14}C values *vs* depth obtained by accelerator mass spectrometry (errors are at 1σ)

Station 27 Depth (m)	18°50S 54°50E Δ^{14}C‰	**Station 28** Depth (m)	17°20S 54°50E Δ^{14}C‰	**Station 32** Depth (m)	12°20S 53°40E Δ^{14}C‰
0	114±10	4	123±12	4	96±17
31	122±11	28	110±13	29	91±12
95	105±12	68	121±13	95	48±12
146	102±11	93	140±12	145	51±13
294	98± 8	142	127±13	294	21± 9
498	94± 8	195	110±13	496	-61±10
800	-79± 9	295	73±12	798	-131±10
1050	-147± 8	496	55±12	1202	-150±11
1205	-144± 8	606	-7±10	1505	-163±12
1504	-168± 8	784	-112±10	1961	-190±13
		1050	-143±13		
		1506	-160±10		
		1949	-182±10		

Station 34 Depth (m)	8°50S 52°20E Δ^{14}C‰	**Station 45** Depth (m)	0°00S 51°00E Δ^{14}C‰	**Station 44** Depth (m)	0°00S 56°30E Δ^{14}C‰
4	65±10	5	85±13	4	78± 9
29	95±13	28	90± 9	29	109± 9
93	58±10	95	90±10	92	70± 8
143	48±10	143	61±11	143	66± 8
296	25±11	295	1±10	296	-2±10
497	-81±10	495	-52±10	498	-76± 9
796	-135± 8	798	-115± 8	795	-122± 9
1052	-171± 9	1048	-160± 8	1052	-144± 8
1207	-160± 9	1504	-172±14	1201	-148±11
1508	-181±10	2266	-172±10	1504	-165±11
		2671	-183± 8		

Station 65 Depth (m)	5°00N 52°00E Δ^{14}C‰
4	82±13
29	60±11
95	62±12
144	68±10
196	28± 9
395	-36±10
602	-115± 8
898	-135±13
1201	-164± 9
1352	-168±10

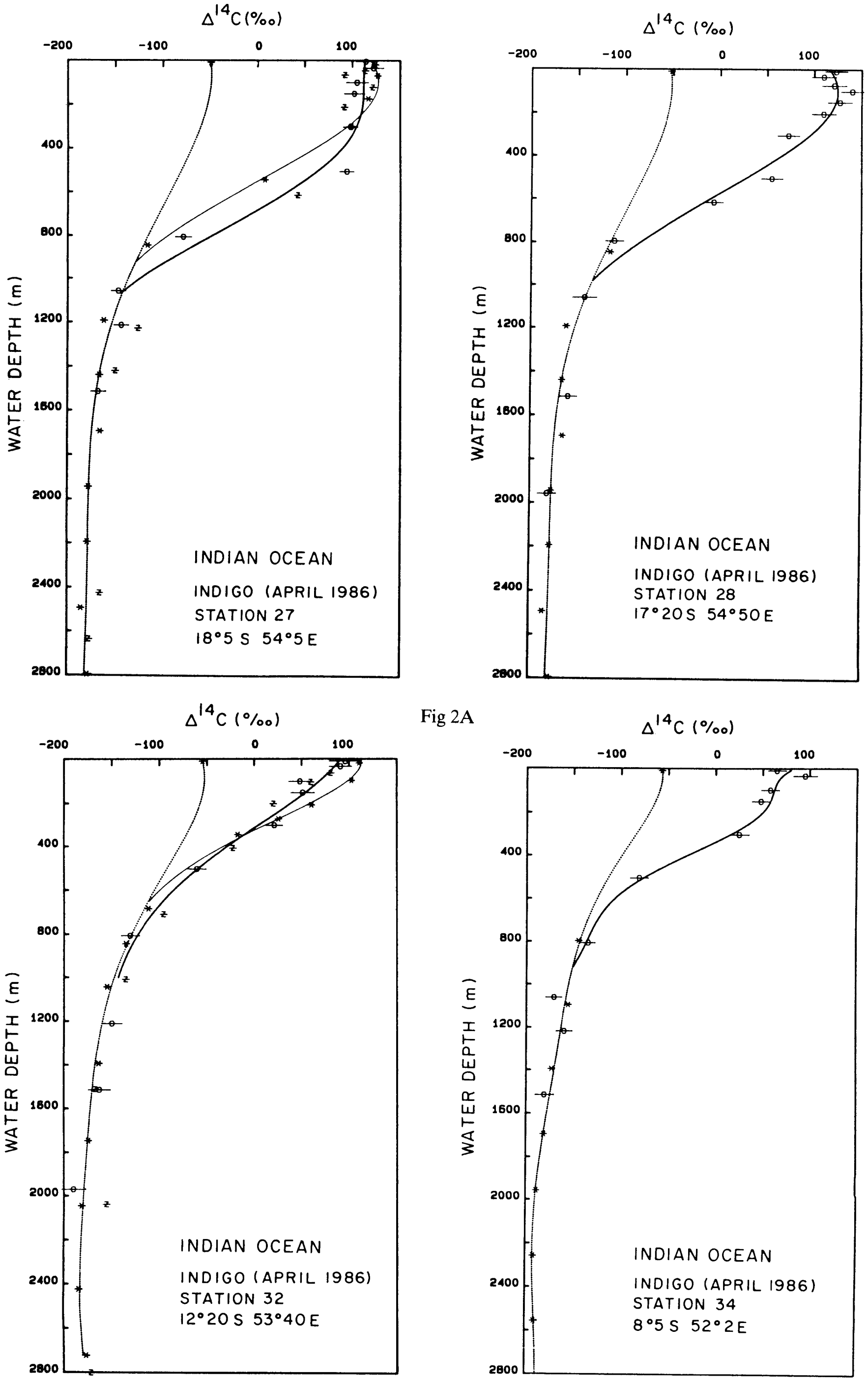
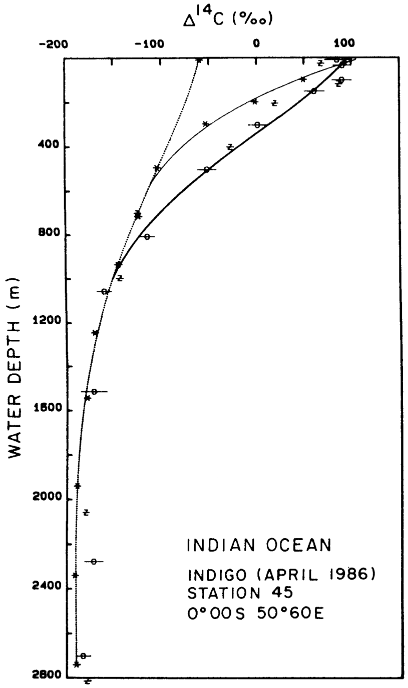
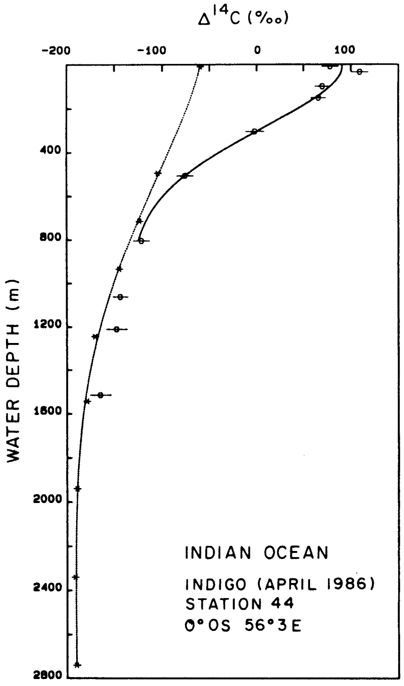

$\Delta^{14}C$ (‰)
-200
-100
0
100
WATER DEPTH (m)
400
800
1200
1600
2000
2400
2800
INDIAN OCEAN
INDIGO (APRIL 1986)
STATION 27
18°5 S 54°5 E
INDIAN OCEAN
INDIGO (APRIL 1986)
STATION 28
17°20S 54°50E
INDIAN OCEAN
INDIGO (APRIL 1986)
STATION 32
12°20S 53°40E
INDIAN OCEAN
INDIGO (APRIL 1986)
STATION 34
8°5 S 52°2E

Fig 2A





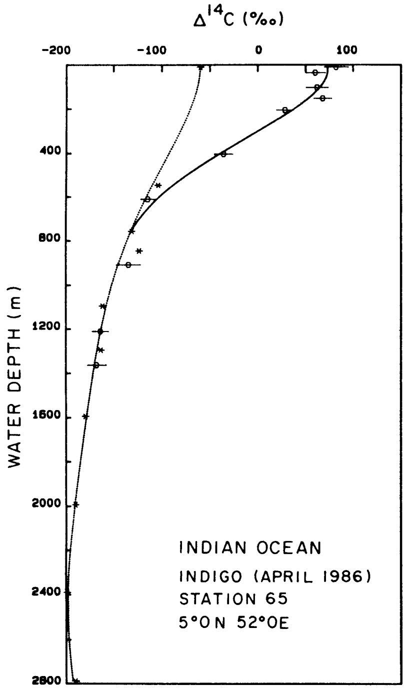


Fig 2B

Fig 2A and 2B. $\Delta^{14}C$ profiles for Stations 27, 28, 32, 34, 44, 45 and 65 of INDIGO expedition (leg II)

* = GEOSECS $\Delta^{14}C$ (β counting)

o = INDIGO $\Delta^{14}C$ (AMS)

z = INDIGO $\Delta^{14}C$ (β counting) (for Stations 27, 32 and 45)

—— = polynomial interpolation for INDIGO measurements (AMS)

—— = polynomial interpolation for GEOSECS measurements

..... = polynomial reconstruction of prenuclear $\Delta^{14}C$ levels

The seven $\Delta^{14}C$ profiles in Figure 2 demonstrate that thermonuclear ^{14}C is present in the upper part of the water column, and that for depths >ca 1000m, the INDIGO measurements agree well with the GEOSECS results, defining the prenuclear level of ^{14}C activity.

Following Broecker *et al* (1985), we estimated mean depth of penetration and water column inventories of bomb-produced ^{14}C by integrating the area between the observed $\Delta^{14}C$ curves and the reconstructed $\Delta^{14}C$ *vs* depth curve for prenuclear time (Z expressed in meters, $\Sigma^{14}C$ expressed in 10^9 atoms of bomb ^{14}C per cm^2). The unfortunate aspect of this calculation convention is that, using $\Delta^{14}C$ values, which refer to terrestrial wood, the inventory is exactly 5% lower than the actual one. Yet, in order to compare the results, we have made the same assumptions as Broecker *et al* (1985): 1) the integration is performed between the $\Delta^{14}C$ *vs* depth curves, 2) the same prenuclear surface water $\Delta^{14}C$ values were used, 3) tritium-free waters are also assumed to be free of bomb ^{14}C.

The inventory errors are on the order of 10-15% for the β-counting $\Delta^{14}C$ profiles and roughly 15-20% for the AMS data (Bard *et al*, 1988). Unfortunately, only the GEOSECS tritium measurements are available at present and our conclusions about the $\Delta^{14}C$ decline with depth cannot be checked with INDIGO tritium data.

OCEANOGRAPHIC IMPLICATIONS

The ^{14}C data demonstrate that during the last eight years, stations between 50°S and 15°S experienced limited changes in $\Sigma^{14}C$ (<30%), and that a broad maximum of $\Sigma^{14}C$ values is still centered at ca 30°S (Fig 3A). By contrast, the bomb ^{14}C water column inventories of the equatorial stations have roughly doubled during the period between the two oceanographic expeditions. Moreover, the $\Delta^{14}C$ profiles obtained for all the locations between 10°S and the equator suggest that there was significant ^{14}C penetration at intermediate depths between 200 and 500m, resulting in the rough doubling of the mean penetration depth, Z, clearly illustrated in Figure 3B (*cf* Östlund & Grall, 1988; Bard *et al*, 1987, 1988).

Based on hydrologic considerations, Bard *et al* (1988) concluded that neither a direct $^{14}CO_2$ diffusion nor a simple advection of water from the south could be responsible for the $\Sigma^{14}C$ and Z doubling during the last eight years in the equatorial zone. The maximum of additional bomb ^{14}C penetration occurred principally at intermediate depths between 300 and 400m, under the influence of westward-flowing waters between the equator and 10°S. This water mass is characterized by relatively low salinity and relatively high oxygen content (Bard *et al*, 1988; the hydrographic data were obtained by the group of physical oceanography of LODYC as part of the INDIGO program). This prominent hydrologic feature is thought to be formed by influx of relatively fresh and oxygenated water from the Pacific through the Banda and Timor Seas. As described by Sharma (1972), Wyrtki (1973) and more recently Gordon (1986), a huge tongue of low salinity layer can be followed throughout the Indian Ocean between 100 and 500m depth, flowing between the equator and 10°S, all the way from the Indonesian archipelago to north Madagascar. As discussed by Sharma (1972), the rela-

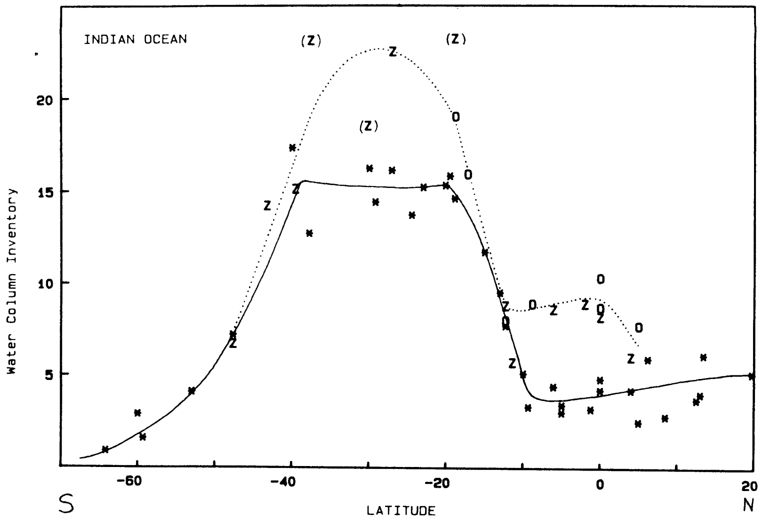


Fig 3A. Plots of water column bomb ^{14}C inventories, expressed in 10^9 atoms/cm^2, as a function of latitude

* = GEOSECS $\Sigma^{14}C$ (1978; β counting)

0 = INDIGO $\Sigma^{14}C$ (leg II, 1986; AMS)

Z = INDIGO $\Sigma^{14}C$ (leg I, 1985 and leg II, 1986; β counting)

__ = GEOSECS level (1978)

.... = INDIGO level (1985–86)

Parentheses indicate when inventories are only given tentatively due to scarcity of the data of the INDIGO profile or to difficulties in reconstructing a prebomb level (no nearby GEOSECS station) (*cf* Bard *et al*, 1988).

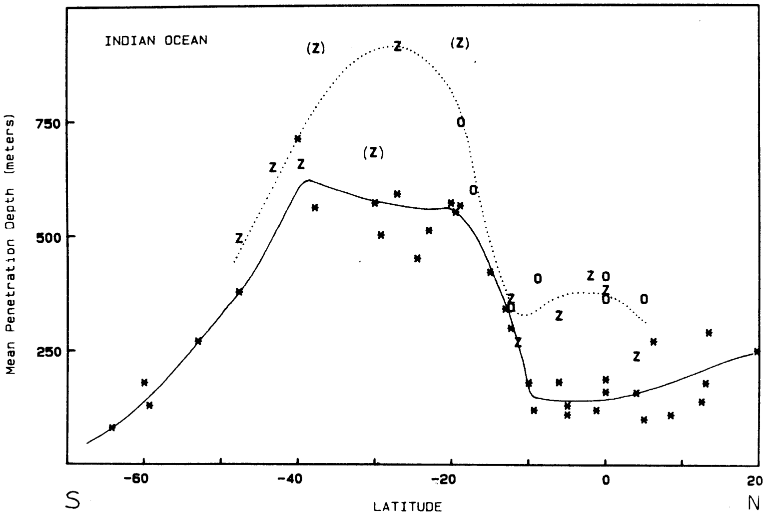


Fig 3B. Plots of mean penetration depth as a function of latitude. For conventions see Fig 3A.

tively high oxygen concentration is enhanced through mixing with high oxygen concentration water from the south during the flow to the west.

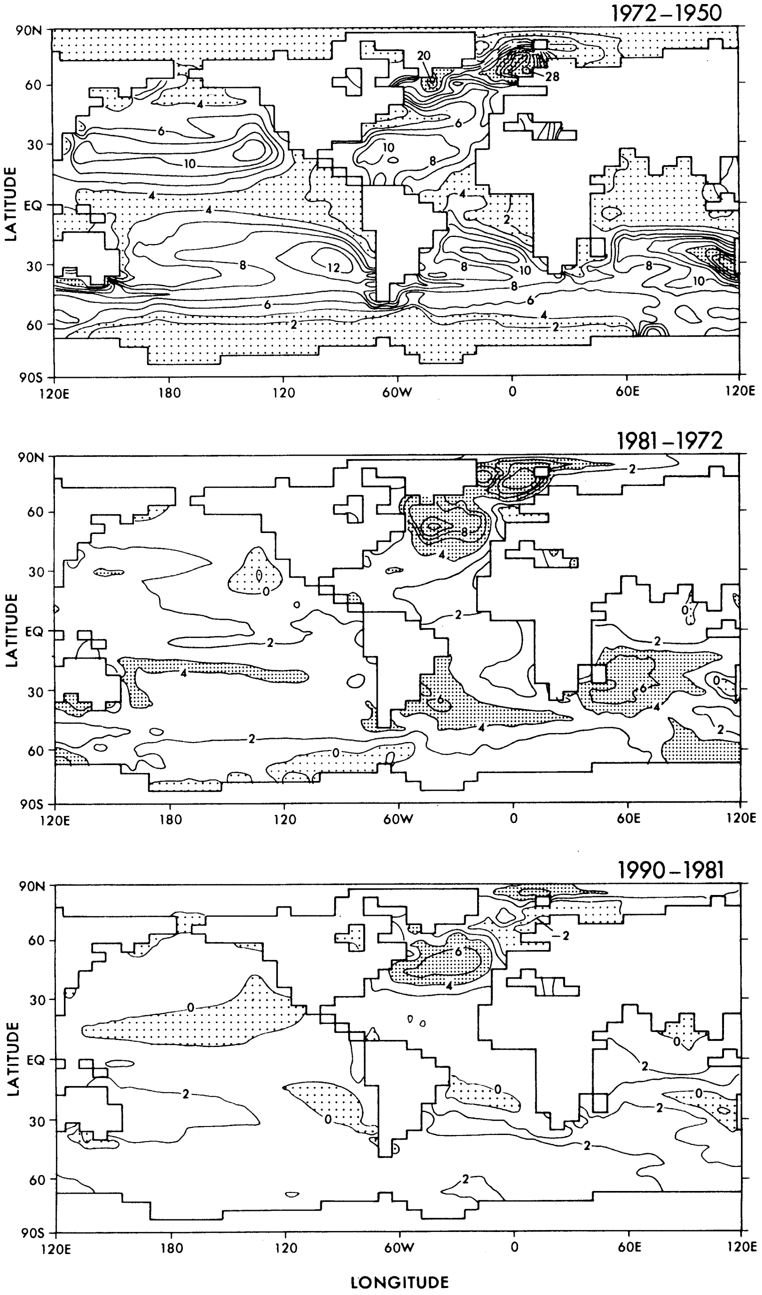
Although it is clear that in the western Indian Ocean, the additional bomb ^{14}C is associated with these westward flowing waters, it is still difficult to pinpoint the ultimate source of the bomb tracer. Indeed, two hypotheses have been proposed (Bard *et al*, 1988): 1) direct provenance from the Pacific mid-latitudes, 2) mixing with Mode Water from the southern gyre of the Indian Ocean.

Modeling results have been obtained using an oceanic general circulation model (GCM built at GFDL, Princeton University). It is a primitive equation world ocean GCM which has a 4.5° Lat, 3.75° Long grid, 12 vertical levels and realistic continental and bottom topography. The model in its prognostic form is forced at the surface by observed, annually-averaged temperature, salinities and wind stresses. For a description of the model, see Bryan & Lewis (1979) and for simulations of the natural and bomb ^{14}C, Toggweiler *et al* (1988a,b).

Figure 4 shows the modeled water column inventories which followed the bomb tests. For the Indian Ocean, latitudinal variations of the modeled inventories agree with the measured variations of Figure 3A; the values between 10°N and 10°S and south of 40°S are typically lower than those for the zone between 20°S and 40°S. Figure 5 presents the time-varying bomb ^{14}C penetration at intermediate depth (level 6:755m) for the Southern Hemisphere. Figure 6 shows Indian Ocean maps of $\Delta^{14}C$ calculated for the year 1990 and horizontal velocity maps for two levels in the model (Fig 6A, level 2: 85m, Fig 6B, level 6: 755m). Both Figures 5 and 6B clearly demonstrate that in the model the penetration of bomb ^{14}C into intermediate depths of the mid-latitude zone of the Indian Ocean, is linked to the westward advection of "Mode Waters" (McCartney, 1982) (in the GCM this water mass is not strictly equivalent to Mode Water since there is no seasonal forcing; nevertheless, the model produces convections in areas of low vertical stability (Toggweiler *et al*, 1988b)). For levels 5 to 7 in the model (respectively 483m, 755m, and 1131m), this "Mode Water" circulation can account for the additional penetration of bomb ^{14}C measured in the equatorial zone.

By contrast, it appears that there is indeed substantial throughflow between the Pacific and Indian Oceans in level 1-4 of the model (respectively, 25m, 85m, 170m, 295m). Consequently, Pacific water may contribute significantly to the Indian Ocean bomb-^{14}C inventories in these levels. The advection of Pacific water and bomb ^{14}C into the Indian Ocean is shown for level 2 (85m) in Figure 6A. These results agree well with the work of Toggweiler and Trumbore (1985) who showed that high bomb ^{90}Sr concentrations in banded corals from the west Indian Ocean can be explained by Pacific water influx through the Indonesian archipelago.

As the $^{3}H/^{14}C$ ratio is far higher in Northern than Southern Hemisphere waters, the INDIGO tritium data would allow further limits to be placed on the origin of the bomb ^{14}C entering the equatorial zone of the western Indian Ocean. Additional oceanographic cruises are also needed in the east Indian Ocean to build a three-dimensional picture of the additional bomb ^{14}C penetration in the Indian Ocean.


1972–1950

1981–1972

1990–1981

Fig 4. Water-column inventories of bomb ^{14}C calculated with the GFDL model. 4A: 1972; 4B: 1981 minus 1972, 4C: 1990 minus 1981. Heavily shaded areas indicate maxima (for 4A: $> 16.10^9$ atoms/cm^2, for 4B and 4C: $>4.10^9$ atoms/cm^2); lightly shaded areas indicate minima (for 4A: $< 4.10^9$ atoms/cm^2, for 4B and 4C: <0).

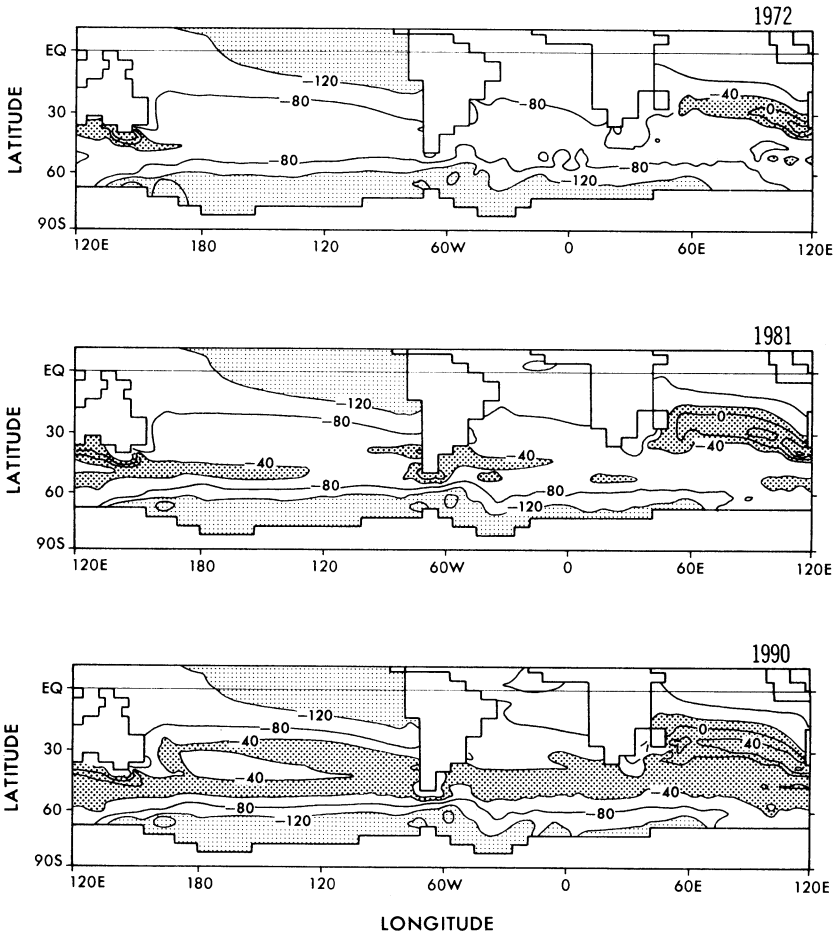


Fig 5. $\Delta^{14}C$ calculated with the GFDL model at intermediate depth (level 6: 755m). (5A: 1972; 5B: 1981; 5C: 1990). Heavily shaded areas indicate maxima ($\Delta^{14}C > -40$°/₀₀); lightly shaded areas indicate minima ($\Delta^{14}C < -120$°/₀₀).

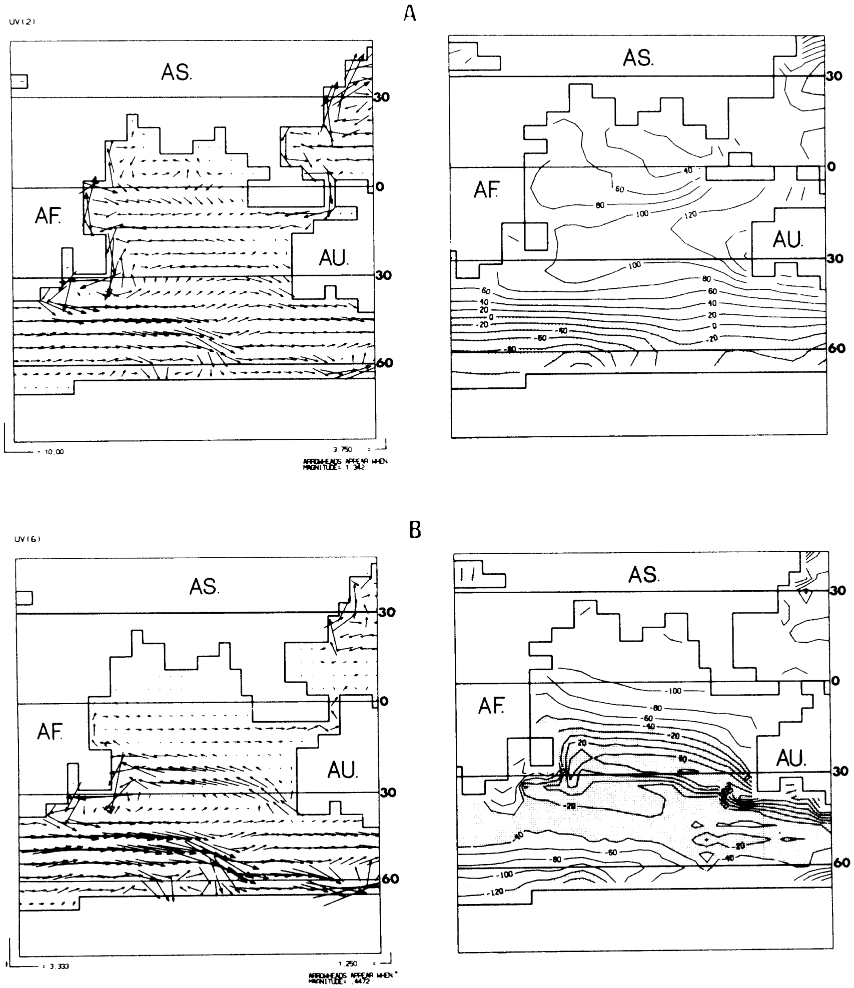


Fig 6. Δ^{14}C for the year 1990 (right) and horizontal velocities (left) calculated with the GFDL model. (Fig 6A: level 2, 85m; Fig 6B: level 6, 755m). Heavily shaded areas indicate maxima $\Delta^{14}\mathrm{C} > -40‰$); lightly shaded areas indicate minima ($\Delta^{14}\mathrm{C} < 0‰$ for 6A and $< -120‰$ for 6B). The arrowheads are proportional to the horizontal velocities and appear when the magnitude is > 1.342cm/sec for level 2 and > 0.4472cm/sec for level 6. The brackets at lower left and lower right beyond the frames scale the arrowshafts in cm/sec. The vector arrows for level 6 have been scaled up by a factor 3 over the scaling used for level 2.

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