BOMB-PRODUCED CARBON-14 IN THE SURFACE WATER OF THE PACIFIC OCEAN

TIMOTHY W LINICK

Mt Soledad Radiocarbon Laboratory, Department of Chemistry, University of California, San Diego, La Jolla, California 92093

ABSTRACT. The distribution of ${}^{14}\text{C}$ concentrations in the dissolved inorganic carbon in the surface waters of the Pacific Ocean is shown to have a primarily latitudinal pattern with $\Delta^{14}\text{C}$ maxima at mid-latitudes in both hemispheres and a minimum at the equator. Oceanographic causes of this phenomenon are discussed.

INTRODUCTION

From 1957 through 1972, the La Jolla Radiocarbon Laboratory collected surface seawater samples from the Pacific Ocean to measure the radiocarbon concentration in the dissolved inorganic carbon. The results of these analyses have been published by Linick (1978). These $^{14}\mathrm{C}$ measurements contribute significant information about the rate of uptake of bomb-produced $^{14}\mathrm{C}$ by the oceans and about ocean mixing processes. The post-bomb pattern of $\Delta^{14}\mathrm{C}$ variation in the surface Pacific may be explained by different modes of water transport.

METHODS

Surface seawater samples were collected mainly on expeditions of research vessels of the Scripps Institution of Oceanography. Water was generally obtained from the ship's non-contaminating seawater pumping system, with water coming from a few meters sub-surface at the ship's bow. A 55-gallon polyethylene-lined steel drum was filled with seawater to within a few inches of its top. The dissolved inorganic carbon was extracted aboard the ship. The extraction system used in the early to mid-1960's is diagrammed in Bien and Suess (1967). The system used from the late 1960's on is shown in figure 1. This is a closed system in which the small amount of air trapped above the seawater is used to strip the CO₂ from the seawater. One 9-pound bottle of concentrated H₂SO₄ solution was added to the seawater, the caps containing the secondary diffuser tube, the immersion heater, and gas outlet were inserted, and the tubing was attached. The small amount of air at the top of the drum was then recirculated through a bottle containing CO₂absorber solution (15M NH₄OH made ca 1M in SrCl₂ to precipitate the carbonate) and then bubbled through the drum of heated, acidified water. The gas circulation was accomplished by using a Masterflex peristaltic pump (using Tygon tubing) with 1550 ml/min output pump head. The water was heated to ca 50°C. By extracting for at least 4 hours, the vast majority of CO₂ was removed. The absorber bottle only was returned to the laboratory, where the solution was decanted and the SrCO₃ precipitate was dried using vacuum and heat. CO₂ was evolved on the high vacuum preparation lines by addition of 2M HCl to the precipitate. The CO₂ was converted to acetylene, the counting gas, via Li₂C₂. Sample activities were measured using the detectors and methods described in Linick (1977). Δ¹⁴C values were calculated relative to 95

percent of 1950 NBS oxalic acid activity (normalized to δ^{13} C of -19%), with sample activities normalized to δ^{13} C of -25% PDB.

RESULTS

A high percentage of samples was collected from central longitudes of the Pacific Ocean, ca 150° W. However, since the tracks of the collection ships were selected for other studies, samples were collected from most other areas of the Pacific as well. The data published in Linick (1978) shows that, due to uptake of ¹⁴C produced by atmospheric nuclear weapons testing, surface seawater Δ^{14} C values rose steadily from 1957 through 1967-1968 and leveled off during the following years. Similar results were found for ¹⁴C in annual coral rings from The Rocks reef off the Florida keys (Druffel and Linick, 1978). From the earliest time of input of significant amounts of bomb-produced ¹⁴C, a strong latitudinal pattern of variation of surface water Δ^{14} C was evident. In Linick and

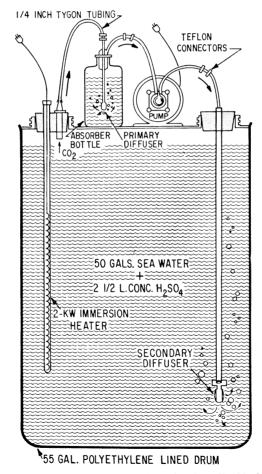


Fig 1. Closed system for shipboard extraction of carbon dioxide from seawater.

Suess (1972), plots of the data points as latitude versus Δ^{14} C were given for the years 1957-1959, 1968, 1969, and 1970 (partial data set). The data used there, as well as in the following discussion, is from this laboratory and the New Zealand Laboratory, measured for the southern hemisphere (Rafter, 1968; Rafter and O'Brien, 1970; 1972). Relatively little variation with longitude along a line of latitude is observable in those cases in which samples were collected on an essentially east-west track (Linick, 1975); the only observable feature of the variation is a slight decrease in Δ^{14} C as the American coastlines are closely approached.

Figure 2 shows the variation of Δ^{14} C with latitude for the central Pacific for all years of sample collection from 1957 through 1972. Samples were not available from a wide latitude range for all years. Very strong maxima are observed at mid-latitudes in both hemispheres with the peaks centered at ca 30° N and S. By the last years of collection, the

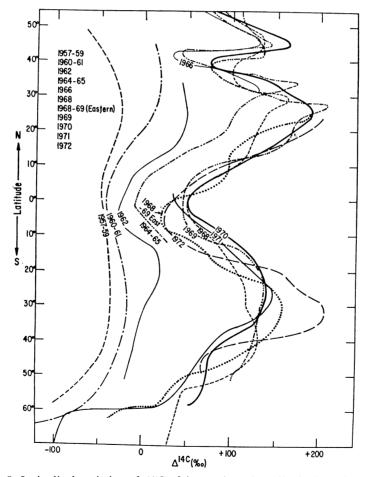


Fig 2. Latitudinal variation of Δ^{14} C of inorganic carbon dissolved in the surface waters of the central Pacific Ocean: 1957 through 1972.

maxima had Δ^{14} C's of about +210%, or approximately 260% above pre-bomb levels. The maximum levels appear to have been reached one or two years earlier in the northern hemisphere than in the southern hemisphere, probably because of the asymmetrical input of bomb 14 C to the atmosphere. 14 C levels remained much lower near the equator than at mid-latitudes. The highest Δ^{14} C values near the equator were ca +50% in 1968-1970, corresponding to an increase of only ca 110% from pre-bomb levels; a decrease from that level is seen in 1971 and 1972. More complex structure is found in the curves at high northern latitudes than at high southern latitudes.

Perhaps the clearest method of expressing variations in Δ^{14} C of surface Pacific seawater is a map indicating lines of constant Δ^{14} C. Such a plot is presented in figure 3. An earlier version of this plot was given by Linick and Suess (1972). Figure 3 is based on all La Jolla and New Zealand Laboratory measurements for samples obtained in 1969-1972, years in which the high bomb 14 C concentrations had leveled off near their maximum values. This map clearly shows the maxima centered in

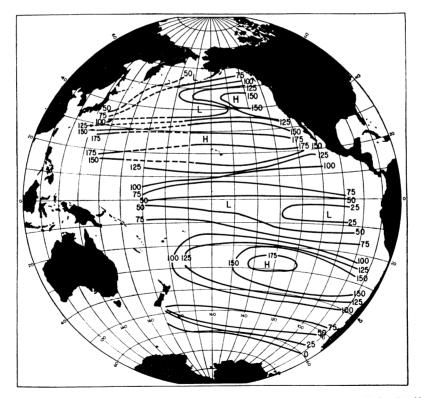


Fig 3. Distribution of Δ^{14} C values in the surface water carbonate of the Pacific Ocean: Isolines of Δ^{14} C drawn from measurements of samples obtained in 1969 to 1972. Solid lines based on actual measurements, broken lines estimated based on data from earlier years or on oceanographic conditions.

the North and South Pacific gyral circulation systems, the minimum at the equator, and other features, to be discussed below, at higher latitudes.

DISCUSSION

Both horizontal and vertical mixing processes can affect the ¹⁴C concentration in the surface water. However, because sub-thermocline water generally has a ¹⁴C level much lower than that of the surface water, vertical mixing processes are usually more evident from surface ¹⁴C measurements. The degree of upwelling of deeper, ¹⁴C-depleted water to the surface is the most important factor in the variability of surface ¹⁴C concentrations. Areas of "divergence," where the flow of near-surface water is away from that region in two or more directions, lead to the upwelling of deeper water. The stability of the surface (mixed) layer and the strength of the thermocline affect the degree of cross-thermocline mixing.

The effectiveness of this barrier may be examined by measuring physical and chemical properties of the water. The dissolved CO₂ partial pressure (p_{CO₂}) relative to equilibrium with the atmosphere is influenced by several factors (Keeling, 1968). Surface p_{CO₂} is increased by upwelling of the usually higher CO₂ deep water and by the rapid warming of upwelled (or otherwise-added) cold water. Photosynthesis by marine organisms uses CO₂ and, thus, decreases p_{CO₂}. The inorganic phosphate–phosphorus (PO₄–P) concentration is lowest at the surface, reaches a maximum several hundred meters down, and declines slightly at greater depths (Reid, 1962). Upwelling brings water of higher PO₄–P concentration to the surface.

Another important factor affecting surface ¹⁴C concentrations is horizontal surface and sub-surface currents. Currents can carry water of a certain ¹⁴C concentration away from its source, affecting ¹⁴C levels along the path of flow.

In the central North Pacific at latitudes from 15° to 35° N, the highest surface Pacific 14 C levels are found. Here, in the gyral circulation system, the surface layer is 100 to 200m thick, and the thermocline is strong. The center of the gyre has a 60ppm (19 percent) deficit in p_{CO_2} relative to equilibrium, with all p_{CO_2} 's being below equilibrium values in this area (Keeling, 1968). In addition, the PO₄–P concentrations in this region are generally less than $0.3\mu g$ -atoms/liter (compared to some other areas with concentrations as high as more than $2.0\mu g$ -atoms/liter) (Reid, 1962). These findings are indicative of a lack of vertical mixing.

A minimum ¹⁴C band is found at 38° to 42° N, a narrow band associated with salinities from 33.60 to 34.15‰. This band corresponds to the boundary region between the Subarctic and Central Water masses. No evidence for substantial vertical mixing is found in pco₂ or PO₄–P concentration measurements. The region is dominated by eastward-flowing water fed from the Kamchatka Current (Oyashio) from the North and the Kuroshio from the South, both bringing water of lower ¹⁴C than the Central Water masses. The ¹⁴C maximum found at 45° to 48° N is

associated with the southern Subarctic Current and is somewhat north of the Subarctic front. Apparently, the surface water in this band is either more isolated from deeper water or is not significantly fed by low ¹⁴C waters. To the north, ¹⁴C concentrations decrease. High PO₄–P (0.5 to 2.0µg-atoms/liter) are found, the winter thermocline is relatively permeable, and the waters are convectively-overturned to great depths by harsh meteorologic conditions.

The general 14C minimum along the equator is quite symmetric about 0°, with low values also found significantly south of the equator at the eastern end. The equatorial Pacific has a well-mixed surface layer 10 to 150m thick, with the thermocline deepest in the west, very shallow in the east (Knauss, 1963). In the central region, there is an area of surface convergence at 5° N, with divergences at 0° and 10° N (Sverdrup, Johnson, and Fleming, 1942). Cromwell (1953) and Rotschi (1970) found evidence of upwelling and divergence along the equator. The equatorial current system is quite complex: the strong North Equatorial Current (10°-20°N) flows to the west fed by the California Current and gyral waters; the weaker Equatorial Countercurrent (5°-8°N) flows easterly and the strong South Equatorial Current (4°N-10°S) flows to the west fed by Peru Current water plus a small amount of Equatorial Countercurrent water. The eastward-flowing Cromwell Current is found 100 to 400m sub-surface at the equator and is a prime factor in vertical mixing (Cromwell, 1953). The strength of the Cromwell Current (Pacific Equatorial Undercurrent) and the degree of upwelling are related. The Peru Current, an area of substantial upwelling (Wooster and Reid, 1963), especially in winter, flows northwesterly into the westward-flowing South Equatorial Current, further decreasing Δ^{14} C values along the equator. The highest excess pco, relative to equilibrium found in the surface Pacific (excesses of more than 90ppm or 29 percent) are found from 0° to 7° S for over 3000km west of the South American coast, with excesses of 30 to 60ppm found within 3° of the equator even at 160° E.

The 14 C maximum in the mid-latitudes of the South Pacific has the same causes as that at 15° to 35° N. A gyral current system dominates, and low p_{CO_2} and low PO_4 –P concentrations are found; there is little upward mixing.

South of 35° S, Δ^{14} C values decrease monotonically, with no complex variation as found at high northern latitudes. South of the Antarctic Convergence (at 55° to 60° S), Δ^{14} C decreases rapidly, with very high (more than 2.0 μ g-atoms/liter) PO₄–P concentrations in a wide band at 60°S. Upwelled water occurs to the south, with the Antarctic Divergence found at 65° to 70° S, caused by east-northeast water flow from 40° to 60°S and west-southwest water flow to the south (Deacon, 1963).

Recommendations for future work

Although desirable, collecting depth profiles at several locations in the ocean every few years would require substantial funding. However, collecting surface samples from a larger area of the Pacific every two to three years would be very worthwhile. This would allow a better understanding of the rates of transfer of CO₂ among reservoirs and of the oceanographic processes than is now possible.

ACKNOWLEDGMENTS

This paper is based on the author's doctoral dissertation (Linick, 1975). Great credit must go to the late G S Bien, who collected, prepared, and measured many of the samples. C S Hutto and her predecessors prepared most of the samples, which were collected by too many people to thank individually. D K Sullivan designed and maintained the electronic equipment. Useful discussions were held with J L Reid and R L Michel. Of course, great thanks go to H E Suess, Director of this Laboratory. This work was funded by US National Science Foundation Grant EAR78-15183 and its predecessor grants, with H E Suess as Principal Investigator.

REFERENCES

- Bien, G S and Suess, H E, 1967, Transfer and exchange of ¹⁴C between the atmosphere and the surface water of the Pacific Ocean, *in* Radioactive dating and methods of low level counting: Vienna, IAEA, p 105-115.
- Cromwell, T, 1953, Circulation in a meridional plane in the Central Equatorial Pacific: Jour Marine Research, v 12, p 196-213.
- Deacon, G E R, 1963, The southern ocean, in Hill, M N, ed, The Sea, vol II: New York, Wiley (Interscience), p 281-296.
- Druffel, E M and Linick, T W, 1978, Radiocarbon in annual coral rings of Florida: Geophys Research Letters, v 5, p 913-916.
- Keeling, C D, 1968, Carbon dioxide in surface ocean waters, 4. Global distribution: Jour Geophys Research, v 73, p 4543-4553.
- Knauss, J. A., 1963, Equatorial current systems, in Hill, M. N., ed, The Sea, vol II: New York, Wiley (Interscience), p. 235-252.
- Linick, T. W., 1975, Uptake of bomb-produced carbon-14 by the Pacific Ocean: PhD dissert, Univ California, San Diego, 255 p.
- 1977, La Jolla natural radiocarbon measurements VII: Radiocarbon, v 19, p 19-48.
- 1978, La Jolla measurements of radiocarbon in the oceans: Radiocarbon, v 20, p 333-359.
- Linick, T W and Suess, H E, 1972, Bomb produced radiocarbon in the surface water of the Pacific Ocean, in Rafter, T A and Grant-Taylor, T E, eds, Internatil conf on radiocarbon dating, 8th, Proc: Wellington, New Zealand, Royal Soc New Zealand, p C87-C93.
- Rafter, T A, 1968, Carbon-14 variations in nature, part 3—14C measurements in the South Pacific and Antarctic Oceans: New Zealand Jour Sci, v 11, p 551-589.
- Rafter, T A and O'Brien, B J, 1970, Exchange rates between the atmosphere and the ocean as shown by recent C-14 measurements in the South Pacific, *in* Olsson, I U, ed, Radiocarbon variations and absolute chronology, Nobel symposium, 12th, Proc: New York, John Wiley & Sons, p 355-377.
- 1972, ¹⁴C measurements in the South Pacific Ocean—a recalculation of the exchange rates between the atmosphere and the ocean, in Rafter, T A and Grant-Taylor, T E, eds, International conf on radiocarbon dating, 8th, Proc: Wellington, New Zealand, Royal Soc New Zealand, p C17-C42.
- Reid, J. L., 1962, On the circulation, phosphate-phosphorus content and zooplankton volumes in the upper part of the Pacific Ocean: Limnol Oceanog, v 7, p 287-306.
- Rotschi, H, 1970, Variations of equatorial currents, in Wooster, W S, ed, Scientific exploration of the South Pacific: Washington, DC, Natl Acad Sci, p 75-83. Sverdrup, H U, Johnson, M W, and Fleming, R H, 1942, The oceans, their physics,
- Sverdrup, H U, Johnson, M W, and Fleming, R H, 1942, The oceans, their physics, chemistry, and general biology: Englewood Cliffs, New Jersey, Prentice-Hall, 1087 p.
- Wooster, W S and Reid, J L. 1963, Eastern boundary currents, in Hill, M N, ed, The Sea, vol II: New York, Wiley (Interscience), p 253-280.

DISCUSSION

Robinson: The GEOSECS Intercalibration Station at about 28.5°N, 121.5°W was sampled for ¹⁴C in both 1969 and 1973. While your data and the data for Druffel indicate that surface ¹⁴C values have largely leveled off since the late 1960's, this station dropped from ca +265 to +190‰ during this interval. The station is about 600km from the coast and probably beyond the effects of coastal upwelling. Do you have any ideas about the interpretation of this effect?

Linick: If one assumes that the 1969 sample was not contaminated, the likely cause of the high Δ^{14} C of it would seem to be the local, temporary formation of a lens of water isolated from the lower ¹⁴C below. It may be possible for such conditions to occur on a scale of days to months.