[RADIOCARBON, VOL 29, NO. 1, 1987, P 18–23]

RADIOCARBON CONTENT OF TROPOSPHERIC CO_2 AT CHINA LAKE, CALIFORNIA 1977–1983

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ABSTRACT. The measurements reported here are a continuation of tropospheric radiocarbon measurements in carbon dioxide carried out since 1961 at our China Lake, California collection facility. The data show a continued decrease in radiocarbon activity from ca 330‰ in 1977 to 215‰ in 1983 in agreement with similar analyses in Europe for the same time interval.

INTRODUCTION

The measurements reported are part of a continuing study of the ^{14}C content of tropospheric CO_2 in the Northern Hemisphere. The program began in 1961 at China Lake, California (35° 32′ N, 117° 41′ W). The sampling location is exposed to clean maritime air in the summer while in the winter there can be episodes of incursions of continental air via Santa Ana winds. The site was chosen to avoid contaminated air from anthropogenic sources such as the metropolitan Los Angeles area or other major producers of fossil fuel emissions. Results of measurements of prior years were reported previously (Berger & Libby, 1966, 1967, 1968; Berger, Fergusson & Libby, 1965).

In the early 1960s, a pronounced maximum in the ^{14}C content of CO_2 in the air occurred approaching a factor of two above the prebomb level. Since then, the ^{14}C content has steadily decreased, due to the uptake of bomb ^{14}C primarily by the world's oceans (Keeling, 1979). This decrease is interesting because it provides information on residence time of CO_2 in the atmosphere and allows the dating of relatively young plant material.

Tropospheric carbon dioxide undergoes exchange with three main reservoirs on time scales of interest to mankind: the stratosphere, the biosphere, and the oceans. Most bomb ^{14}C is injected into the stratosphere (Berger, 1979). Hence, only mixing with stratospheric CO_2 can increase ^{14}C in the troposphere. The oceanic reservoir is a complex system containing the bulk of bomb-produced ^{14}C; it is actually a series of interrelated reservoirs with varying time scales for exchange. Most of the upper layers equilibrate with the ^{14}C of the bomb transient on a decadal time scale. The time scales for equilibration with most deeper waters are longer, on the order of centuries (Linick, 1980; Bien, Rakestraw & Suess, 1965).

METHODS

Samples were collected in a photographic tray ca 20 x 40 x 6cm containing 2L of 2N carbonate-free sodium hydroxide. This tray is located inside a wire-mesh-protected "bird-house" remote from any contaminating sources of anthropogenic fossil fuel emissions. The height of the collection

station is 2m above ground level to reduce the amount of dust collected. The unexposed sodium hydroxide solution, as well as the exposed mixture, is shipped in tightly sealed plastic laboratory bottles. Exposure time to atmospheric air is typically one week.

In the laboratory, the contents of the plastic bottles are transferred to a round-bottomed flask of 5L capacity. CO_2 is gradually liberated by the addition of dilute hydrochloric acid. CO_2 produced in the above manner is converted to acetylene by reaction with molten lithium metal to produce Li_2C_2, followed after cooling, to room temperature by hydrolysis with tritium-free well water. The acetylene is purified for counting by passage through traps of dry ice, traps filled with glass balls coated with P_2O_5, and traps filled with activated charcoal at 0°. Samples were allowed to age for at least 3 weeks to allow any radon present to decay before radiocarbon analysis. After ^{14}C assay, a small sample of the acetylene was recombusted to CO_2 without isotopic fractionation by adding O_2 and circulating over CuO at 600°C. The $\delta^{13}C$ of the CO_2 was then measured mass spectrometrically (Linick, 1977).

RESULTS AND DISCUSSION

Results reported here are based on 95% of the NBS oxalic acid activity normalized to $\delta^{13}C = -19‰$ (PDB). All sample activities have been normalized to $\delta^{13}C = -25‰$ (PDB). Mass spectrometric $\delta^{13}C$ measurements were made on all samples and standards using, as previously, CO_2 prepared by recombusting an aliquot of the acetylene counting gas sample. Two of the gas proportional counters described by Linick (1977, 1979) were used, *ie,* 2L quartz counters.

The data from our measurements are given in Table 1 and shown in Figure 1. The continuing decrease following the ^{14}C peak produced by the bomb transient is evident from the data. The e-folding time for the excess $^{14}CO_2$ is found to be 14–15 years. This is very similar to the time scale found from data prior to our sampling period. As noted, the time scale results from a series of complex interactions between the earth's various reservoirs, as well as *in situ* production.

From Figure 1, a seasonal structure in tropospheric $^{14}CO_2$ is seen to be superimposed on the basic decrease. A small increase in ^{14}C appears to occur about the middle of each year, which is barely detectable above the downward trend. Then a major decrease occurs each winter. This structure is due to a combination of atmospheric and oceanic effects. Mixing between the stratosphere and troposphere is not constant, but reaches a maximum around April of each year. This phenomenon is known as the "spring leak." It introduces excess $^{14}CO_2$ each year. The effect is compounded in the oceans by the warming of surface waters which increases stratification and inhibits mixing of $^{14}CO_2$ into deeper waters resulting in a few per mil increase in the atmosphere. Thus, a combination of these processes can produce a small $^{14}CO_2$ maximum in the northern hemispheric summer. During the winter the thermocline deepens, resulting in an increased uptake of atmospheric ^{14}C and a more rapid decrease in the tropospheric level of $^{14}CO_2$. Inasmuch as the sampling location is very iso-

TABLE 1
^{14}C and ^{13}C measurements from China Lake, California

La Jolla no.	UCLA no.	Date sampled	$\delta^{13}C$	$\Delta^{14}C$
5729	2600	1/14–1/21/77	−22.99	325.7 ±3.5
5728	2601	2/25–3/4/77	−24.66	318.8 ±3.4
5739	2602	3/28–4/4/77	−24.61	340.0 ±7.0
5730	2603	4/27–5/4/77	−23.51	323.5 ±3.5
5737	2604	6/13–6/20/77	−22.85	323.9 ±3.4
5732	2605	7/31–8/7/77	−21.84	330.2 ±3.3
5731	2606	8/29–9/5/77	−21.82	321.1 ±3.5
5734	2607	9/22–10/4/77	−22.45	327.5 ±3.2
5733	2608	10/28–11/4/77	−22.93	322.9 ±3.3
5735	2609	11/27–12/4/77	−23.08	311.8 ±3.3
5736	2610	12/28–1/4/78	−23.10	312.4 ±3.3
5727	2611	5/23–5/30/78	−22.83	322.0 ±3.5
5726	2612	7/5–7/12/78	−22.05	321.7 ±3.5
5725	2613	7/28–8/4/78	−22.01	310.9 ±3.4
5724	2614	8/28–9/4/78	−22.01	309.9 ±3.3
5738	2615	9/27–10/4/78	−21.46	301.1 ±3.4
5722	2616	10/28–11/4/78	−23.33	309.1 ±3.3
5741	2617	2/12–2/19/79	−24.87	302.4 ±7.0
5740	2618	3/12–3/19/79	−25.71	291.4 ±6.8
5704	2619	5/28–6/4/79	−23.09	274.1 ±6.9
5706	2620	7/4–7/11/79	−23.23	282.1 ±3.3
5719	2621	9/1–9/8/79	−22.32	285.9 ±3.4
5700	2622	9/28–10/5/79	−22.22	279.0 ±3.6
5721	2623	10/28–11/4/79	−23.51	284.5 ±3.4
5718	2624	11/28–12/5/79	−23.82	264.5 ±3.3
5723	2625	12/29–1/5/80	−23.86	269.3 ±3.3
5686	2626	4/1–4/8/80	−24.89	267.8 ±3.9
5688	2627	4/27–5/4/80	−23.03	266.4 ±3.4
5690	2628	5/28–6/4/80	−23.06	263.7 ±3.5

https://doi.org/10.1017/S0033822200043526 Published online by Cambridge University Press

TABLE 1 (continued)

La Jolla no.	UCLA no.	Date sampled	$\delta^{13}C$	$\Delta^{14}C$
5692	2629	6/27–7/4/80	−21.64	273.4 ±3.6
5694	2630	7/29–8/5/80	−21.78	263.5 ±3.1
5696	2631	8/28–9/4/80	−21.95	270.7 ±3.6
5698	2632	9/27–10/4/80	−21.63	260.7 ±3.9
5702	2633	10/28–11/4/80	−23.14	257.7 ±3.4
5720	2634	11/27–12/4/80	−23.88	255.1 ±3.6
5711	2635	2/25–3/4/81	−24.38	244.2 ±3.1
5710	2636	3/28–4/4/81	−21.90	243.3 ±3.2
5716	2637	7/28–8/4/81	−23.63	250.2 ±3.2
5717	2638	8/28–9/4/81	−21.92	250.9 ±3.4
5673	2639	2/5–2/12/82	−24.66	241.8 ±7.2
5674	2640	3/1–3/8/82	−24.57	242.6 ±2.9
5675	2641	3/28–4/5/82	−25.00	233.6 ±3.9
5680	2642	4/27–5/4/82	−23.45	237.9 ±3.5
5681	2643	5/28–6/4/82	−23.65	239.3 ±3.8
5676	2644	6/27–7/4/82	−21.39	225.7 ±3.1
5683	2645	8/4–8/11/82	−22.16	233.9 ±3.6
5682	2646	8/31–9/7/82	−24.04	237.5 ±3.6
5677	2647	9/27–10/4/82	−24.00	233.0 ±3.2
5678	2648	10/28–11/4/82	−23.56	226.9 ±3.6
5679	2649	11/27–12/4/82	−24.96	226.9 ±3.2
5684	2650	12/29–1/5/83	−24.67	212.6 ±3.3
5708	2651	5/18–5/25/83	−24.00	218.1 ±3.1

lated, a seasonal varying influence of fossil fuel CO_2 must be exceedingly small.

In comparison, our radiocarbon results lie well within the date envelope of a similar study for central Europe (Levin *et al,* 1985). Taken together both investigations show that the global troposphere, except for some differences in fine structure, is well mixed between 35° and 48°N latitude on an intercontinental scale for the period of 1977–1983. In addition, information about the tropospheric residence time of CO_2 can be derived

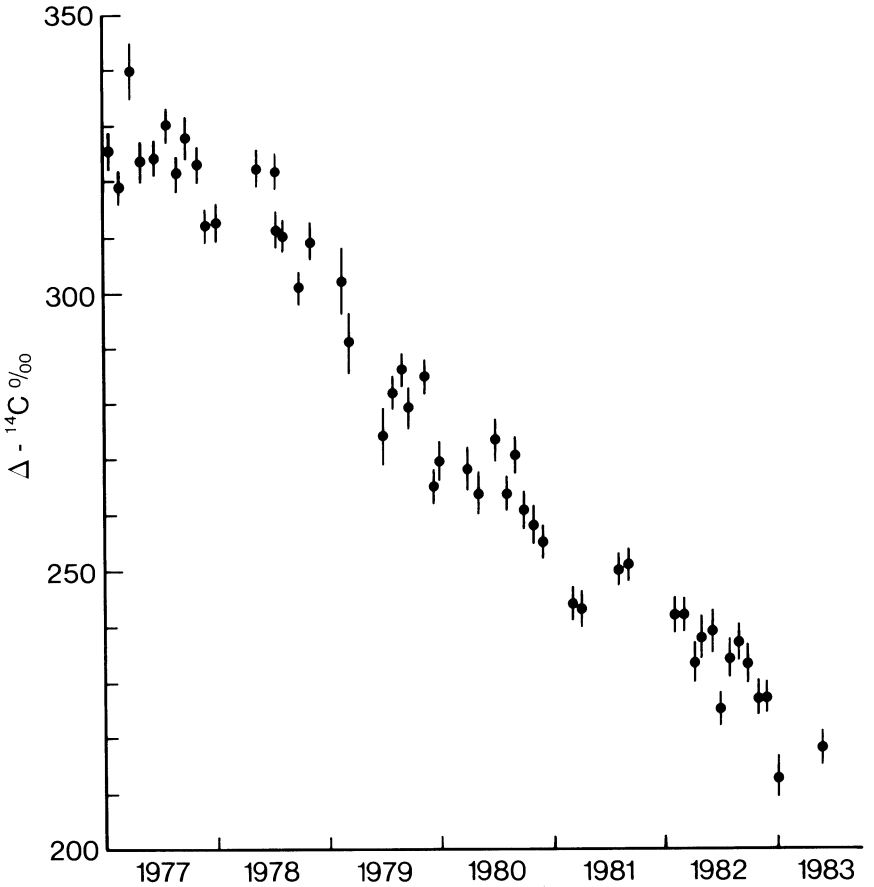

Fig 1. Atmospheric $\Delta^{14}C$ at China Lake, California (35° 32′ N, 117° 41′ W)

through a comparison of the atmospheric data with radiocarbon assays of banded corals (Druffel, ms in preparation).

ACKNOWLEDGMENTS

G Plain collected the China Lake samples and deserves our gratitude. This study was funded in part by the State of California Air Resources Board to provide baseline data. This is publication no. 2635 of the Institute of Geophysics and Planetary Physics, UCLA.

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