# Radiocarbon from Nuclear Tests, II

Future concentrations predicted for this isotope in the earth's carbon cycle suggest its use in tracer studies.

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In the testing of nuclear weapons to date, large amounts of radiocarbon have been synthesized and injected into the earth's carbon cycle, specifically into the atmosphere (1-7). From this reservoir, such radiocarbon makes its way into other carbon reservoirs through numerous mixing processes, with the result that its concentration at various places in the carbon cycle is rising. Thus, scientists are in possession of a large-scale tracer that potentially can elucidate the mechanisms and rates of many natural processes that involve the element carbon. It is the purpose of this article (8) to point out several potential tracer applications of bombproduced carbon-14. In order to evaluate these tracer possibilities it is necessary to have an estimate of the total amount of bomb-produced C14 and to know its present as well as its future distribution in the carbon cycle. Such estimates are given below, along with an evaluation of the experimental data used as the basis for the inventory estimates

# Reliability of Vegetation as an Indicator of Atmospheric Levels

In a previous report (4) on radiocarbon produced during nuclear tests, by Broecker and Walton, it was noted that the  $C^{14}/C^{12}$  ratios in tree-leaf samples collected in Palisades, New York, were significantly lower than those in samples collected at the same time in other areas. In order to determine the reason for this difference, a series of carefully chosen samples collected in the Palisades area during the late spring of 1959 were analyzed. Samples of atmospheric CO<sub>2</sub>, leaves, and soil CO<sub>2</sub> were analyzed. The samples of atmospheric CO<sub>2</sub> were collected by exposing pans of "CO2-free" KOH solution to the atmosphere; the samples of soil CO<sub>2</sub> were collected in a similar way except that the pans were set under an inverted, open-top oil drum driven into the soil. As determined from the quantity of CO<sub>2</sub> collected, the escape rate of CO<sub>2</sub> from the soil was approximately 7 gram molecules per square meter per month. It should be emphasized that this rate depends on both the time of year and the type of soil; (in this case the time was late spring and the soil was of the great soil group, Sol Brun Acide, which has here a humic-rich zone down to about 8 inches and, below that, a yellow subsoil).

The analytical results are given in Table 1, where they are expressed in  $\Delta C^{14}$  units (9) (that is, per-millage difference between the fractionation-corrected value for radioactivity of the sample and 0.95 times the activity of the National Bureau of Standards oxalic acid C<sup>14</sup> standard). Values for samples grown just before bomb-testing began range from -20 to -40 per mill on this scale, and age-corrected values for samples of pre-1900 woods are close to zero.

The two samples of leaves taken from a forest in Palisades (L528A and L528E) have  $\Delta C^{14}$  values about 50 per mill lower than a sample of leaves collected at the same time in Carson City, Nevada (L516E), a difference consistent with findings in earlier comparisons between Palisades and Great Basin samples (4). The difference cannot be explained by a uniformly lower  $C^{14}/C^{12}$  ratio in the air of the Palisades area because values for two samples from a nearby area agree well with the findings for Carson City leaves. One of these samples (L528B) was aircollected atop a 400-foot tower in Alpine, New Jersey, and the other (L528F) was a leaf sample from a lone tree on a golf course at Rockleigh, New Jersey. Furthermore, the low values for the two forest samples cannot be attributed to incorporation by the growing leaves of soil CO2 which had accumulated in the forest air, since a sample of atmospheric  $CO_2$  (L528C) taken within 3 feet of the growing leaves had a value 57 per mill higher than the average for the two leaf samples and had the same ratio as the CO<sub>2</sub> collected atop the 400-foot tower.

Three possible explanations remain: (i) the leaves photosynthesized most of their carbon prior to air sampling, at a time when the Palisades area had a considerably lower atmospheric  $C^{14}/C^{12}$ ratio than it had at the time leaves were collected; (ii) the leaves of certain trees receive a major fraction of their carbon from organic compounds stored for significant periods of time in the tree roots; or (iii) the forest leaves received about 30 percent of their carbon from soil CO<sub>2</sub> taken in by their roots. All three explanations obviously pose the problem of why the Rockleigh sample, taken only 2 miles west of the Palisades samples, does not show a similar effect. Since the value for Rockleigh leaves matches the value for the atmosphere and since none of the leaves in the Palisades area sprouted until early May, the first explanation given above is very unlikely. It is also difficult to believe that in one tree (a maple) a large fraction of the carbon incorporated in leaves came from stored organic materials while in another (a black cherry) only a small fraction came from this source.

The third explanation is qualitatively consistent with work cited by Rabinowitch (10), but whether it is quantitatively possible is another question. Tracer experiments by Kursanov *et al.* (11) indicate that as much as 20 percent of the carbon photosynthesized by bean seedlings can be introduced through the roots. However, further work, by Stolwijk and Thimann (12) casts some doubt on this high percentage. Whether under certain circumstances tree roots can provide a third of all photosynthesized carbon is at present not known.

If appreciable uptake of soil  $CO_2$  through the roots of certain trees is

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accepted as the true explanation, there remains the problem of explaining why significant uptake from the soil has occurred in the case of the Palisades samples and not in the case of the Rockleigh samples. A possible explanation based on the different growth environments is as follows: The Rockleigh tree grows in an open area and in one where deep root penetration is possible; on the other hand, the Palisades tree grows in a densely vegetated area where there are only 2 to 4 feet of soil overlying bedrock (Palisades diabase). Thus, the tree in the Rockleigh area may take up less soil  $CO_2$  because (i) there is less decaying vegetation and hence less soil CO<sub>2</sub> available and (ii) a smaller percentage of its roots are concentrated near the zone of maximum soil CO<sub>2</sub> production.

Certainly more evidence must be obtained before this problem can be solved completely. It can only be said at this point that there is almost certainly a real lag between the  $C^{14}/C^{12}$  ratio in leaves from the Lamont Observatory forest and the ratio in the atmosphere. Even though this may be an unusual case, it shows that monitoring of atmospheric CO2 is most reliably accomplished through direct collection of atmospheric CO2. Since only a few air samples were taken in the Northern Hemisphere prior to 1958, results on vegetation are the only source of information for this period. Their correct evaluation is hence a matter of importance. The information available indicates that vegetation samples are generally reliable indicators of atmospheric  $C^{14}/C^{12}$  ratios but that they may occasionally provide only minimal estimates.

### **Bomb-Produced Carbon-14 Inventory**

Additional samples defining the tropospheric C<sup>14</sup>/C<sup>12</sup> ratio extend Broecker and Walton's curve (4) from March 1958 to June 1959. The additional data, given in Table 2 and Figure 1, show that the rate of rise of the C<sup>14</sup>/C<sup>12</sup> ratio has increased in both hemispheres—from 5 up to 11 percent per year for the Northern Hemisphere and from 4 up to 7 percent per year in the Southern Hemisphere. At the same time, the ratio of the two hemispheric bomb effects has remained about the same (that is, C<sup>14</sup>/C<sup>12</sup><sub>South</sub> = 0.67 C<sup>14</sup>/C<sup>12</sup><sub>North</sub>).

Data published by Willis (2) and by Tauber (3) suggest that the tropo-16 SEPTEMBER 1960

Table 1. Data relating to the reliability of vegetation samples as indicators of atmospheric  $C^{14}/C^{12}$  ratios.

Collection date	Material	Location	Physical surroundings	$\delta C^{14}$	δC <sup>13</sup>	$\Delta C^{14}$	Sample No.
5/16/59 to 6/7/59	Atmospheric CO <sub>2</sub>	Alpine, N.J. 40°57'N, 73°55'W	Top of 400-ft radio tower	251 ± 6	- 18.9	249 ± 6	L528-B
5/16/59 to 6/7/59	Atmospheric CO <sub>2</sub>	Palisades, N. Y. 41°00'N, 73°55'W	3 ft above ground in dense forest	264 ± 7	-16.9	257 ± 7	L528-C
6/5/59	Willow leaves	Carson City, Nev. 39°07'N, 119°53'W	5 ft above ground in a clump of willows	249 ± 8	-23.8	259 ± 8	L516-E
6/9/59	Oak leaves	Rockleigh, N.J. 41°00'N, 73°58'W	On knoll in middle of golf course	232 ± 8	-25.8	<b>246</b> ± 8	L528-F
5/16/59	Maple leaves	Palisades, N.Y. 41°00'N, 73°55'W	5 ft above ground on small tree in dense forest	178 ± 6	-25.9	189 ± 6	L528-A
6/7/59	Maple leaves	Palisades, N.Y. 41°00'N, 73°55'W	5 ft above ground on small tree in dense forest	198 ± 7	-26.2	211 ± 7	L528-E
5/16/59 to 6/7/59	Soil CO <sub>2</sub>	Palisades, N.Y. 41°00'N, 73°55'W	Leaf-matted ground in dense forest	106 ± 7	-23.2	108 ± 7	L528-D

spheric  $\Delta C^{14}$  value for northern Europe rose to about 300 by 1 July 1959. On the other hand, Hayes et al. (5) have analyzed lemon grass grown in low latitudes of the Northern Hemisphere and find that, as of June 1959, there has been a 26.8 percent rise over prebomb levels. This is equivalent to about 250 per mill on the  $\Delta C^{14}$  scale and agrees well with data given in this article. From the above differences it appears that true geographical variations do exist within the troposphere of the Northern Hemisphere, presumably the result of finite mixing rates for new C<sup>14</sup> added from the stratosphere. Munnich and Vogel (13) have also demonstrated rather marked seasonal oscillations within the Northern Hemisphere, which are apparently related to the spring maximum of stratospheric Sr<sup>90</sup> fallout (14). On the basis of an average Southern Hemisphere  $\Delta C^{14}$  value of +160 in June 1959 and a Northern Hemisphere average of +250, the tropospheric inventory was  $6.3 \times 10^{27}$ atoms on 1 July 1959 as compared to  $3.6 \times 10^{27}$  on 1 July 1958. Because of inhomogeneities in the troposphere of the Northern Hemisphere, the 1959 estimate has an uncertainty of perhaps 15 percent.

Table 2. Data relating to the increase in the tropospheric  $C^{14}/C^{12}$  ratio between the fall of 1958 and the fall of 1959.

Collection date	Location	Material	$\delta C^{14}$	$\delta C^{13}$	$\Delta C^{14}$	Sample No.
Frank de la factoria		Northern Hemispher	е			
8/8/58 to 8/11/58	Mediterranean Sea 36°N, 7°E	Atmospheric CO <sub>2</sub>	214 = 8	- 5.1	176 ± 8	L493-O
8/15/58 to 8/19/58	Atlantic Ocean 39°N, 30°W	Atmospheric CO <sub>2</sub>	$200 \pm 8$	- 5.8	$162 \pm 8$	L493-P
$\frac{10/22}{58}$ to $\frac{10}{24}$	Atlantic Ocean 33°N, 76°W	Atmospheric CO <sub>2</sub>	197 ± 5	- 5.1	159 ± 5	L529-A
11/8/58 to 11/10/58	Atlantic Ocean 11°N, 76°W	Atmospheric CO <sub>2</sub>	193 ± 8	-6.6	$159 \pm 8$	L529-M
4/4/59	Tucson, Ariz. 32°N, 111°W	Tree leaves	193 ± 9	- 20.7	193 ± 8	L523
5/16/59 to 6/7/59	Palisades, N.Y. 41°N, 74°W	Atmospheric CO <sub>2</sub>	264 ± 7	-16.9	257 ± 7	L528-C
6/5/59	Carson City, Nev. 39°N, 120°W	Tree leaves	$249~\pm~8$	-23.8	259 ± 8	L516-E
6/9/59	Rockleigh, N.J. 41°N, 74°W	Tree leaves	$232~\pm~8$	-25.8	246 ± 8	L528-F
5/16/59 to 6 7/59	Alpine, N.J. 41°N, 74°W	Atmospheric CO <sub>2</sub>	251 ± 6	-18.9	249 = 6	L528-B
10/12/59 to 10/18/59	Alpine, N.J. 41°N, 74°W	Atmospheric CO <sub>2</sub>	236 ± 6	24.0	246 ± 6	L547-A
		Equatorial Region				
12/1/58 to 12/4/58	Pacific Ocean 1°N, 84°W	Atmospheric CO <sub>2</sub>	176 ± 9	-6.1	140 ± 9	L537-A
		Southern Hemispher	e			
4/5/58 to 4/18/58	Atlantic Ocean 35°S, 24°E	Atmospheric CO <sub>2</sub>	118 ± 5	- 5.7	$82 \pm 5$	L493-L
4/28/58 to 5/2/58	Atlantic Ocean 27°S, 46°E	Atmospheric CO <sub>2</sub>	$125 \pm 6$	- 5.4	<b>87</b> ± 6	L493-M
5/13/58 to 5/17/58	Atlantic Ocean 13°S, 65°E	Atmospheric CO <sub>2</sub>	120 ± 5	-3.6	78 ± 5	L493-N
1/28/59	Pacific Ocean 49°S, 75°W	Atmospheric CO <sub>2</sub>	144 ± 7	-7.2	111 = 7	L537-L
3/16/59	Lima, Peru	Grass	$135 \pm 7$	- 10.1	135 = 7	L519-B

Table 3. Data relating to bomb-produced C14 concentration in surface ocean water (24).

Sample	Collection	Collection Location		δC <sup>13*</sup>	$\Delta \mathbf{C}^{14}$ †	Difference from pre-bomb av.	
No. 0	uate		-			Measured	Predicted
		North Atlan	tic (pre-bom	b av., −4	19 <sup>0</sup> / <sub>00</sub> )(15)		
L465 <b>-Z</b>	11/13/57	34°N,65°W	$-1 \pm 5$	+1.5	$-54 \pm 8$	5	10
L464-B	11/16/57	30°N,62°W	$26 \pm 5$	+4.3	$-33 \pm 8$	16	10
L464-D	11/17/57	29°N,61°W	$13 \pm 6$	-1.8	$-33 \pm 6$	16	10
L464-O	11/26/57	15°N,40°W	$1 \pm 8$	- 1.0	$-47 \pm 8$	2	10
L529-I	11/15/58	16°N,75°W	$20 \pm 7$	+3.8	$-38 \pm 7$	11	20
L538-I	5/14/59	10°N,45°W	$19 \pm 6$	+4.1	$-39 \pm 7$	10	27
L538-P	7/10/59	35°N,74°W	$28 \pm 8$	+0.7	$-23 \pm 8$	26	29
		South Atlan	tic (pre-bom	nb av., -58	8 <sup>0</sup> / <sub>00</sub> )(15)		
L464-R	12/5/57	1°N,33°W	$-28 \pm 6$	-1.1	$-76 \pm 6$	-18	10
L464-T	12/16/57	10°Ś,34°W	$-8 \pm 7$	(-1.0)	$(-56 \pm 9)$	2	10
L465-D	12/26/57	23°S,38°W	7 ± 7	-2.1	$-39 \pm 7$	19	10
L465-X	4/6/58	34°S,18°E	$-2 \pm 5$	+1.0	$-54 \pm 5$	4	12
		South Pac	ific (pre-bon	ıb av., -:	$54^{0}/_{00}(7)$		
R359	12/17/57	41°S,174°W	$15 \pm 5$	(-2.0)		23	11
R388/1	3/4/58	36°S,174°E	$1 \pm 9$	(-2.0)	$(-45 \pm 9)$	9	12
R376	3/5/58	41°S,174°E	$0 \pm 5$	(-2.0)	$(-46 \pm 5)$	8	12
R388/3	3/6/58	28°S,176°E	$8 \pm 6$	(-2.0)	$(-38 \pm 8)$	16	12
R388/5	3/8/58	21°S,177°E	9 ± 6	(-2.0)	$(-37 \pm 9)$	17	12
R388/10	3/18/58	20°S,175°E	$13 \pm 5$	(-2.0)	$(-33 \pm 5)$	21	12
R388/11	3/19/58	21°S,173°E	$13 \pm 5$	(-2.0)	$(-33 \pm 5)$	21	12
R388/12	3/21/58	18°S,172°E	$12 \pm 5$	(-2.0)	$(-34 \pm 5)$	20	12
R388/15	3/23/58	15°S,178°E	$12 \pm 5$	(-2.0)	$(-34 \pm 5)$	20	12
R388/21	3/31/58	32°S,175°E	5 ± 5	(-2.0)	$(-41 \pm 5)$	13	12

\* Parentheses in column 5 denote estimates rather than measured values.  $\dagger$  Parentheses in column 6 indicate that the  $\Delta C^{14}$  value is based on an estimated  $\delta C^{13}$  value.

Table 4. Estimates of the distribution of bomb-produced  $C^{14}$  on 1 July for a number of different years (A.D.) (units,  $10^{27}$  atoms).

Reservoir				Year					Steady-state distribution
	1958	1959	1965	1976	2000	2100	2350	7520	of natural radiocarbon
Strato- sphere	8.4*	8.0-17.0	3.1-6.4	1.1-2.4	0.6-1.0	0.2–0.3	0.1–0.2	0.0	7
Tropo- sphere	3.6	6.3	6.7–10.1	4.5–7.2	2.6-4.0	0.8–1.2	0.4-0.6	0.2	28
Biosphere	0.2	0.3	0.8-1.0	1.0-1.6	0.7-1.2	0.2-0.3	0.1	0.0	8
Ocean	0.8	1.4	5.4-7.5	9.4-13.8	12.0-18.7	14.5-22.7	14.4-22.6	7.8-12.3	1960
Total	13.0	16.0-25.0	16.0-25.0	16.0-25.0	15.9-24.9	15.7-24.5	15.0-23.5	8.0-12.5	2000
	-								

\* Hagemann et al. (6).

Table 5. Parameters for reservoirs making up model dynamic carbon cycle.

	Eleva- tion		Carbon content	Steady-	Mean resi-		
Reservoir	range (m)	tropo- sphere (m²)	(gram mole- cules)	Meas- ured*	Com- puted†	dence time (yr)	
Stratosphere	>14,000	$510 \times 10^{12}$	$11 \times 10^{15}$		+75	5	
Troposphere	<14,000		$46 \times 10^{15}$	0		4	
Terrestrial biosphere			$13 \times 10^{15}$			10	
Pacific plus Indian Ocean (<50°S) sur- face water	<100	$220 \times 10^{12}$	$50 \times 10^{15}$	-32		8	
North Atlantic (5°N– 55°N) surface water	<100	$40 \times 10^{12}$	9 × 1015	-35		5	
South Atlantic (50°S- 5°N) surface water	<100	$40 \times 10^{12}$	$9 \times 10^{15}$	(-46)	-65	5	
Pacific plus Indian Ocean (<50°S)‡ deep water	>100		1980 × 1015	-200		800	
Atlantic (50°S-55°N) deep water	>100		655 × 1015	-104		650	
Antarctic Ocean (>50°S)	Entire	$45 \times 10^{12}$	420 × 1015	(-113)	- 120	70	
Arctic Ocean (>55°N)‡	Entire	$15 \times 10^{12}$	40 × 10 <sup>15</sup>	(-28)	- 30	30	

\* Numbers in parentheses were not used in steady-state computations. See values derived from computations (col. 6) and compare with measured values. † The computation was based on the assumption that stratospheric mean residence time is 5 years and that half of the natural radiocarbon production is above the tropopause. ‡ Bering Strait isolates the Arctic Ocean from the Pacific.

On the basis of the same assumptions used previously (4), the amount of bomb-produced C<sup>14</sup> stored in living organic materials is calculated to be  $0.3 \times 10^{27}$  atoms. Although the uncertainty in this estimate is large, bombproduced C<sup>14</sup> in the organic reservoir continues to be so small a part of the total inventory as to be unimportant.

The amount of bomb-produced C<sup>14</sup> which has entered the oceans has also been computed, according to the method of Broecker and Walton (4) except that the return flux of bomb-produced C<sup>14</sup> from the ocean to the atmosphere has been taken into account (in accordance with the circulation model discussed below). Based on a CO<sub>2</sub> exchange rate of 22 gram molecules per square meter per year (15), a value of 1.4  $\times$  10<sup>27</sup> C<sup>14</sup> atoms is obtained for the 1 July 1959 oceanic inventory. It should be noted that this value is essentially independent of the oceanic mixing model selected (see 15).

If all of this oceanic C<sup>14</sup> had remained in the upper 100 meters of water, the C<sup>14</sup>/C<sup>12</sup> ratio of the surface layer should have risen 30 per mill between 1 March 1955 and 1 July 1959. According to the more elaborate model discussed below, which takes into account loss to deep-water masses, the average value for surface water should have risen 29 per mill by 1 July 1959. The corresponding increases for 1 July 1958 and 1 July 1957 are, respectively, 14 and 7 per mill.

Ocean-water measurements available to date are summarized in Table 3. Since the differences are small, it can only be said that they are consistent with the values predicted by the model. Future measurements should provide a good check on the circulation model adopted here, since the rise in surface ocean  $\Delta C^{14}$  values will be such that measurement errors will not be so significant.

If the rate of addition of bombproduced C<sup>14</sup> to the troposphere is assumed to be linearly dependent on the stratospheric inventory, then the rising slope of the curve in Fig. 1 indicates a rising stratospheric inventory. Specifically, the rise from July 1958 to July 1959 is more than twice the annual increase for the previous two years; hence, Hagemann's average inventory (6), measured for the period 1955-1958 (namely, 7.2  $\times$  10<sup>27</sup> atoms), should be scaled up to  $17 \times 10^{27}$  atoms in order to obtain the stratospheric inventory on 1 July 1959. Much of this inventory increase is presumably due to Russian tests during the fall of 1958.

A nearly identical estimate of the stratospheric inventory is obtained by taking Hagemann's estimate (6) of the total amount of C<sup>14</sup> thus far produced by bombs ( $25 \times 10^{27}$  atoms) and subtracting the amount of this C<sup>14</sup> which had passed into the other reservoirs (troposphere, biosphere, and oceans) up to 1 July 1959. The latter amount is given below as  $8 \times 10^{27}$  atoms, and therefore the stratospheric inventory, by difference, is  $17 \times 10^{27}$  atoms.

Despite the agreement between these two estimates of stratospheric inventory, there is reason to believe that they may represent only an upper limit. As suggested by Martell (16), nuclear debris added to the polar stratosphere may exhibit a significantly shorter residence time than that added to the stratosphere of middle latitudes. Before July 1958 most bomb C<sup>14</sup> was probably produced by testing in the equatorial Pacific, whereas subsequently the major part of the C<sup>14</sup> was added by Russian testing in the Arctic. Thus, the increased slope may represent rapid transfer of C14 out of the polar stratosphere rather than a twofold increase in stratospheric inventory. Furthermore, Hagemann et al. (6) admit that their estimate of total bomb-derived radiocarbon produced up to the testing moratorium is only a rough figure. They obtained it by scaling up the January 1957 measured inventory according to the total number of megatons exploded, allowing as well for a difference in the proportion of tests conducted in the air and at surface level.

Clearly, then, there is considerable uncertainty in the stratospheric inventory of July 1959. As a result, only a range is given in Table 4. The upper limit,  $17 \times 10^{27}$ , is the value discussed above, while the lower limit is taken as the measured inventory in 1957—approximately  $8 \times 10^{27}$  atoms (6).

### **Future Distribution**

About 90 percent of the carbon within the ocean-atmosphere-biosphere system is in the ocean. Consequently, prediction of the future distribution of bomb-produced C<sup>14</sup> among the various carbon reservoirs depends mainly on having an adequate model of ocean circulation and ocean-atmosphere interaction. The model used here (shown schematically in Fig. 2) is based on 16 SEPTEMBER 1960

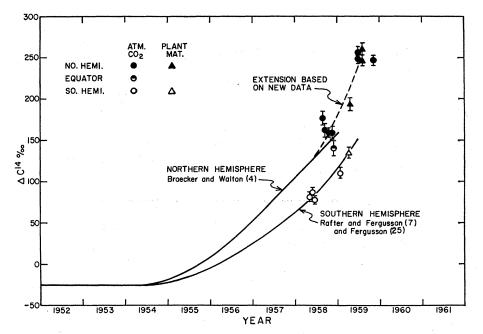


Fig. 1. The C<sup>14</sup>/C<sup>12</sup> ratios [expressed as  $\Delta$ C<sup>14</sup> values (see 9)] for the Northern and Southern Hemisphere tropospheres, plotted as a function of time. The solid curves are those published by Broecker and Walton (4) and by Fergusson (7, 25). The data on which the extension points and dashed curve are based are given in Table 2.

estimates of the steady-state  $C^{14}/C^{12}$ ratios in the ocean and atmosphere; full details are given in (15). The oceanatmosphere-biosphere system has been divided into ten reservoirs, which are defined in detail in Table 5; each is assumed to be so well mixed internally that its C<sup>14</sup> concentration is uniform. Transfer of CO<sub>2</sub> between the reservoirs is as indicated by the arrows (Fig. 2), which are numbered to denote the rates of transfer required for steady-state distribution of  $C^{14}$  throughout the system (15). The mean residence time (in years) of a carbon atom within a given reservoir is indicated by the numbers in parentheses under the reservoir names. Because there is some uncertainty about the number and mean residence time of carbon atoms in the humus of terrestrial soils, this reservoir has not been included in the model. As shown be-

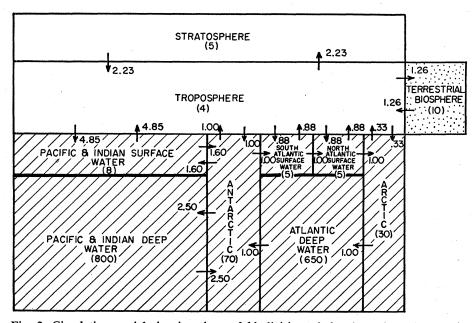


Fig. 2. Circulation model showing the tenfold division of the dynamic carbon cycle. Arrows indicate modes of mixing, and the numbers associated with each arrow give the rate of transfer of CO<sub>2</sub> (in gram molecules/yr  $\times$  10<sup>-15</sup>). The numbers in parentheses are the mean residence times of CO<sub>2</sub> molecules in the reservoirs (see 15 for details).

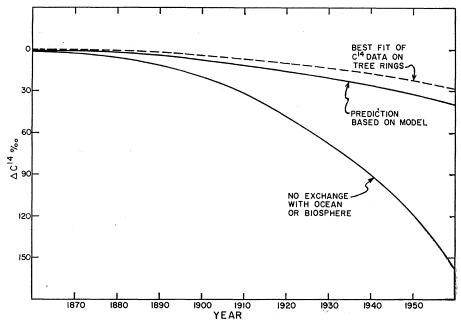


Fig. 3. The C<sup>14</sup>/C<sup>12</sup> ratios (expressed as  $\Delta C^{14}$  values) in atmospheric CO<sub>2</sub> plotted against time; the plot is based on theory (solid curves) and on measurements (dotted curve). The decrease in  $\Delta C^{14}$  is due to the addition to the atmosphere of C<sup>14</sup>-free CO<sub>2</sub> produced by the combustion of coal and oil. The curve based on measurements is primarily from the work of Fergusson (26).

low, there is evidence that the mean residence time of carbon atoms in humus is measured in hundreds rather than tens of years; thus it is of little present importance in connection with the problem of bomb-produced  $C^{14}$ .

The stratospheric residence time given in Fig. 2 is 5 years, as determined by Hagemann et al. (6) on the basis

of C<sup>14</sup> data. From consideration of Sr<sup>90</sup> inventories, Feely (17) has suggested that the residence time may be closer to 1.5 years. It is possible, however, that Sr<sup>90</sup> is transferred to the troposphere faster than C<sup>14</sup> because Sr<sup>90</sup> becomes attached to particles sufficiently large to settle, whereas C<sup>14</sup> remains as a gas. In addition, the

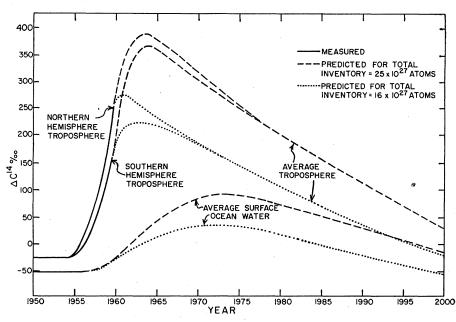


Fig. 4. Predicted  $C^{14}/C^{12}$  ratios (expressed as  $\Delta C^{14}$  values) for the troposphere and surface ocean water from A.D. 1960 to 2000. Two sets of curves are given in order to reflect the limits of uncertainty in the stratospheric inventory of bomb-produced  $C^{14}$ . Estimates include the projected combustion-produced  $CO_2$  effect. Direct measurements are available up to 1 July 1959 for the troposphere and through 1957 for surface ocean water. The dates (abscissa) refer to 1 January of the year in question.

stratospheric residence time may depend on altitude and latitude of injection—a longer residence time for low latitudes and high elevations.

In order to check whether the ocean model adequately represents mixing within the dynamic reservoir, predictions based on it have been made, for the period 1850 to 1955, of the  $C^{14}/C^{12}$ ratio of atmospheric CO<sub>2</sub> resulting from man's release of C14-free CO2 by burning coal and oil. The amounts of such CO<sub>2</sub> added to the tropospheric reservoir each decade since 1860 have been summarized by Revelle and Suess (18). Calculations in which their data are applied to the model suggested here give the upper solid curve in Fig. 3. For comparison, a dotted curve based on the available analyses of tree-ring samples is plotted on the same graph. When corrected for radioactive decay between growth and measurement and normalized to a common C13/C12 ratio (normalization corrects for isotope fractionation), tree-ring data should provide a basis for estimating the  $C^{14}/C^{12}$  ratio in the atmosphere at the time of ring growth. Also plotted in Fig. 3 is the  $C^{14}/C^{12}$  ratio the atmosphere would have had if there had been no communication between atmospheric CO<sub>2</sub> and bicarbonate dissolved in the ocean or between atmospheric CO<sub>2</sub> and organic matter in the terrestrial biosphere. The theoretical model is seen to provide a fairly good fit to the data, predicting a slightly greater effect of combustion-produced CO2 than is actually observed. Thus, it is reasonable to conclude that the model provides a fair approximation to the true transfer rates between the atmosphere and other reservoirs.

With the model proposed here, it is possible to predict future distributions of bomb-produced C<sup>14</sup> throughout the dynamic carbon reservoir. Table 4 gives such results; one set is based on the upper limit of the stratospheric inventory and one on the lower. These predictions are made on the assumption that there will be no further increase in inventories of C<sup>14</sup> from bomb testing. The total inventory decreases with time as the result of radioactive decay (1 percent per 80 years). On the other hand, the inventory of cosmic-rayproduced C<sup>14</sup> (given for comparison in column 10) should remain essentially constant.

Inventory values such as those given in Table 4 can readily be converted to  $\Delta C^{44}$  values in order to show per-millage changes from pre-bomb levels. This conversion has been made for the tropospheres of the two hemispheres as well as for surface ocean water (the upper 100 meters); the results are plotted in Fig. 4 to A.D. 2000. The solid portions of the curves are fixed by measurements, while the dashed portions are based on the model. The dashed curves also take into account the effect of further combustion of fossil fuels; consumption estimates given by Revelle and Suess (18) were used. The predicted reductions in the atmospheric  $C^{14}/C^{12}$  ratio resulting from future increments of combustion-produced CO<sub>2</sub> are 65 per mill in 1975 and 140 per mill in 2000. The comparable estimates for the surface ocean are, respectively, 40 and 95 per mill.

The results given in Fig. 4 suggest that the atmosphere will reach its peak  $C^{14}/C^{12}$  level before 1963 and that the surface ocean will reach its peak between 1970 and 1975. The ratio for the atmosphere will have dropped to one-half the peak value between 1975 and 1980, and the ratio for the surface ocean will have dropped proportionately by about 1990. In about 50 years the  $C^{14}/C^{12}$  level in the atmosphere will have returned to the pre-bomb level. From this time on, the fossil-fuel effect will dominate. All of these conclusions are based on an assumption that there will be no further additions of C<sup>14</sup> from bomb detonations and on an assumed stratospheric mean residence time of 5 years.

Just how valid the model is will be revealed by continued monitoring of atmospheric and oceanic  $C^{14}/C^{12}$  ratios. As discussed below, future monitoring of bomb-produced  $C^{14}$  throughout the ocean-atmosphere-biosphere system will also provide considerable information regarding residence times and modes of mixing. If the present model is not adequate, the information that shows its inadequacy will aid in developing a better model. Measurements over the next year or two should resolve the uncertainty in the total inventory.

## Potential Tracer Applications of Bomb-Produced Carbon-14

The radiocarbon produced by nuclear tests provides a potentially useful tracer in the study of a number of problems in the natural sciences. In order to suggest the potentialities of this tracer to scientists in various fields, we cite four examples of its use below. These examples involve the fields of soil science.

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biochemistry, limnology, and oceanography.

Soil science. An important variable in the field of soil science is the mean life of organic materials in active soils. Theoretically, the  $C^{14}/C^{12}$  ratio in soil carbon is an indicator of mean life. Prior to the addition of bomb-produced C<sup>14</sup>, however, the C<sup>14</sup> method was a relatively insensitive tool for this purpose, as is indicated by the curves marked "1900" and "1950" in Fig. 5. In pre-bomb days, soil organic fractions with mean lives between 4 and 256 years had  $C^{14}/C^{12}$  ratios almost within the range of error of precise radiocarbon measurements ( $\pm 5$  per mill). To some extent this lack of sensitivity stems from the post-1900 decrease in the atmospheric  $C^{14}/C^{12}$ ratio, which depended on combustionproduced CO<sub>2</sub>.

Figure 5 also shows how organic materials of soils will react to the addition of bomb-produced C<sup>14</sup>. By 1964 it should be possible to measure residence times with an accuracy of better than 10 percent. Since active soils contain a mixture of organic materials which undoubtedly have different residence times, interpretation of soil data will no doubt be a complex problem. This will be especially true if there exists a chemical decay sequence of organic fractions such that one fraction is derived from the next. For example, if A decays to B, B to C, C to D, and so on, then D might have a very short residence time in the soil and still not show any bomb-produced C<sup>14</sup> for many years. Fractions A, B, and C would act as buffers in the passage of bombproduced  $C^{14}$  to fraction D. Nevertheless, measurements carried out over a number of years on various mechanical and chemical fractions of a given soil should provide valuable information. Provided no more above-ground bomb testing is carried out, the period of greatest sensitivity for organic fractions of short residence time will be between now and 1965. Sometime within this period the C14/C12 ratio of the atmosphere will begin decreasing, and thus additional complications in the interpretation of soil data will enter.

Since a knowledge of the mean residence time of carbon in organic fractions in soil is necessary to a complete understanding of the geochemical cycle of carbon, a program of radiocarbon measurements on such fractions has been initiated at the Lamont Observatory. To date, five measurements have been made, all on fractions of a soil sample taken in early June 1959 in a wooded area on the Lamont Observatory grounds in Palisades, New York. The sample descriptions and measurement data are given in Table 6.

Carbon-dioxide given off by the soil had a  $\Delta C^{14}$  value slightly lower than that of plants formed during the summer of 1958-a finding which suggests, as might be expected, that this CO<sub>2</sub> is largely derived from the decay of the previous year's growth. The result on the humic acids suggests a mean life of between 50 and 250 years for this fraction (see the curve for 1958 in Fig. 5). On the other hand, the organic fractions remaining after humic acid extraction are old enough to be measured by natural radiocarbon, having an age of 2000 years (equivalent to a  $\Delta C^{14}$  value of -229). Aside from the escaping CO<sub>2</sub>, the coarse organic materials from the 21-inch composite are the only fraction which might have taken on bomb-produced C<sup>14</sup>; this fraction probably has a mean residence time of 50 years or less. However, the fine organic materials from the same composite show an appreciable age of about 1100 years ( $\Delta C^{14} = -132$ ). The

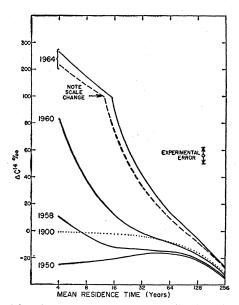


Fig. 5. A plot showing the  $C^{14}/C^{12}$  ratio (expressed as  $\Delta C^{14}$  along the ordinate) which one would find in Northern Hemisphere soil organic material if the organic material had a mean residence time of x(the abscissa) and were collected and measured in the year M. Two sets of curves are given for 1964 in order to reflect the limits of uncertainty in the present stratospheric inventory of bombproduced C<sup>14</sup>. Curves are given for M =The 1900, 1950, 1958, 1960, and 1964. curves are based on the model of the carbon cycle described in this article. The dates refer to 1 January of the year in question.

Table 6. Radiocarbon analyses on various fractions of a soil sample from a wooded area in Palisades, N.Y., collected in the late spring of 1959.

Material	Separation procedure	Carbon (gm per kg of soil*)	δC14	δC <sup>13</sup>	ΔC <sup>14</sup>	Sample No.
CO <sub>2</sub> given off by soil	Absorption in KOH lo- cated under barrel in- verted over soil	<b>†</b>	107 ± 7	-23.2	108 ± 7	L528-D
Humic acids from surface soil (upper 6 in.)	NaOH soluble portion of fine fraction ( <40 mesh)	5 <b>±</b> 2	$-25 \pm 7$	-24.2	$-28 \pm 7$	L528-G
Residual organic fraction in surface soil after removal of humic acids	NaOH insoluble portion mesh of fine fraction (<40)	8 <b>±</b> 2	$-218 \pm 9$	-24.7	-229 ± 9	L528-P
Coarse 100-percent organic fraction of bulk soil (composite from 0 to 21 in. depth)	Stayed on screen (ordi- nary window screen) when soil was washed through	4 ± 2	-1 ± 7	24.4	$-2 \pm 7$	L528-K
Fine fraction of bulk soil (composite from 0 to 21 in. depth)	Went through window screen (includes fine or- ganic)	10 <b>±</b> 2	-123 = 9	-23.0	$-132 \pm 9$	L528-L

\* Weight of soil is based on a depth of 21 in. Since most organic matter is in the upper third of this depth, values in column 3 would be tripled if only 7 in. depth were taken.  $\uparrow$  At the observed rate of escape of CO<sub>2</sub> from the area under the barrel (that is, 7 gram molecules/m<sup>2</sup> month), about 1.2 percent of the total soil organic carbon was being converted to CO<sub>2</sub> in 1 month.

Table 7. Data relating to the time lag in  $C^{14}/C^{12}$  ratio between human blood and the atmosphere.

Sample No.	C	ollection	- Material	δC <sup>14</sup>	δC <sup>13</sup>	$\Delta C^{14}$	Date of equivalent in	Time
	Date	Location	- Materiai	(0/00)	(0/00)	(0/00)	atmosphere	lag (yr)
			Northern	Hemisphere				
L505-A (20)	1/1/59	Palisades, N.Y.	Breath CO <sub>2</sub>	106 ± 8	-21.8	105 ± 8	1/58	1.0
L505-B (20)	1/1/59	Palisades, N.Y.	Blood	$115 \pm 8$	-16.5	102 = 8	1/58	1.0
L516-A	3/4/59	Carson City, Nev.	Blood	118 ± 7	-16.4	104 = 7	1/58	1.2
L516-D	6/1/59	Carson City, Nev.	Blood	$135 \pm 6$	18.0	126 ± 6	4/58	1.1
L516-G	12/17/59	Carson City, Nev.	Blood	$183 \pm 6$	-17.8	175 ± 6	12/58	1.0
L569-A	1/19/60	Palisades, N.Y.	Blood	$194 \pm 6$	-16.7	$184 \pm 6$	1/59	1.0
L569-B	1/13/60	Palisades, N.Y.	Breath CO <sub>2</sub>	$213 \pm 6$	-18.2	$207 \pm 6$	2/59	1.1
			Southern	Hemisphere				
L519-A	3/6/59	Lima, Peru	Blood	$70 \pm 6$	-17.2	$56 \pm 6$	11/57	1.4
L516-C	3/15/59	Ushuaia, Argentina	Blood	<b>33 ± 8</b>	-19.2	$20 \pm 8$	11/56	2.4

Table 8. Effect of bomb-produced C<sup>14</sup> in rivers and lakes.

Sample No.	Collection date	Material	$\delta C^{14}$	δC13	$\Delta C^{14}$	Annual increase ( <sup>0</sup> / <sub>00</sub> )	Fraction of atmospheric increase
			ruckee River, C	Calif.			
L415-NN (21)	Aug. 1957	Plants living under water	$-40 \pm 7$	-18.8	-53 ± 7		
L415-00 (21)	Aug. 1957	Mollusk shell	6 ± 7	-4.7	$-34 \pm 7$		
L415-PP (21)	Aug. 1957	Mollusk meat	$-23 \pm 7$	-27.0	-20 <u>`</u> = 7	< 20	< 0.40
L487-H	Aug. 1958	Dissolved HCO3	$-1 \pm 7$	8.5	$-34 \pm 7$		
L487-F	Aug. 1958	Mollusk shell	$-8 \pm 6$	-3.1	$-52 \pm 6$		
L487-G	Aug. 1958	Mollusk meat	$-22 \pm 6$	-20.3	$-32 \pm 6$		
			Weber River, U	Itah			
L415-S	Aug. 1957	Plants living under water	$-146 \pm 6$	-32.2	$-141 \pm 6$	17 ± 11	$0.34 \pm 0.22$
L487-O	Sept. 1958	Plants living under water	$-116 \pm 9$	-23.7	$-124 \pm 9$		
		G	reat Salt Lake,	Utah			
L415-FFF (21)	June 1958	Brine shrimp	$-111 \pm 7$	-16.3	$-132 \pm 7$		
L415-GGG	June 1958	Dissolved HCO3	$-93 \pm 6$	+3.8	$-150 \pm 6$	67 ± 12	$0.64 \pm 0.12$
L542	Aug. 1959	Brine shrimp	$-39 \pm 9$	-15.0	-60 = 9		

fact that separation into fractions of different age is possible by straight sieving is consistent with the prediction that comminution of organic litter is timedependent. Just how far this method of separation by age can be carried is not known from the data now available. An interesting and consistent fact to point out is that a recombination of the humic acids with the residual organic fractions gives an organic mixture with almost the same  $\Delta C^{14}$  value as that measured for the fine fraction from the soil composite.

Along the same line, Tamm and Ostlund (19) in Sweden have recently published radiocarbon measurements on total organic matter found in three horizons of a soil profile. They find a progressive decrease in radiocarbon with depth, total organic matter in the deepest layer having a mean residence time of about 400 years. Further analyses of organic materials from soils collected over the next few years will make it possible to draw firmer conclusions. The data reported here merely illustrate the type of information which can be obtained.

Biochemistry. Broecker et al. (20) pointed out the value of bomb-produced  $C^{14}$  as a tracer in determining the mean life of carbon in various tissues of animals and men. Just as Fig. 5 gives quantitative data for soils, so Fig. 6 gives information on residence time for human tissue. Fig. 6 gives data only for relatively short residence times and differs from Fig. 5 in that blood, rather than atmospheric CO<sub>2</sub>, is taken to be the source of the carbon in human tissue. Where data from direct analyses of the blood are not available, it is assumed that the  $\Delta C^{14}$  value for blood is the same as that for atmospheric CO<sub>2</sub> one year earlier. This assumed one-year lag is based on the data given in Table 7 for three residents of the United States (five samples). To be sure, the actual lag depends on such factors as food storage time and the relative proportions of terrestrial and aquatic foods in the diet. No doubt dietary differences account for the high lag times for the Southern Hemisphere listed in Table 7. In any given case, however, an analysis of the blood will indicate the actual lag time. Without such an analysis, it is probably safe to assume a lag time of one year if the person in question ate what might be called a typical American diet.

To date, only one sample of human tissue has been analyzed. Perry Hudson provided us with a composite sample of blood-free muscle tissue taken during an autopsy of a New York City resident who died in January 1960. The  $\delta C^{13}$  and  $\Delta C^{14}$  results are, respectively, -16.2 and  $+ 143 \pm 10$  per mill. On the basis of the one-year lag time suggested above, the mean residence time of carbon atoms in muscle tissue is  $0.8 \pm$ 0.2 year. Again, it is well to emphasize that studies such as this should be carried out while the atmospheric curve is still rising, a situation which even now may be coming to an end.

Limnology. An understanding of the CO2 cycle in fresh-water systems is important in a number of scientific problems, such as metabolism studies in lakes, studies of probable sources of ground and thermal waters, and radiocarbon dating of materials formed in fresh water. One parameter that is necessary in many of these studies is the invasion rate of CO<sub>2</sub> from the atmosphere into the body of water. Broecker and Walton (21) estimated such invasion rates for several closed lakes in the Great Basin, using the steady-state C<sup>14</sup> concentrations in these lakes and in the rivers which supply them. Another way to estimate these invasion rates is to observe the rate of build-up of bomb-produced C<sup>14</sup> in the lakes. Because of its low bicarbonate concentration and its large ratio of surface area to volume, Great Salt Lake should show a far more rapid increase in  $C^{14}/C^{12}$  ratio than the other lakes studied. Therefore, it is the best lake to sample periodically in order to provide data for accurately estimating the invasion rate of CO2. Measurements spaced 14 months apart [June 1958 to August 1959 (see Table 8)] show that  $\Delta C^{\mbox{\tiny 14}}$  values for Great Salt Lake rose  $67 \pm 12$  per mill during this period. This rise can be used to predict the invasion rate in the following manner.

Let us assume that the total  $CO_2$  concentration in the lake is at steadystate and that the annual water input is equal to the water lost by evaporation. The following equation will then express the rate of change in the C<sup>14</sup> concentration of the dissolved bicarbonate in the lake:

$$\frac{d\Delta \mathbf{C}^{\mathbf{I4}}_{\mathrm{L}}}{dt} = \frac{l_{\mathrm{E}}k_{\mathrm{R}}}{\overline{h}\,k_{\mathrm{L}}} \left(\Delta \mathbf{C}^{\mathbf{I4}}_{\mathrm{R}} - \Delta \mathbf{C}^{\mathbf{I4}}_{\mathrm{L}}\right) + \frac{R}{k_{\mathrm{T}}\overline{h}} \left(\Delta \mathbf{C}^{\mathbf{I4}}_{\mathrm{A}} - \Delta \mathbf{C}^{\mathbf{I4}}_{\mathrm{L}}\right) \quad (1)$$

where  $\Delta C^{14}{}_{L}$ ,  $\Delta C^{14}{}_{R}$ , and  $\Delta C^{14}{}_{\Lambda}$  are, respectively, measures of the C<sup>14</sup> concentration in the dissolved bicarbonate of the lake, in the dissolved bicarbonate 16 SEPTEMBER 1960

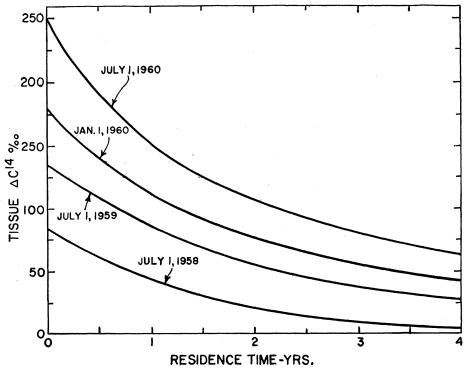


Fig. 6. A plot showing the  $C^{14}/C^{12}$  ratio (expressed as  $\Delta C^{14}$  along the ordinate) in body tissue as a function of mean residence time (abscissa). A 1-year time lag between blood and atmospheric CO<sub>2</sub> is assumed. The date on each curve represents the date of collection of a tissue sample.

of average river water entering the lake, and in CO<sub>2</sub> from the atmosphere over the lake;  $l_{\mathbb{H}}$  is the mean annual linear evaporation rate;  $\overline{h}$  is the mean depth of the lake; and  $k_{\mathbb{L}}$  and  $k_{\mathbb{H}}$  are, respectively, the total CO<sub>2</sub> concentration in the lake and in average river water entering the lake. R is the rate of invasion of CO<sub>2</sub> from the atmosphere into the lake.

Mean  $\Delta C^{14}$  values based on actual measurements are +190 for  $\Delta C^{14}$  and -100 for  $\Delta C^{14}$ <sub>L</sub>. Since the rivers draining into Salt Lake are as yet only slightly influenced by bomb-produced  $C^{14}$  (as discussed below), the mean value assumed for  $\Delta C^{14}{}_{R}$  is the one measured by Broecker and Walton for the Bear River at Preston, Idaho (21)that is, -255. The values of  $l_{\rm E}$ ,  $\overline{h}$ ,  $k_{\rm L}$ , and  $k_{\rm R}$  are, respectively, 1.1 meter per year, 3.6 meters, 6.2 gram molecules per cubic meter, and 2.8 gram molecules per cubic meter (21). The only variables in Eq. 1 which are as yet unspecified are  $d\Delta C^{14}L/dt$  and R. The former can be calculated from the 67 per mill per year rise given above. Solution for the remaining variable Rgives an invasion rate of 6 gram molecules per square meter per year. When all the uncertainties are considered, it is probable that the actual value is not less than 3 nor greater than 8 gram molecules. This agrees well with the

best estimate of 4 gram molecules per square meter per year obtained with the steady-state assumptions (21).

Bomb-produced C<sup>14</sup> is also a means of determining the dominant source of bicarbonate found in fresh waters. In most cases there are three sources: (i) sedimentary carbonates which are free of radiocarbon, (ii) atmospheric CO<sub>2</sub>, and (iii) CO<sub>2</sub> produced in the decay of organic materials either in the drainage-basin soils or in the sediments underlying the body of water. As shown by Munnich and Vogel (22) and by Broecker and Walton (21), measurements of natural C14 can indicate the relative contributions of source (i) and of sources (ii) and (iii) combined. However, the contributions of sources (ii) and (iii) were not separable by this means, since concentrations of C<sup>14</sup> in soil CO<sub>2</sub> and in atmospheric CO<sub>2</sub> were very nearly the same prior to bomb testing. With the addition of bomb-produced C<sup>14</sup> to the atmosphere, soil CO2 now lags considerably behind atmospheric  $CO_2$  in  $C^{14}/C^{12}$  ratio; hence, under certain circumstances a differentiation between sources (ii) and (iii) may be made on this basis.

A third area of application is that of determining the transit time of ground water. Suppose that the relative contributions of the three sources of dissolved bicarbonate are known, Table 9. Radiocarbon data bearing on the replacement rate of Cariaco Trench water.

Water mass	Date	No. of sam- ples	Av. ΔC <sup>14</sup> ( <sup>0</sup> / <sub>00</sub> )	S.D.* (º/₀)
Caribbean surface water (measured)	1955	8	- 57	±9
Caribbean surface water (estimated from model)	1890		-43	
Cariaco trench deep water (measured)	1957	2	-40	±7
*Standard deviation				

\*Standard deviation.

through an analysis of the type described above. Suppose, too, that the  $C^{14}/C^{12}$  ratios in each can be estimated. Then a measurement of the  $C^{14}/C^{12}$  ratio in a sample of ground water will indicate how long it has been since the water picked up its  $CO_2$ . It should be emphasized, however, that estimates of both travel time and source cannot be made from the same  $C^{14}$  measurement. Hence, periodic  $C^{14}$  measurements should be made and combined with other information, such as stable carbon isotope ratios and tritium measurements.

Other examples of the use of  $C^{44}$  data from fresh-water systems are measurements of the Truckee River at Truckee, California, and of the Weber River at Ogden, Utah, which show that the  $C^{14}/C^{12}$  ratio is rising at a considerably lower rate in these rivers than in the atmosphere (see Table 8). This suggests either that the atmosphere is not the major source of C14-bearing CO2 or that the water, after taking on atmospheric CO<sub>2</sub>, resides more than three years in the ground-water system before reaching the river. If the transport time through the ground-water system is actually short, then CO<sub>2</sub> from organic decay is the major source of  $C^{14}$ -bearing bicarbonate and the decaying organic materials must have mean lives of more than three years.

Thus, while the C<sup>14</sup> concentration in the atmosphere continues to rise rapidly, limnic processes which involve carbon and which occur over periods of from 1 to 100 years can be effectively studied by periodic measurements of C<sup>14</sup> concentrations.

Oceanography. Of all the fields mentioned above, perhaps oceanography should benefit most from the study of bomb-produced C<sup>14</sup>. Already natural radiocarbon has provided much valuable information concerning the turnover time for deep-water masses which exhibit residence times of the order of hundreds of years (7, 15). But for studying near-surface masses which communicate with the surface on a

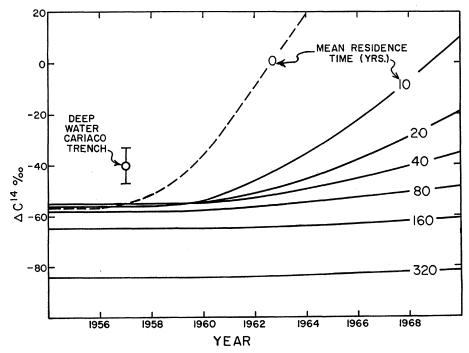


Fig. 7. A plot showing the  $C^{14}/C^{12}$  ratio (expressed as  $\Delta C^{14}$  along the ordinate) as a function of time for various deep-water residence times (T) in the Cariaco Trench. T is defined with respect to the mixing with Caribbean surface water. The curves are based on the model of the carbon cycle described in this article and on an upper limit for the stratospheric inventory of bomb-produced C<sup>14</sup>. The average of two measurements on deep-water samples from the Cariaco Trench is shown.

time scale of tens of years, natural radiocarbon is of little use. It is in the study of such water masses, however, that bomb-produced  $C^{14}$  is most useful.

Consider, for example, the Cariaco Trench in the southern Caribbean, off Venezuela. The circulation times in this trench are of particular interest because it is stagnant in the sense that dissolved oxygen is absent while H<sub>2</sub>S is present. The C<sup>14</sup> measurements for various water samples from the Cariaco Trench are listed in Table 9. They show that the deep waters of the trench have as high a  $C^{14}/C^{12}$  ratio as the surface waters supplying the trench. This suggests that mixing occurs in tens rather than in hundreds of years. With the atmospheric C<sup>14</sup> concentration rising so rapidly, however, the deep and surface waters will begin to show measurable differences unless the mixing occurs in as short a period as several months. The situation is graphically represented in Fig. 7, where the radiocarbon concentrations predicted for the deep waters of the trench are shown for various mean residence times (times of residence prior to mixing with the surface waters of the Caribbean). It is obvious that in 1956 (a representative pre-bomb period) residence times of less than 100 years could not be determined from radiocarbon concentrations. By 1970, however, as shown in Fig. 7 (far right), radiocarbon concentrations will indicate residence times in the range from 0 to 100 years rather effectively. In addition to indicating the mean residence time in the trench, C<sup>14</sup> data should also make it possible to determine the mode of injection and the rate of mixing of new water added to the trench. For the minimum C<sup>14</sup> inventory, curves similar to those of Fig. 7 could be drawn. In such a figure each point would be only two-thirds as far above the pre-bomb base level as the corresponding point in Fig. 7.

### Conclusions

Radiocarbon measurements in simultaneously collected tree-leaf and atmospheric  $CO_2$  samples show that the concentrations are often the same but that situations arise where tree leaves give a lower value. A possible explanation for this lack of agreement is that the leaves receive part of their  $CO_2$ through the roots of the tree. Consequently, it appears that direct collection of CO<sub>2</sub> from the air is the most reliable way to monitor the atmospheric  $C^{14}/C^{12}$  ratio.

Further monitoring of tropospheric  $C^{14}/C^{12}$  ratios in both hemispheres indicates that the rate of rise roughly doubled during the period from 1 July 1958 to 1 July 1959. This increase may be due either to a doubling of the stratospheric inventory from about  $8 \times 10^{27}$  to around  $17 \times 10^{27}$  C<sup>14</sup> atoms or to quite rapid fallout of debris added to the polar stratosphere.

Estimates of the future distribution of bomb-produced C<sup>14</sup> throughout the atmosphere-biosphere-ocean system have been made; for this purpose a mixing model based on steady-state concentrations of natural radiocarbon was used. The predicted inventories are given in Table 4. From these inventories, combined with estimates of the future effects of combustion-produced CO2, the  $C^{14}/C^{12}$  ratios for the atmosphere and for surface ocean water can be predicted (see Fig. 4). The ratio for the atmosphere of the Northern Hemisphere will rise before 1963 to a maximum of from 1.3 to 1.4 times the pre-bomb value, whereas for surface ocean water the maximum will be between 1.09 and 1.15 times the prebomb value and will occur between 1970 and 1975. By 1980 the ratio for the atmosphere will have dropped halfway back to the pre-bomb level, while the ratio for the surface ocean will drop proportionately by about 1990. Early in the next century the combustion-produced CO2 effect will be dominant in the atmosphere, and the  $C^{14}/C^{12}$ ratio will have dropped below the prebomb value. As illustrated by several examples, bomb-produced C14 can serve as a potential tracer in the fields of soil science, biochemistry, limnology, and oceanography.

Due to continuous spiking of the troposphere with bomb-produced radiocarbon, changes in C<sup>14</sup> concentration throughout the carbon cycle are now occurring faster, by several orders of magnitude, than changes due solely to radioactive decay. Consequently, processes carried out in from 1 year to 100 years are now measurable, or will be so in the next few years. It is hoped that the examples discussed in this article will stimulate the imagination of scientists in many fields, so that the many potential applications of this world-wide tracer may be realized (23).

#### **References and Notes**

- 1. G. S. Bien and H. E. Suess, Z. Physik 130, G. S. Bien and H. E. Suess, Z. Physik 130, 309 (1959); H. de Vries, Science 128, 250 (1958); K. O. Munnich and J. C. Vogel, Naturwissenschaften 45, 327 (1958); R. L. Patterson, Jr., and I. H. Blifford, Jr., Science 126, 26 (1957).
  E. H. Willis, Nature 185, 552 (1960).
  H. Tauber, Science 131, 921 (1960).
  W. S. Broecker and A. Walton, *ibid.* 130, 309 (1959).
  F. N. Haves, E. Hansbury, V. N. Kerr, D. J.

- 5 F. N. Hayes, E. Hansbury, V. N. Kerr, D. L. Williams, Z. Physik 158, 374 (1960).
  6 F. T. Hagemann, J. Gray, Jr., L. Machta, A. Turkevich, Science 130, 542 (1959).
  7 T. A. Rafter and G. J. Fergusson, New Zea-

land J. Sci. Technol. **B38**, 871 (1957); paper (A/Conf 15/P/2128) presented at the 2nd United Nations International Conference on the Peaceful Uses of Atomic Energy, New Zealand, 11 June 1958.

- 8. This research was carried out at the Lamont Inis research was carried out at the Lamont Geological Observatory of Columbia Uni-versity. Financial support for the work was provided by the Atomic Energy Commission [grant AT (30-1)1656]. M. Zickl, J. Hubbard, R. McPherson, and F. Senn aided in the laboratory work.
- W. S. Broecker and E. A. Olson, Am. J. Sci. Radiocarbon Suppl. 1, 111 (1959).
   E. I. Rabinowitch, Photosynthesis and Related

- E. I. Rabinowitch, Photosynthesis and Related Processes (Interscience, New York, 1951), vol. 2, pt. 1, p. 910.
   A. L. Kursanov, N. N. Krjukova, B. B. Vartapjetjan, Doklady Akad. Nauk S.S.S.R. 85, 913 (1952).
   J. A. J. Stolwijk and K. V. Thimann, Plant Physiol. 32, 513 (1957).
   K. O. Munnich and J. C. Vogel, paper pre-sented at the Radiocarbon Dating Conference held Sept, 1959 at Groningen, Netherlands. See H. Godwin, Nature 184, 1365 (1959).
   U.S. Atomic Energy Comm. Health and Safety Lab. Rept. No. 69 (1959), p. 11.
   W. S. Broecker, R. Gerard, B. C. Heezen, M. Ewing, J. Geophys. Research 65, in
- M. Ewing, J. Geophys. Research 65, in press.
- 16. E. A. Martell, Science 129, 1197 (1959).
- 17. H. W. Feely, *ibid.* 131, 645 (1960). 18. R. Revelle and H. E. Suess, *Tellus* 9, 18 (1957)
- 19. C. O. Tamm and H. G. Ostlund, Nature 185, 706 (1960).
- W. S. Broecker, A. Schulert, E. A. Olson, Science 130, 331 (1959).
   W. S. Broecker and A. Walton, Geochim. et Cosmochim. Acta 16, 15 (1959).
   K. O. Munnich and J. C. Vogel, Naturwissen-schaften 5, 168 (1959).
   Thie orticle is Longet Conference Operatory.
- 23. This article is Lamont Geological Observatory Contribution No. 423.
- Contribution No. 423.
  24. The Atlantic Ocean surface-water samples were collected by the Columbia University research vessel Vema. We appreciate the cooperation and help given by R. Gerard in this phase of the program. The Pacific results were obtained by Rafter and Fergusson (7).
  25. G. L. Fergusson paper prepared for presented for pres
- G. J. Fergusson, paper prepared for presentation at the International Oceanographic Congress, New York, Sept. 1959.
   <u>6</u>, *Proc. Roy. Soc. (London)* A243, 561 (1958).

# Brenton Reid Lutz, Vascular Physiologist

Brenton Reid Lutz was born in Woodlawn, Nova Scotia, on 2 June 1890. He graduated from Melrose High School, Melrose, Massachusetts, in 1909. He attended the College of Liberal Arts and the Graduate School of Boston University, earning the degrees of S.B., A.M., and Ph.D. (1917) in physiology. During his early professional years he was awarded the 16 SEPTEMBER 1960

Jacob Sleeper fellowship at Boston University (1914) and a teaching fellowship at Harvard University (1915-1917). In 1918 he married Edna Baldwin. From 1919 to 1939 he held joint appointments at Boston University in the College of Liberal Arts (from instructor to professor of biology in 1927) and in the Medical School (from instructor to associate professor of

physiology in 1930). He succeeded Arthur W. Weysse as chairman of the biology department in 1927. He continued to give, annually, a series of lectures in physiology at the Medical School until 1939.

Lutz made blood-pressure measurements in man with Weysse, who introduced the auscultatory method to America in 1913. He worked with Walter B. Cannon in the physiology laboratory at Harvard Medical School. As a result, he developed a life-long interest in the autonomic nervous system and the cardiovascular system, and he became adept in mammalian surgery and physiological instrumentation.

In World War I, Lutz served as a lieutenant in the Sanitary Corps and pioneered in aviation physiology at Mineola, Long Island, under Edward C. Schneider. An important series of