a cutthroat—competition among ideas is essential to research. It seems to me that a researcher should have no freedom to make other able people work on his ideas rather than on their own, beyond what he can make them want to do through persuasion based on the merits of his ideas.

Fortunately, today good people don't have to work under any other conditions. There is tremendous competition for good workers with good ideas. A man who really suffers from the tyranny of the boss can go to some other department or to some other company. And, if a boss is so tyrannous that good men leave him, he won't last forever.

Still, men do complain about the tyranny of jobs and bosses. It is my observation that these are seldom the best men. In fact, often they are men who have amply demonstrated their inability to do research when left completely to themselves, and sometimes they are men who should not try to do research under any circumstances.

Clearly, freedom is vitally important to research, but other somewhat inconsistent things are important, too. Responsibilities of one sort or another keep us from following up every idea or inclination we have. Apparatus ties us down. Concentration on one subject lessens our ability to tackle another. Then too, we may lose our freedom foolishly. Snobbishness may dictate our course of action. Or we may work in a poor environment, inadequately provided for, or with a tyrannous boss to browbeat us. But, in this day and age we are foolish if we put up with such things, unless we really aren't good enough to find another environment—or unless conditions aren't as bad as we think they are after all.

Stratospheric Carbon-14, Carbon Dioxide, and Tritium

The program of high-altitude balloon sampling reveals new information about the stratosphere.

French Hagemann, James Gray, Jr.,
Lester Machta, Anthony Turkevich

This article reports the results of measurements made in recent years of the carbon-14, carbon dioxide, and tritium in the stratosphere. The purpose of the measurements was to obtain information on the stratospheric concentrations of carbon-14 and tritium produced by the explosion of nuclear devices and to study the changes in these concentrations with time. Such data can be expected to furnish new insight into the circulation of the stratosphere, as well as to contribute to the evaluation of the hazards from nuclear explosions in the atmosphere.

The sampling program for carbon-14 was started in late 1953 at Minneapolis, Minn., and extended in 1955 to three other locations in the Western Hemisphere: San Angelo, Tex.; Canal Zone, Panama; and São Paulo, Brazil. The Minneapolis collection was shifted in June 1958 to Sioux City, Iowa. Carbon-14 was determined by measuring the specific activity of the carbon dioxide from air collected at altitudes between 45,000 and about 100,000 feet. Air samples were collected on a monthly schedule at four altitudes at each location unless operational difficulties prevented collection. The tritium measurements were performed only on samples collected at Minneapolis, primarily in the period 1957–58.

This article presents all of the significant original data and discusses the experimental errors which lead to correction of some of the values and rejection of others. Many of the technical details are omitted (*1*).

Sampling System

Basic equipment. A balloon-borne system was developed by General Mills (*2*) to collect samples of whole air from the stratosphere. The collection system (Figs. 1 and 2) consisted of four major components: a lift balloon, a collection bag, an armored vessel, and a control unit.

The lift balloon was a nonextensible plastic film (2-mil polyethylene) balloon of the "Skyhook" type. When the lift balloon reached its full size, excess helium was vented through a duct system so that the balloon would float at a predetermined altitude during the collection period. After collection of the sample, an electrically operated trap-door-type valve in the duct released the exact amount of lift gas necessary to insure a controlled, safe descent of the system. A parachute was suspended directly beneath the balloon to help smooth the rate of descent and to prevent free fall of the equipment in case of failure of the balloon.

The collection bag was a large balloon fabricated of the same material as the lift balloon. Two sizes of bags were used, depending on the collection altitude: a bag 47.5 feet in diameter was used at altitudes above 80,000 feet, and a bag 34.6 feet in diameter was used at lower altitudes. A collection blower of the centrifugal type, powered by a 24-volt direct-current motor, was located below the collection bag.

When measurements were to be made of tritium, a measured amount of deuterium tracer, in the form of heavy water, was introduced into the collection bag at the time of sampling. To accomplish this, a dispenser containing the tracer was located between the blower and the collection bag. The dispenser was kept at a constant temperature of 95°C. A fine orifice in the dispenser was opened during the time the blower was in operation so that the water vapor was introduced uniformly throughout the collection period. The amount of tracer introduced, 2 to 5

Dr. Hagemann and Dr. Gray are staff members at Argonne National Laboratory, Lemont, Ill. Dr. Machta is on the staff of the U.S. Weather Bureau, Washington, D.C. Dr. Turkevich is on the staff of the Enrico Fermi Institute for Nuclear Studies, University of Chicago, Chicago, Ill.

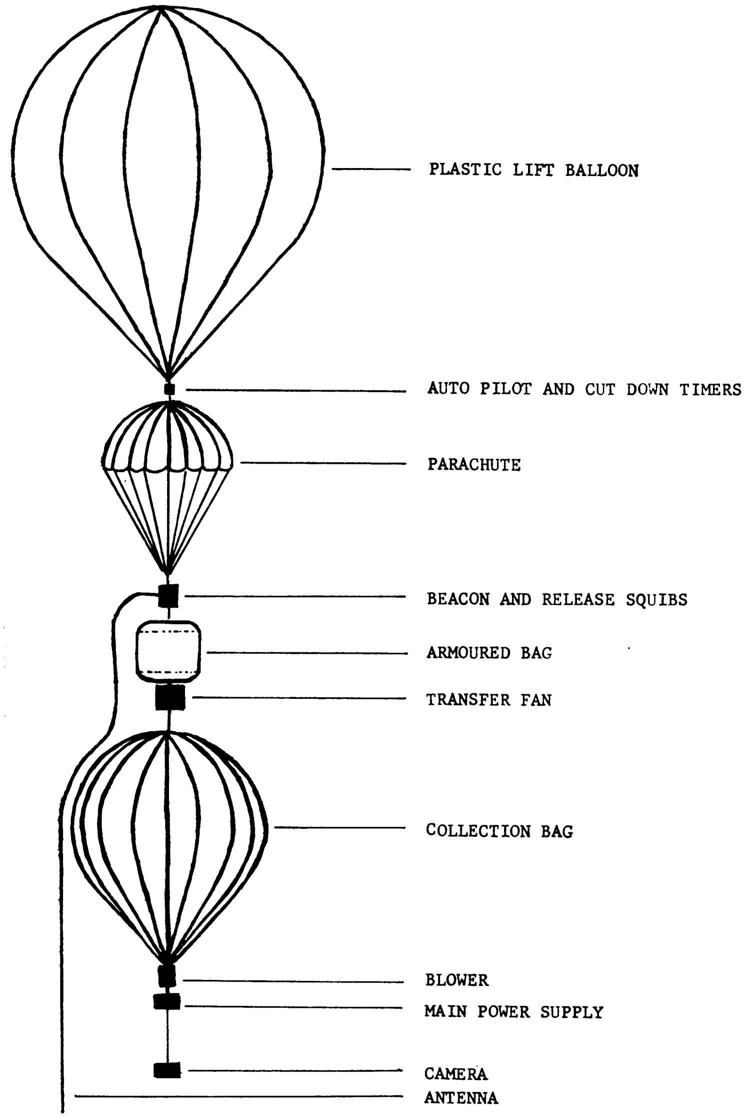
grams, was determined by weighing the dispenser before and after collection of the sample.

The final airborne sample container was a 1400-cubic-foot armored vessel to which the air sample was transferred from the collection bag during descent. It was constructed in three layers to prevent loss of the sample upon impact with the ground. The inner liner, of gas-tight 4-mil polyethylene, was covered with a nylon fabric for protection against the heavy, coarse canvas "armor" of the outer bag. An axial flow blower was used to transfer the air from the collection bag to the armored vessel.

The control unit consisted of a power supply, master control unit, and barograph, all housed in a large, insulated, red bag. An upward-viewing sequence camera suspended below the control unit provided a record of the expansion of the lift balloon and the deployment of the collection bag.

Collection procedure. The following sequence of events took place in a typical sample collection operation. The system was launched and rose to its floating altitude. The intake trap-door valves on the collection blower were opened, and the blower was turned on for the predetermined time required to fill the collection bag. After the collection blower was turned off, a trap-door valve was closed, sealing the sample in the collection bag. The collection time varied from about ½ to 1½ hours, resulting in a sample of about 2000 standard cubic feet of air. A spring-loaded valve on the balloon then opened, allowing helium to escape, and the system began its descent. At approximately 33,000 feet, about 500 standard cubic feet of the 2000 cubic feet of air collected were transferred from the bag to the armored vessel. A barometric control automatically shut off the transfer fan when the 3000-foot level was reached.

During these operations a recovery crew tracked the course of the flight visually and with the aid of signals generated by a small radio transmitter included in the balloon train. The recovery crew proceeded by truck to the impact site and transferred the air sample with a four-stage compressor from the armored vessel to high-pressure cylin-



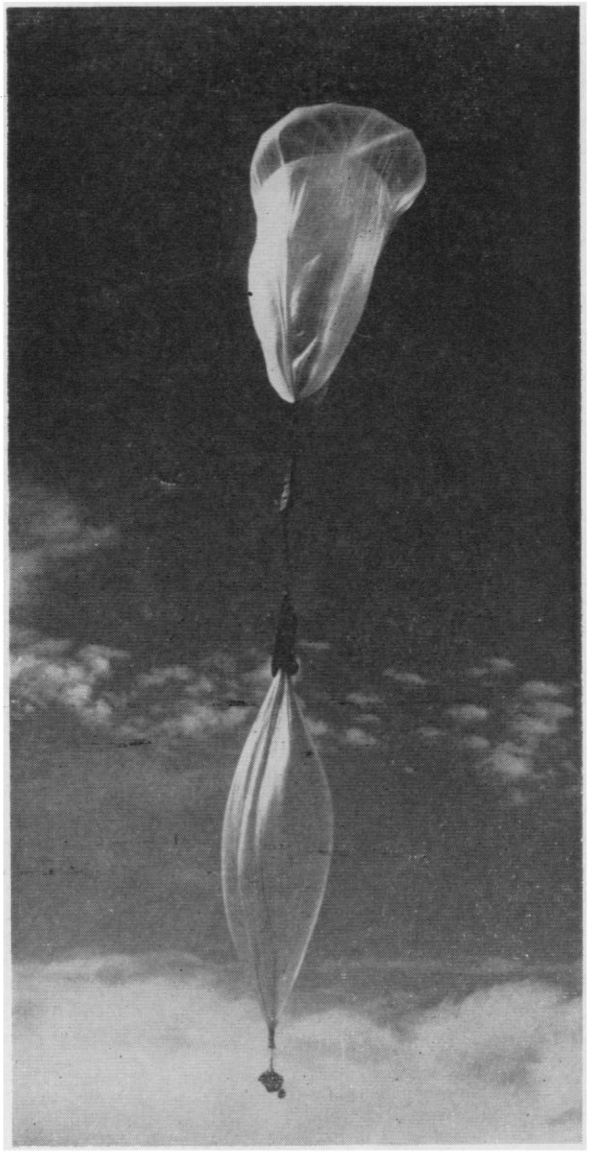


Fig. 1 (Left). Balloon train used in the collection of the stratospheric radiocarbon, carbon dioxide, and tritium samples. Fig. 2 (Right). Photograph of balloon train. See Fig. 1 for identification of visible parts of the train. The view shown is during descent, and the large collection bag has not yet transferred its air to the armored bag which hangs limply above it. The larger package at the bottom of the train is the filter apparatus which collects particulate stratospheric radioactivity. It is the size and shape of an ash can.

ders for shipment to the laboratory and subsequent processing and measurement.

Carbon dioxide was removed from some of the air samples, prior to their entering the compressor, with a trap refrigerated with liquid nitrogen. When a tritium analysis was to be made, a portion of the water vapor was frozen out of the air in a similar manner.

Carbon Dioxide

Advantage was taken of the availability of stratospheric air collected in this program to measure the concentration of carbon dioxide at high altitudes. A series of special small samples for this purpose were taken directly from the armored vessel to avoid possible alteration of the composition of the air in passage through the compressor and storage in the high-pressure cylinders. These samples were collected in small (1.7 liter) stainless-steel tanks, previously evacuated to a pressure of less than 10^{-3} mm-Hg in the laboratory. Aliquots of the air samples were taken near the start and end of the transfer from the armored vessel to the cylinders. Such samples were obtained at each of the four collection stations during the summer and fall of 1956.

The carbon dioxide concentration was determined by pumping a measured amount (500 to 1000 cm^3) of air, previously dried by passage through anhydrone, through a capillary trap cooled with liquid nitrogen. The separated carbon dioxide was distilled from a − 80°C bath into a second trap cooled with liquid nitrogen, which was part of a capillary system of calibrated volume. The carbon dioxide was allowed to warm and expand, and the pressure was measured with a constant-volume capillary manometer. At least two replicates were run on each sample. The standard error of a single determination was estimated to be 0.5 percent, based upon observed recoveries from carbon dioxide-free air to which measured volumes of carbon dioxide had been added.

A similar series of direct air samples had been collected earlier at Minneapolis during the winter of 1954. In this case single aliquots were taken in glass flasks fitted with stopcocks. The method for determining carbon dioxide was similar to that described above, but with an estimated standard error of 1.0 percent.

The results of the measurements of the stratospheric concentrations of carbon dioxide are shown in Table 1. The table indicates no significant difference in the values obtained at Minneapolis in 1954 and in 1956. The agreement between the duplicate samples collected in 1956, with a few exceptions, is good. The samples collected at the start of transfer operations at Minneapolis on 1 August 1956 and at the Canal Zone on 1 August and 4 October 1956 gave values differing widely from samples collected at the end of the transfer and from the average. These values can be ignored as being nonrepresentative. The average value at Minneapolis for all altitudes studied is 312 ppm; at San Angelo, 310 ppm; at the Canal Zone, 312 ppm; and at São Paulo, 310 ppm. The average at about 50,000 feet for all locations is 311 ppm; at about 65,000 feet, 310 ppm; at about 80,000 feet, 313 ppm; and at about 90,000 feet, 310 ppm. The average concentration of carbon dioxide at all locations at altitudes of about 50,000 feet or greater is 311 ppm with an average deviation of less than 1 percent and a range of ± 2 percent. This value is in close agreement with recent determinations of the concentration and variability of carbon dioxide in the troposphere (*3*).

Table 1. Stratospheric concentration of carbon dioxide.

Collection date	Altitude (10^3 ft)	CO_2 concentration (ppm)	
	Minneapolis, Minn.		
19 Oct. 1954	50	318	
4 Nov. 1954	50	316	
4 Dec. 1954	51	311	
27 Dec. 1954	61	308	
10 Jan. 1955	60	309	
23 Jan. 1955	60	308	
15 Oct. 1954	64	315	
27 Oct. 1954	66	312	
2 Nov. 1954	66	309	
19 Dec. 1954	65	310	
29 Dec. 1954	66	308	
27 Dec. 1954	71	308	
6 Jan. 1955	73	310	
20 Oct. 1954	79	317	
30 Nov. 1954	79	316	
6 Dec. 1954	79	311	
		Start	*End*
2 Oct. 1956	52	308	309
12 Sept. 1956	65	307	310
1 Aug. 1956	71–61	337	309
16 May 1956	83	309	310
13 Aug. 1956	80	311	323
30 Aug. 1956	90	319	315
	San Angelo, Tex.		
6 Aug. 1956	47	308	312
7 Aug. 1956	65	307	309
16 Oct. 1956	79	312	310
25 Aug. 1956	94	309	309
	Canal Zone		
7 Aug. 1956	54	319	314
1 Aug. 1956	67	258	314
21 Oct. 1956	87–83	312	318
4 Oct. 1956	97–90	346	304
	São Paulo, Brazil		
18 Aug. 1956	50	309	309
29 Aug. 1956	68–65	309	307
17 Aug. 1956	82–80	315	309
16 Aug. 1956	92–90	311	311

Carbon-14

Sample processing and measurement (*4*). The initial separation of carbon dioxide from the compressed air samples was accomplished by standard freeze-out techniques. In those cases in which freeze-out traps were used in the field, the traps were closed with valves and the separated carbon dioxide was shipped to the laboratory.

The separated carbon dioxide was purified for counting by the following procedure: (i) freezing and pumping at − 196°C; (ii) reaction with CaO at 700° to 750°C to form $CaCO_3$; (iii) pumping the $CaCO_3$ to a high vacuum at 450°C; (iv) liberating the CO_2 by heating the $CaCO_3$ to 900°C; (v) freezing and pumping again at − 196°C.

The radiocarbon was measured in proportional counters, the purified carbon dioxide being used as the counter fill. Considering the large number of samples to be measured, a sample size in the range of 1 to 2 liters of carbon dioxide was chosen as the best compromise between the convenience of sample preparation and the accuracy of the measurement.

The counters, 3 inches in diameter and 25 inches long, were constructed of stainless steel with a 1-mil stainless steel anode center wire; they had a sensitive volume of about 2.2 liters. When surrounded by a ring of anticoincidence Geiger counters and 12 inches of steel shielding, the counters, filled with 1 atm of inert carbon dioxide, had backgrounds of about 17 count/min.

As a routine procedure each gas sample was measured in at least two counters at pressures in the range of 30 to 76 cm-Hg. Each sample was usually counted for a period long enough to assure a statistical uncertainty of no more than 1 percent in the radiocarbon activity. The sensitivity of the counters was set at about 0.9 kev, using the monoenergetic x-rays from an Fe^{55} source as an internal energy standard. The number of undetected beta particles possessing less than 0.9 kev of energy was estimated to be less than 0.5 percent and was neglected. The end effect losses were experimentally determined, and the measurements were corrected appropriately. Although no direct experimental measurements were made, the wall losses were estimated to be no greater than about 1 percent, and the correction was neglected.

Results and errors. The results of the stratospheric carbon-14 measurements are listed in Table 2. The estimated errors in the altitudes shown in column 2 are ± 2500 feet for altitudes below about 70,000 feet and ± 5000 feet for the higher altitudes. When the collection occurred over a larger range of altitude than 1000 feet, the actual range is shown. All samples were stratospheric except those marked with the letter T, which were definitely tropospheric, and those marked U, which could not be definitely assigned to the stratosphere. In column 3, the measured carbon-14 specific activities are given in units of disintegrations per minute per gram of carbon. The measurement errors of the unmarked values are estimated to be ± 3 percent or less; of those marked with an asterisk, in the range ± 3 to 5 percent; and of those marked with a dagger, in the range of ± 5 to 10 percent. Values marked with the letter F were obtained from samples collected by a freeze-out trap in the field.

In a majority of samples the concentration of carbon dioxide was determined on aliquots of the compressed air, and the results are shown in column 4. Examination of these measured concentrations shows that about 60 percent of the values are between 295 and 327 ppm. About 17 percent of the values are higher than 389 ppm and only 0.5 percent are lower than 295 ppm. Comparison of the more than 50 cases where carbon-14 measurements were made for the same sample upon both field cold-trap-separated carbon dioxide and carbon dioxide separated from the compressed air showed that the specific activities of the field-trapped samples averaged about 10 percent higher than those of the compressed samples. These observations lead to the conclusion that inert carbon dioxide was introduced during the transfer of the air sample from the collection bag to the cylinder. In view of these considerations the measured specific activities were corrected somewhat arbitrarily as follows:

1) When both field trap and compressed air values were measured, the higher value of the specific activity, usually that of the trap sample, was chosen.

2) No correction was made when the measured concentration was between 295 and 327 ppm.

Table 2. Stratospheric concentration of carbon-14. In column 2, the letter *T* indicates that the sample was definitely collected in the troposphere. The letter *U* indicates uncertainty whether the sample was collected in the stratosphere or troposphere. Unmarked values indicate that the sample was collected in the stratosphere. In column 3, an asterisk indicates a measurement error of 3 to 5 percent, a dagger a measurement error of 5 to 10 percent. Unmarked values have measurement errors less than 3 percent. Values marked with the letter *F* indicate that carbon dioxide was separated in the field with a freeze-out trap. A carbon half-life of 5600 years and a concentration of carbon dioxide of 311 ppm were used to calculate the values given in column 5. After correction for variation in CO_2 concentration (see text) a natural background of 71×10^5 atoms per gram of air was subtracted to give excess carbon-14.

Collection date	Altitude (10^3 ft)	Observed specific activity (disintegration/min per g of carbon)	Measured CO_2 concentration (ppm)	Excess C^{14} (10^5 atom/g of air)
Minneapolis, Minn. (lat. 45° N)				
1953				
24 Sept.	81	16.8*		29
9 Oct.	82	17.8		36
5 Nov.	82	26.8†		89
12 Nov.	64	22.1*		61
17 Dec.	53	16.1†		25
23 Dec.	64	15.1*		19
1954				
28 Jan.	80	23.9*		72
28 Jan.	52	15.0*		18
28 Jan.	65	19.5*		46
6 Feb.	68–66	21.8		59
16 Feb.	79	30.8		113
22 Feb.	47	13.3*		9
22 June	98	27.6*		94
8 July	82	28.6		100
10 Sept.	49	16.1*		25
22 Sept.	79	36.8		149
15 Oct.	64	42.2		181
19 Oct.	50	14.3*		14
20 Oct.	79	27.3*		93
27 Oct.	66	46.9*		209
2 Nov.	66	52.0		240
8 Nov.	67	28.4		99
22 Nov.	95	36.1		145
22 Nov.	66	33.7		131
30 Nov.	79	104.6		554
4 Dec.	51	13.5*		10
6 Dec.	79	122.2		658
10 Dec.	65	34.1		133
19 Dec.	65	47.6		213
27 Dec.	61	31.8		128
27 Dec.	71	79.3*		402
29 Dec.	66	50.0		228
1955				
6 Jan.	49U	27.5*		94
6 Jan.	73	68.7		339
29 Jan.	66	30.8		113
30 Jan.	80	56.5		267
8 Feb.	71–68	66.4		326
8 Feb.	65	38.3		158
14 Feb.	59	29.0		102
1 Mar.	66	60.6		291
2 Mar.	49U	15.1*		19
9 Mar.	92	23.0		66
16 Mar.	77	67.8		333
16 Mar.	59U	20.8		53
28 Mar.	70	73.5		368
2 Apr.	60U	22.2		62
7 Apr.	50	23.5		70
25 Apr.	70	30.0†		108
25 Apr.	78	41.9		180
6 May	49–47U	13.2†		7
11 May	77	56.9		269
18 May	76	68.6		339
20 May	70	61.4		295
14 June	61–58	26.7		88
15 June	76	62.8		304
23 June	71	61.2		294
7 July	50	14.5*		15
18 July	76	70.4		350
20 July	59	17.8		36
8 Aug.	68	43.4		188
10 Aug.	60	17.6		34
11 Aug.	78	42.1F		160
1 Sept.	57	25.4		81
7 Sept.	65	32.6		124
23 Sept.	77	71.7F		322
24 Sept.	59	24.7		76
28 Sept.	48U	15.8*		24
10 Oct.	80	55.3F		232
13 Oct.	58	31.5		118
18 Oct.	80	56.1F		236
21 Oct.	48U	13.7*	331	9
26 Oct.	69	35.4*F		123
28 Nov.	50	18.7*F		31
30 Nov.	59	48.6*F		195
30 Nov.	69	31.4†		116
5 Dec.	70	37.6F		135
6 Dec.	48	18.9†F		33
9 Dec.	56U	31.6		118
9 Dec.	69	33.8		131
12 Dec.	49	17.1	316	23
21 Dec.	83	54.6F		228
1956				
4 Jan.	59	24.5F		63
12 Jan.	50	25.4F		68
13 Jan.	82	51.0*F		208
19 Jan.	70	55.2†F		231
30 Jan.	59U	32.0		120
7 Feb.	68	45.6	323	179
18 Feb.	81	67.3	294	298
27 Feb.	91	85.1F		395
28 Feb.	52	23.5	306	58
29 Feb.	50	16.1†F		17
8 Mar.	48	23.4F		57
16 Mar.	66	35.4	312	123
26 Mar.	94	59.6†F		255
30 Mar.	83	70.9F		317
9 Apr.	95	40.5	313	151
10 Apr.	68	37.8	306	136
30 Apr.	49	18.6	393	
7 May	66	36.8	307	131
16 May	83	53.1	240	220
21 May	49	17.2	352	37
27 May	92	58.0	326	247
5 June	80	51.4	296	211
8 June	65	14.0†	314	6
11 June	94	52.0	308	214
19 June	54	16.4	593	
21 June	68	14.2*	316	7
21 June	88	48.9	339	222
24 June	80	41.9	326	159
2 July	80	41.8	282	158
6 July	65	39.4	307	145
12 July	50	17.5	325	25
16 July	90	38.1	443	
1 Aug.	71–61	28.3	573	
5 Aug.	50	15.8	595	
13 Aug.	80	43.1	412	
30 Aug.	90	63.5F		277
12 Sept.	65	31.9	305	104
14 Sept.	90	54.8	349	265
18 Sept.	80	46.6	289	184
23 Sept.	50U	18.3	282	29
2 Oct.	52U	25.9	354	91
12 Oct.	66	32.6	312	108
16 Oct.	84	55.6	336	259
19 Nov.	80	52.4	395	
23 Nov.	66	12.8	314	– 1
27 Nov.	48	13.3	331	7
13 Dec.	52U	31.8	328	113
20 Dec.	83	39.6	408	
1957				
10 Jan.	67	69.0	311	307
29 Jan.	90–85	65.1	324	286
6 Feb.	50U	14.9	529	
13 Feb.	82	63.3	308	274
19 Feb.	67	57.5	322	244
9 Mar.	68	59.4	356	300
17 Mar.	47	29.5	351	112
30 Mar.	83	47.1	341	212

Collection date	Altitude (10^3 ft)	Observed specific activity (disintegration/min per g of carbon)	Measured CO_2 concentration (ppm)	Excess C^{14} (10^5 atom/g of air)
10 Apr.	84	43.4	322	167
14 Apr.	67	55.5	460	
15 Apr.	48	28.0F		82
25 Apr.	48U	26.4F		74
27 May	63	28.1	392	
28 May	49	16.2*F		18
29 May	73	46.9	308	186
19 June	91	37.2	365	169
20 June	76	50.4F		205
27 June	50	24.1F		61
11 July	50	15.4		13
11 July	62	33.7F	356	141
15 July	77	57.2F		242
19 July	85–82	48.5	363	240
23 July	65	32.5	348	128
2 Aug.	49U	11.3*	370	3
10 Aug.	90	38.6	376	183
16 Aug.	71	59.2F		253
22 Aug.	82	50.3F		205
24 Aug.	65	53.9F		224
4 Sept.	46	16.2	334	27
6 Sept.	77	47.1	407	
25 Sept.	89	58.0F		247
2 Oct.	79	46.5	316	184
2 Oct.	49	16.6	342	29
26 Oct.	65	62.8*F		273
29 Oct.	90	50.1F		203
5 Nov.	91	47.9		214
7 Nov.	108–105	41.6	333	173
25 Nov.	79	50.7	327	207
26 Nov.	50	16.7	326	20
30 Nov.	64	51.1F		209
4 Dec.	47	21.0F		44
12 Dec.	81	49.4F		200
19 Dec.	88	51.5F		211
21 Dec.	64	45.1F		176
1958				
8 Jan.	48U	15.2	328	17
10 Jan.	64	47.1F		187
22 Jan.	78	49.1F		198
31 Jan.	92	40.2	369	190
5 Feb.	90–88	49.8	328	217
7 Feb.	77	51.3F		210
13 Feb.	46	25.5	353	88
20 Feb	64	44.9*F		175
6 Mar.	47	19.1		43
12 Mar.	64	38.9	340	162
25 Mar.	81	42.4F		161
26 Mar.	93	41.1	341	176
7 Apr.	90	44.6		195
8 Apr.	78	47.1F		187
9 Apr.	47	33.6F		113
16 Apr.	65	48.5†F		195
9 May	91	45.2F		177
12 May	48	23.2F		56
15 May	78	38.8F		142
6 June	91	28.9		202
7 June	62	49.8†F		101
11 June	47U	25.3F		68
17 June	77	47.8F		191
20 June	61	40.9F		153
San Angelo, Tex. (lat. 32° N)				
1955				
22 Mar.	50U	15.9		21
24 Mar.	62	26.8		85
29 Mar.	71	70.7		339
30 Mar.	74	64.4		303
5 Apr.	62	25.7		78
8 Apr.	72	50.8		224
14 Apr.	78	68.5		327
21 Apr.	53–51	15.1		17
25 Apr.	71	41.7		171
13 May	70	42.8*		177
14 May	53–50U	16.6		25
16 May	79	72.6		350
17 May	63	20.4		48
23 June	71	44.8		189
5 July	77	48.7		212
12 July	61	20.2		47
2 Aug.	53–51	14.9†		16
3 Aug.	70	31.1		109
7 Aug.	59	20.7		49
10 Aug.	78	40.1		162
8 Sept.	71	28.8		96
9 Sept.	60	17.2*		29
27 Sept.	77	40.6		164
28 Sept.	93	62.2		290
29 Sept.	93	62.7		293
19 Oct.	71	21.3		53
25 Oct.	60	20.4		48
3 Nov.	83–80	42.7	323	163
4 Nov.	71	54.1	301	225
9 Nov.	50	20.8	299	43
21 Nov.	62	20.1	310	39
25 Nov.	81	55.8*	376	299
4 Dec.	51	13.1	377	16
7 Dec.	62	25.3	382	100
10 Dec.	82	48.2	329	208
12 Dec.	72	37.7	331	149
1956				
25 Jan.	50U	15.2	312	12
6 Feb.	84	56.1	325	236
20 Feb.	66	29.7	310	92
8 Mar.	69	12.5*	303	– 3
16 Mar.	83	63.2	316	275
22 Mar.	50	15.7	291	15
23 Mar.	98	69.8	311	311
4 Apr.	95	69.6	336	341
14 May	52–51	13.2	320	1
16 May	68–63	40.7	617	
17 May	84	58.1	314	247
8 June	50U	15.7†	313	15
18 June	65	28.9	327	87
19 June	81	50.7	338	231
6 July	52	16.0	398	
8 July	83	44.5	351	204
18 July	66	28.3	373	115
22 July	93	51.8	379	275
6 Aug.	47T	14.4	556	
7 Aug.	65	38.1	619	
9 Aug.	84	41.5	483	
11 Aug.	94	57.8	331	266
25 Aug.	94	52.2	328	231
5 Sept.	80	48.1	405	
7 Sept.	94	23.6	475	
8 Sept.	50T	14.2	388	26
11 Sept.	65	27.1	333	87
5 Oct.	96	44.8	374	224
11 Oct.	65	32.1	480	
16 Oct.	79	34.2	1880	
22 Oct.	47T	13.3	690	
6 Nov.	94	53.6	312	223
13 Nov.	66	38.2	312	138
16 Nov.	44	13.4*	329	6
17 Nov.	77	51.7	298	212
15 Dec.	63	40.5	500	
17 Dec.	95	52.6	307	217
1957				
12 Jan.	100	44.7	427	
17 Jan.	67	42.8	381	216
1 Feb.	97	42.5	308	162
10 Feb.	80	69.7	304	311
25 Feb.	67–65	30.6	348	117
7 Mar.	96	46.4	338	205
12 Mar.	81	68.7	340	340
18 Mar.	50	13.9*	411	
23 Apr.	81	51.2	340	235
29 Apr.	66	52.9	321	219
2 May	82	65.5	310	288
3 May	66	43.4	294	167
19 May	89–87	45.0	243	176
21 May	49U	13.8*	301	5
6 June	91	49.5	311	200
10 June	67	44.2	296	171
12 June	48U	15.7*	326	15
19 June	82	57.4	343	275
7 July	88	56.7	392	
9 July	67	36.3	306	128
11 July	85–78	55.9	296	235
13 July	48	14.0	302	6
6 Aug.	65	37.7	364	171
7 Aug.	91	38.2	483	
9 Aug.	82–78	58.4	308	249
20 Aug.	50	14.8	418	
4 Sept.	90	53.2	333	241
6 Sept.	81	40.1	306	149
11 Sept.	65	24.4	383	94
13 Sept.	49	13.7	376	20
1 Oct.	50	10.1*	402	
3 Oct.	91	27.7	475	
4 Oct.	83	54.1	323	225
5 Oct.	65	24.4	337	74
2 Nov.	90	57.7	311	245
14 Nov.	48	19.5	309	36
20 Nov.	84	39.6	309	146
26 Nov.	66	46.9	310	186
2 Dec.	82–80	54.3	313	226
4 Dec.	49T	13.2	306	1
7 Dec.	90	55.4	315	232
10 Dec.	67	40.0	320	148
1958				
7 Jan.	81–79	55.6		252
8 Jan.	90	46.5	348	214
9 Jan.	50	15.5	349	24
15 Jan.	62	42.7	331	178
1 Feb.	50U	13.5*	324	3
2 Feb.	92	49.2	330	215
7 Feb.	65	40.5	309	151
15 Feb.	80	49.7	313	201
3 Mar.	92	43.0*	311	165
7 Mar.	80	49.4	314	200
9 Mar.	67	41.7	308	157
10 Mar.	51U	19.2	309	34
1 Apr.	92	42.3	317	161
3 Apr.	65	28.9	315	87
7 Apr.	82	47.6	313	190
8 Apr.	49	14.8	318	10
3 May	48U	20.2*	318	40
4 May	66	36.5	317	129
7 May	92	45.7	312	179
10 May	49	26.0	314	71
19 May	81	53.3F		221
3 June	81	47.2		204
5 June	89	43.6F		168
7 June	65	47.2		204
8 June	52U	13.5†F		3
2 July	82	46.7F		185
3 July	67	41.3F		155
12 July	50U	24.0F		60
18 July	89	45.3		192
1 Aug.	50U	16.8*		27
2 Aug.	92	44.7		189
3 Aug.	66	38.8		155
5 Aug.	81	52.7F		218
1 Sept.	90	47.4F		189
3 Sept.	80	49.2F		199
4 Sept.	51U	17.7F		26
9 Sept.	66	38.0F		137
Canal Zone, Panama (lat. 9° N)				
1955				
30 Mar.	75	29.7		112
31 Mar.	50–48T	12.3*		4
11 May	69	21.5		61
12 May	63	12.1*		3
27 May	79	40.3		177
11 June	51U	13.4†		11
13 June	61	13.1		10
29 June	79	36.3		152
30 June	70–69	19.2		47
7 July	78	32.3		127
18 July	51T	14.0		15
28 July	62	14.0*		15
30 July	70	13.7†		13
6 Aug.	77	27.5		98
9 Aug.	60	17.8		39
26 Aug.	52	13.4		11
10 Sept.	77	16.6*		31
13 Sept.	70	12.9*		9
22 Sept.	94–91	46.5		215
18 Oct.	92–89	44.2		200
28 Oct.	77	26.3		90
30 Oct.	61	14.9*		21
2 Dec.	83	26.1	317	72
11 Dec.	51T	3.2†	1500	
14 Dec.	71	18.2	367	47
1956				
20 Jan.	50T	11.1*	336	– 5
21 Jan.	62	15.4	325	13
12 Feb.	62	18.6	325	31
3 Mar.	52T	12.0*	321	– 5
4 Mar.	71	11.7*	355	2
14 Mar.	51	11.7*	345	0
28 Mar.	80	22.3	309	51
4 Apr.	52T	11.2	341	– 4
5 Apr.	68	19.7	317	37
9 Apr.	82	18.0	309	28
10 Apr.	93	20.7	307	42
6 May	50T	12.2	368	8
15 May	81	17.8	396	
22 May	61	16.6	422	

Collection date	Altitude (10^3 ft)	Observed specific activity (disintegration/min per g of carbon)	Measured CO_2 concentration (ppm)	Excess C^{14} (10^5 atom/g of air)
1 June	51U	12.5*	351	6
11 June	66	16.7	439	
15 June	82–80	19.2	716	
6 July	55	11.2†	365	1
7 July	67	23.4	326	57
1 Aug.	67	32.7	336	122
7 Aug.	54T	12.2*	301	– 4
4 Sept.	52	12.7*	329	3
6 Sept.	69–66	91.3	343	480
4 Oct.	97–90	27.5	534	
15 Oct.	67	37.1	381	178
16 Oct.	54	11.0†	338	– 6
21 Oct.	87–83	35.3	295	122
1 Nov.	51T	11.9*	348	2
1 Nov.	70	50.4	321	205
19 Nov.	92	29.2	350	109
27 Nov.	81–79	34.7	330	131
1 Dec.	52T	10.7*	382	1
4 Dec.	93–88	29.8	359	117
10 Dec.	67	24.1	372	87
1957				
20 Feb.	84–82	47.5	361	231
21 Feb.	51T	11.3*	352	– 1
22 Feb.	66	17.7	343	36
1 Mar.	77	28.2	552	
3 Mar.	65	12.3†	377	10
4 Mar.	47T	3.9†	459	
7 Mar.	91–87	29.1	341	103
4 Apr.	82	57.1	388	319
11 Apr.	66–64	15.3*	296	13
13 Apr.	50T	10.3*	374	– 4
4 May	65	10.9	351	– 3
7 May	49T	9.5	353	– 12
10 May	78	43.6	316	167
22 May	65	11.2†	378	3
7 June	81–79	47.3	361	230
9 June	65	22.4	309	52
1 July	49U	12.5*	355	7
14 July	67	26.6	321	75
19 Aug.	66	23.2	368	79
20 Aug.	50U	13.2	424	
20 Sept.	65	21.6	327	47
8 Oct.	89	48.4	324	194
21 Oct.	49T	12.9	322	0
23 Oct.	67	20.4	312	40
30 Oct.	80	29.4	321	90
1 Nov.	82	29.7	534	
3 Nov.	89	27.1	324	77
5 Nov.	52T	13.2	319	1
6 Nov.	66	24.1	319	61
1 Dec.	46T	12.2	319	– 4
2 Dec.	90–88	31.0*	363	127
4 Dec.	66	20.9	396	
15 Dec.	78–76	27.9	323	81
1958				
2 Jan.	51T	10.8†	334	– 8
5 Jan.	81	28.1	381	118
13 Jan.	92	37.0	338	140
15 Jan.	65	40.2	348	145
5 Feb.	67	26.2	327	73
10 Feb.	82	30.7	327	97
16 Feb.	93	18.9	515	
4 Mar.	92	30.3	347	114
8 Mar.	50T	12.3†	347	4
1 Apr.	94	39.9	358	181
3 Apr.	52T	13.1*	401	
4 Apr.	81	37.6	369	173
14 Apr.	65	10.0†	512	
1 May	65	22.9	322	54
2 May	51T	14.6	348	19
4 May	85	33.2	334	124
9 May	80	42.2	337	179
2 June	65	24.4	328	70
6 June	81	45.7	380	234
8 July	61	44.9	332	192
14 July	51U	11.3*	459	
1 Aug.	65	183.3	313	933
6 Aug.	51T	15.6	244	14
3 Sept.	65	64.6F		283
8 Sept.	52	14.3		16
São Paulo, Brazil (lat. 23° S)				
1955				
22 Mar.	61	13.4*		6
29 Mar.	50T	13.9*		9
17 Apr.	62	12.7*		3
26 Apr.	79	25.1		73
27 Apr.	70	17.3		29
28 Apr.	52T	14.3*		11
9 May	78	22.4		58
11 May	60	13.2*		1
12 May	50U	12.9*		4
27 May	69	15.2*		16
14 June	78	21.8*		54
15 June	61	12.8†		3
5 July	79	24.2		69
7 July	71	16.0		21
10 July	62	11.2		– 7
3 Aug.	78	21.2*		51
6 Aug.	61	14.7		14
30 Aug.	70	14.5		12
14 Sept.	60	12.9		4
28 Sept.	90	98.0		493
11 Oct.	91	105.7		537
12 Oct.	77	33.7		123
13 Oct.	68	17.9		32
30 Oct.	61	14.4*	398	
1 Nov.	76	37.7	321	136
20 Nov.	69	20.6	305	42
25 Nov.	52U	15.3	301	13
26 Nov.	62	13.7†	317	4
2 Dec.	75–69	20.7	589	
6 Dec.	52T	11.7*	434	
7 Dec.	82–80	39.2	306	144
8 Dec.	62	15.7	308	15
27 Dec.	81	36.4	278	128
1956				
10 Jan.	62	15.6*	375	31
11 Jan.	71	17.6	292	25
12 Jan.	81	39.3	298	144
16 Jan.	52T	13.4†	285	2
1 Feb.	52T	13.5	308	3
6 Feb.	62	14.7	300	10
7 Feb.	69	22.3	305	51
24 Feb.	81	26.9	327	76
5 Mar.	80	26.2	281	73
13 Mar.	52T	12.8	276	– 1
14 Mar.	70	17.7	307	26
3 Apr.	83–81	21.7	380	74
26 Apr.	96	33.7	399	
27 Apr.	53–50U	12.8†	296	– 1
18 May	81	18.1	298	28
19 May	68–65	15.6	291	14
20 May	91	22.0	313	50
21 June	51	14.5	310	8
23 June	68–65	25.2*	377	96
30 June	80	19.3	380	59
17 July	50U	15.1	517	
19 July	67–65	23.2	425	
20 July	80	47.4	429	
22 July	90	42.6	300	162
16 Aug.	92–90	23.7	292	59
17 Aug.	82–80	37.1	308	132
18 Aug.	50T	14.7	282	10
29 Aug.	68–65	26.4	310	74
11 Sept.	53–50U	15.1	304	12
12 Sept.	68–65	45.1	407	
15 Sept.	90	34.8	293	120
16 Sept.	80	30.7	380	134
2 Oct.	54–50U	13.8	326	5
4 Oct.	67–65	22.7	306	53
26 Oct.	90	24.5*	344	77
29 Oct.	81	50.8	322	207
30 Oct.	91	37.7	301	136
1 Nov.	90	19.8	314	37
19 Nov.	81	29.4	554	
10 Dec.	90–87	26.0	317	71
12 Dec.	90	27.5	332	90
21 Dec.	66	21.2	331	52
1957				
18 Jan.	90	43.2	344	191
28 Jan.	90	35.3	389	170
29 Jan.	84–80	33.4	424	
11 Feb.	93–90	36.1	419	
12 Feb.	83–80	32.8		118
16 Feb.	68–65	12.8*	468	
19 Feb.	50T	11.0*	379	2
19 Mar.	90	38.4	308	139
20 Mar.	80–78	24.7	303	64
25 Mar.	66	21.2	298	45
26 Mar.	50–46	13.1	295	1
10 Apr.	90	35.2	413	
25 Apr.	84	38.3	308	139
26 Apr.	68–66	19.0†	307	33
27 Apr.	49T	12.8*	313	– 1
14 May	65	19.4	313	35
15 May	95	39.5	297	145
17 May	81	39.6	320	146
18 May	50T	13.2	325	1
19 May	80	37.6*	308	135
31 May	80	47.0	301	186
1 June	90	37.9	404	
2 June	64	17.8	349	39
13 June	94	34.2	306	116
14 June	81–78	37.6	331	148
15 June	49T	12.2*	410	
9 July	66	15.8	481	
10 July	93	41.0	308	154
11 July	93	39.9	363	185
12 July	80–74	39.1	298	143
13 July	49U	14.3*	345	16
3 Aug.	68	33.7	298	114
5 Aug.	49U	16.2*	522	
8 Aug.	80	43.3	313	166
10 Aug.	90	39.7	495	
16 Aug.	89	35.4	357	152
17 Aug.	90	38.5	369	179
5 Sept.	49U	15.2*	339	20
6 Sept.	65	30.2	310	94
10 Sept.	80	39.3	311	144
12 Sept.	81	37.4	319	134
15 Sept.	87	36.1	314	127
21 Nov.	92	35.3	326	122
22 Nov.	82–75	37.7	316	136
24 Nov.	80	32.9	315	109
25 Nov.	65	28.4	322	85
10 Dec.	82–80	30.8	323	98
12 Dec.	66	32.2	320	105
14 Dec.	51U	12.7	312	– 1
19 Dec.	96–93	42.6	323	162
1958				
7 Jan.	79	28.5†	326	85
8 Jan.	89	36.9	323	131
10 Jan.	65	27.7	391	
11 Jan.	51U	12.4*	337	3
3 Feb.	66	25.9*	314	71
9 Feb.	80	39.7	304	146
10 Feb.	90	37.0	316	132
5 Mar.	80	38.4	313	139
6 Mar.	90	38.8	323	142
7 Mar.	80	35.4	312	128
13 Mar.	66	22.9	320	54
14 Mar.	50T	12.8	308	– 1
2 Apr.	52U	15.1	310	12
3 Apr.	69	34.4	325	117
4 Apr.	81	33.1	316	110
5 Apr.	90	34.7	314	119
9 May	65	23.0†	329	62
11 May	89	33.8	308	114
12 May	51T	13.9†	316	5
16 May	88	38.6	317	140
17 May	80	38.7	325	151
4 June	64	22.5	316	52
5 June	88	41.4	319	156
8 June	80	37.3		143
10 June	48T	15.5		18
13 June	79	40.8F		153
9 July	90	36.1F		127
10 July	63	33.3		120
11 July	80	36.3		138
13 July	52U	16.0		21
5 Aug.	93	37.5*F		134
7 Aug.	81	36.8		141
8 Aug.	53T	17.8F		27
9 Aug.	65	39.1		154
2 Sept.	95	31.9		113
4 Sept.	53U	13.2		5
Sioux City, Iowa (lat. 42.5° N)				
1958				
5 July	50U	17.7	314	26
6 July	66	47.3	301	188
7 July	81	46.1	369	229
28 July	93	45.2*	334	195
2 Aug.	90–88	43.7	317	168
3 Aug.	66	45.5	313	178
7 Aug.	49	19.2F		34
9 Aug.	80	48.7F	315	196
1 Sept.	88	44.5	329	187
1 Sept.	50U	17.6	311	25
8 Sept.	62	40.3		150
15 Sept.	83	48.0		192

3) When the measured concentration was between 327 and 389 ppm, the specific activity was corrected by the following ratio: measured concentration/311.

4) No correction was made in the few cases in which the measured concentration was less than 295 ppm on the assumption that such low values were due to sampling errors which did not affect the specific activity.

5) Samples whose measured concentration was greater than 389 ppm were considered too unreliable to be used.

6) For samples for which no measurement of concentration was available, the specific activity values were corrected by the following factors, representing the ratios of the average of the measured values of the compressed air samples at each station (excluding values greater than 389 ppm) to 311 ppm: Minneapolis, 1.09; San Angelo, 1.06; Canal Zone, 1.12; São Paulo, 1.05; Sioux City, 1.00.

An investigation is being made of the C^{13}/C^{12} ratios of the carbon dioxide samples in the hope that more exact corrections may be applied to the data.

Column 5 of Table 2 lists what are considered to be the best values for the concentration of artificially produced carbon-14—that is, the excess over the natural cosmic ray-produced carbon-14 background, expressed in units of 10^5 atoms of carbon-14 per gram of air. These values were obtained from the measured specific activity values, which were corrected for variations in concentration in the manner just described and converted to units of 10^5 atoms per gram of air on the basis of a carbon-14 half-life of 5600 years and a uniform stratospheric concentration of carbon dioxide of 311 ppm. The assumed natural background is 71×10^5 atoms of carbon-14 per gram of air, corresponding to a specific activity of 13 disintegrations per minute per gram of carbon. The uncertainty of this assumed stratospheric background is probably small compared to the magnitude of most of the excess carbon-14 observations and does not seriously affect the integrated inventory of artificially produced carbon-14 in the stratosphere.

These excess carbon-14 results are presented graphically in Fig. 3, which shows the measurements in four altitude groups as a function of time at the four collection stations. The lines connecting the points, especially in time regions of few measurements, are for visual aid only. Indicated at the top of the figure

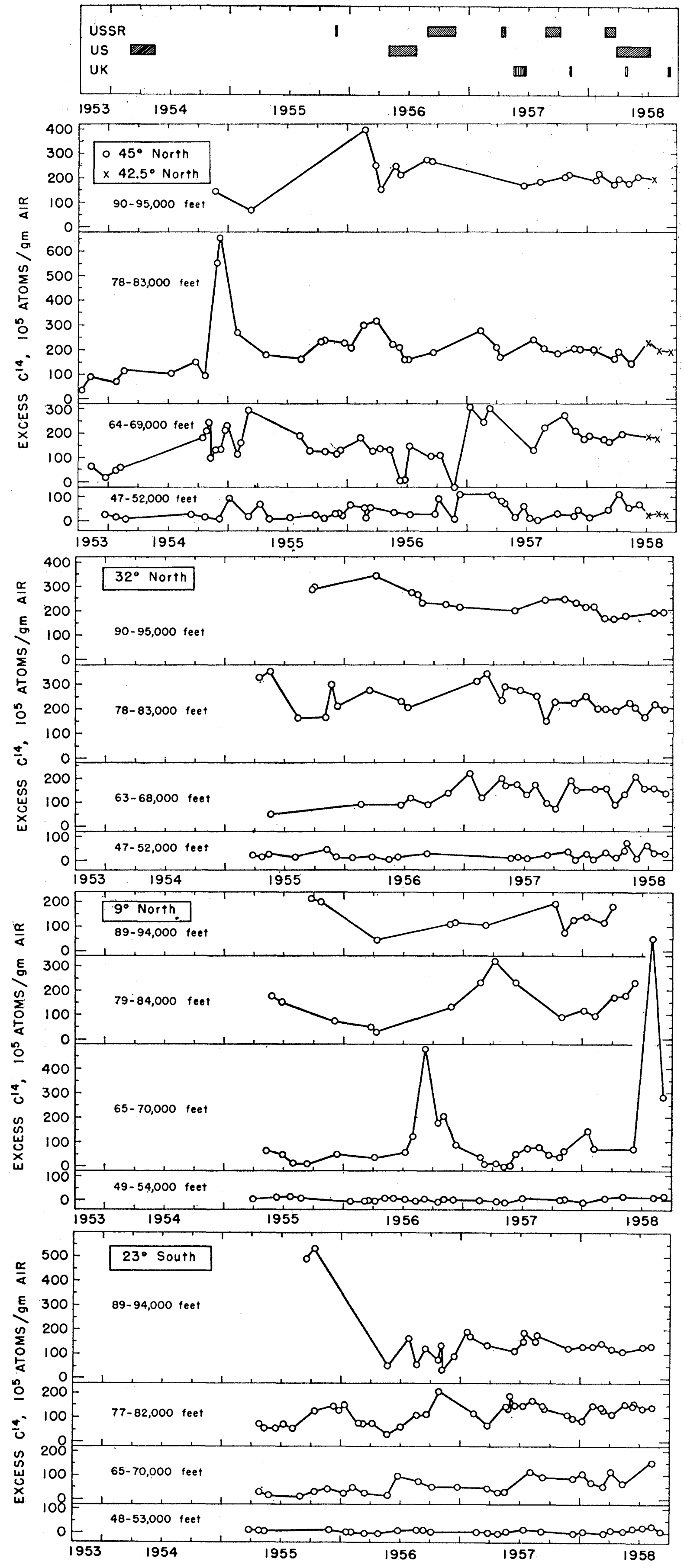


Fig. 3. Excess carbon-14 as a function of time at various altitudes and latitudes. Bars at top of figure indicate large stratospheric injections of radiocarbon by nuclear tests.

are the times of known large stratospheric injections of carbon-14 by nuclear detonations. These have been taken from the compilation of Telegadas (*5*). The detonations of each country are indicated separately. In the case of the United States, only the Pacific test series are shown. It should be borne in mind that the U.S.S.R. tests have been carried out north of 40°N; the U.S. tests have been carried out at 11°N; and the U.K. tests have been carried out primarily at 2°N. The lengths of the bars show the duration of the test series and do not necessarily bear any relationship to the amount of carbon-14 injected into the stratosphere.

Tritium

Sample processing and measurement. The recovered samples of water from the stratospheric air collections, usually 1 to 10 milliliters in volume, were split into two fractions. The concentration of the deuterium was measured in one fraction by mass spectrometric or infrared analysis, the concentration of the deuterium in the original tracer water having been determined previously. About 0.5 to 0.75 milliliter of the second fraction was reduced to hydrogen gas by magnesium, and the tritium content of the gas was measured with a Geiger counter surrounded by anticoincidence counters and steel shielding. The majority of the samples had counting rates at least one order of magnitude greater than background.

The total tritium collected was obtained by multiplying the tritium-to-deuterium ratio in the samples by the total deuterium added as tracer at the time of collection. The volume of air collected was estimated by measuring the total number of revolutions of the blower, which had been calibrated under simulated conditions, and by photographing the inflated collection bag to estimate its dimensions. The mass of air sample was calculated from the density of the air at the collection altitude.

Results and errors. The concentration of tritium per gram of air may be ascertained even though water is added or subtracted indiscriminately prior to the laboratory separation into the two fractions, provided that the added water contains no appreciable tritium or deuterium (tropospheric water would produce no significant error). However, the tritium-to-hydrogen ratio in the stratosphere is not determined by this method. It can only be inferred from the water vapor content of stratospheric air, which is poorly known.

Following is an evaluation of the errors associated with the tritium measurements. The amount of tracer added during the collection of the air is estimated to be accurate to about ± 1 percent. The deuterium concentrations are accurate to ± 4 percent. Errors in the measurement of the tritium activity are about ± 10 percent; about half are attributable to statistical counting errors and half to calibration errors. The tracer deuterium oxide contained less than 250 disintegrations of tritium per minute per gram. This was normally only about 5 percent of the total tritium in a sample and, therefore, introduced only a small correction.

The method of determining the volume of air collected is estimated to have a standard error of about ± 5 percent. However, the density must be known to convert the volume to mass of air and, due to uncertainties in the absolute altitude of the collection, the mass of air collected is not known to better than ± 15 percent below 70,000 feet and ± 25 percent above 70,000 feet. Thus, the error in determining the mass of air containing the tritium is the predominant error.

Combining all the sources of error, it is estimated that a single determination of the tritium concentration should be accurate to ± 30 percent, with greater accuracy at the lower levels of the stratosphere.

A few early results were obtained in the period between June 1955 and August 1956 and are listed in Table 3. In these early collections, tracing was performed by adding deuterium oxide to the collection bag before the balloon was launched. The error associated with these measurements may be somewhat larger than the errors associated with similar measurements made during the period 1957–58 obtained by the collection method described above. The latter results are given in Table 4.

Discussion

General comments. Figure 3, showing the excess of carbon-14 at the four locations as a function of time, presents a different pattern at the different altitudes. The excess carbon-14 values are small at the lowest altitudes (approximately 50,000 feet) and show little fluctuation with time. On the other hand, the data at the higher altitudes show large excess carbon-14 concentrations and marked variability. Some of this variability may be due to experimental errors. However, several striking peaks —for example, those at the Canal Zone at 65,000 to 70,000 feet in September 1956 and July 1958—are closely related to known injections. On the other hand, the large peaks at Minneapolis at 80,000 feet in December 1954 and at São Paulo at 90,000 feet in October 1955, 6 and 16 months after United States tests in the Pacific, indicate a lack of homogeneity of the stratosphere long after the injection of the carbon-14 has occurred.

Table 3. Tritium concentrations in 1955–56 in the stratosphere above Minneapolis.

Collection date	Altitude (10^3 ft)	Tritium (10^5 atom/g of air)
30 Nov. 1955	59	78
4 Jan. 1956	59	62
26 Oct. 1955	69	43
19 Jan. 1956	70	72
15 June 1955	76	82
11 Aug. 1955	78	140
23 Sept. 1955	77	115
10 Oct. 1955	80	89
18 Oct. 1955	80	38
26 Mar. 1956	94	86
30 Aug. 1956	90	107

The concentrations at Minneapolis and San Angelo in the Northern Hemisphere are greater than at São Paulo in the Southern Hemisphere. Broecker and Walton (*6*) also found higher carbon-14 concentrations in Northern than in Southern Hemisphere tropospheric air,

Table 4. Tritium concentrations in 1957–58 in the stratosphere above Minneapolis.

Collection date	Altitude (10^3 ft)	Tritium (10^5 atom/g of air)
28 May 1957	49	11
27 June 1957	50	25
11 July 1957	50	9
6 Mar. 1958	47	27
9 Apr. 1958	47	42
16 Aug. 1957	71	118
24 Aug. 1957	65	107
26 Oct. 1957	65	111
30 Nov. 1957	64	89
21 Dec. 1957	64	123
10 Jan. 1958	64	89
20 Feb. 1958	64	69
7 June 1958	62	58
20 June 1958	61	85
20 June 1957	76	71
15 July 1957	77	107
22 Aug. 1957	82	59
22 Jan. 1958	78	63
7 Feb. 1958	77	71
25 Mar. 1958	81	73
15 May 1958	78	67
17 June 1958	77	152
25 Sept. 1957	89	124
5 Nov. 1957	91	61
19 Dec. 1957	88	99

A final observation from Fig. 3 is the higher average values at Minneapolis and San Angelo than at the Canal Zone at almost all altitudes and times covered by this study. This is particularly striking in view of the large injections by the United States in the latitude closest to the Canal Zone. The significance of this observation is discussed below.

Stratospheric inventories (*7*). The data permit estimates of the stratospheric content of radiocarbon produced by nuclear weapons. In computing these inventories, charts such as Fig. 4 were prepared from concentrations averaged over 6-month intervals centered on 1 January and 1 July. The stratospheric content was found by graphical integration and is given in Table 5. It is felt that the errors in these numbers are no greater than ± 30 percent.

Table 5. Stratospheric content of artificial radiocarbon.

Date	Radiocarbon content (10^{27} C^{14} atoms)
1 July 1955	8.6
1 January 1956	8.0
1 July 1956	5.6
1 January 1957	6.6
1 July 1957	7.4
1 January 1958	6.1
1 July 1958	8.4

Münnich and Vogel (*8*) have estimated that there were about 4.4×10^{27} artificially produced carbon-14 atoms in the troposphere, the biosphere, and the surface layers of the oceans in mid-1957. Broecker and Walton (*6*) estimate the number to be 4.8×10^{27} carbon-14 atoms in March 1958. Correcting these numbers to 1 January 1957, using an average of the growth curves for the two hemispheres given by Broecker and Walton, gives about 3.2 and 2.4×10^{27} atoms, respectively. The same figures, corrected to 1 July 1958, are 6.8 and 5.2×10^{27} carbon-14 atoms. Since Broecker and Walton have more reliable estimates of the oceanic content, numbers near the lower part of the range are probably to be preferred. Thus, the total content in the stratosphere, troposphere, biosphere, and oceans was about 9.0×10^{27} carbon-14 atoms on 1 January 1957 and about 14×10^{27} carbon-14 atoms on 1 July 1958. [Libby (*9*) suggests that some carbon-14 may fall back to the ground or sea as calcium carbonate. This local fallout of carbon-14 is not included in the above inventories.] The 1 January 1957 inventory agrees with Libby's estimate (*9*) of about 10×10^{27} atoms produced by nuclear devices.

The 9×10^{27} carbon-14 atoms present on 1 January 1957 were produced by a total of 89 megatons of nuclear explosives (*10*). During the period 1957–58, an additional 85 megatons (*10*) were detonated. However, a larger fraction (85 percent versus 30 percent) of the tests were conducted in the air in the latter years and, therefore, probably introduced more carbon-14 per megaton into the atmosphere. For this reason, the increment in 1957–58 is estimated as roughly 16×10^{27} carbon-14 atoms. The sum, 25×10^{27} carbon-14 atoms, is the present estimate based upon available data of the total number of artificial radiocarbon atoms released into the atmosphere up to the suspension of atomic tests on 31 October 1958.

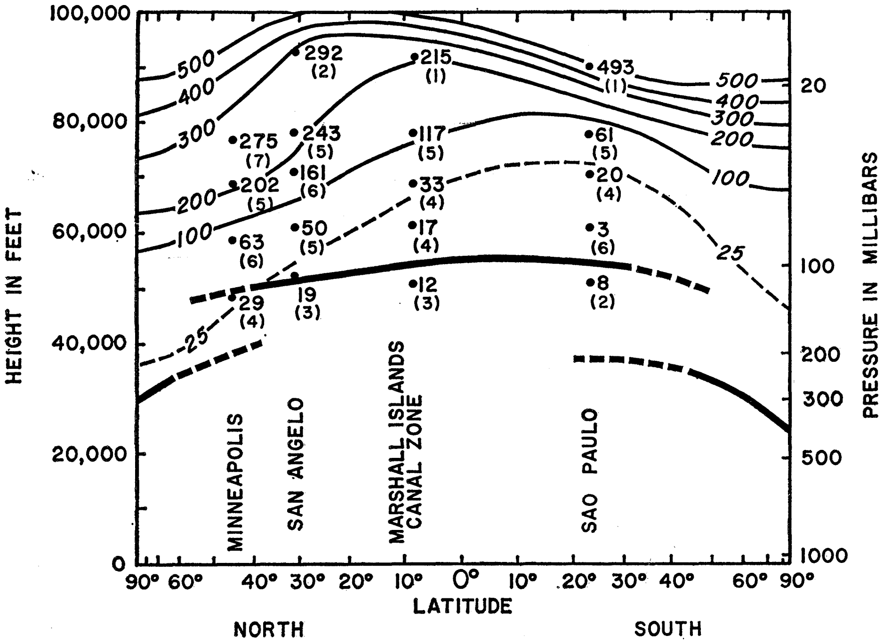


Fig. 4. Altitude-latitude cross section of the atmosphere showing the mean excess radiocarbon distribution as of 1 July 1955. Concentrations in 10^5 atoms per gram of air are indicated near points identifying the altitude. Numbers in parentheses show the number of samples from which the mean concentration was computed. Thin lines are isolines of carbon-14; heavy lines indicate the position of the tropopause.

The fate of artificially produced carbon-14 is of importance in considering the biological hazards from atomic tests. According to Broecker and Walton (*6*), the carbon-14 content of the Northern Hemisphere troposphere had increased by 17 percent by mid-1958. If the 25×10^{27} carbon-14 atoms that have been produced up to the present were mixed throughout the world atmosphere only, the total tropospheric concentration of carbon-14 would be 1¾ times the natural concentration. However, it is not likely that this total will exceed 1½ times the natural concentration because of uptake by the oceans. Continued mixing (*11*) with the surface layers of the oceans will reduce the carbon-14 concentration in the atmosphere from the peak value to a world-wide average of about 1⅓ times the natural value. The bulk of the exchangeable carbon reservoir lies in the deep oceans, where there is over 50 times more exchangeable carbon than in the atmosphere. The mixing between the surface layers of the ocean and the deep ocean probably takes place over a period of several tens to several hundreds of years. Thus, it is estimated that mixing of the artificial radiocarbon with the entire global reservoir over many tens or hundreds of years will reduce the radiocarbon increment from nuclear tests carried out to the present date to below 1 percent of the natural background. The biological significance of increases in the carbon-14 content of the atmosphere has been discussed by Leipunsky (*12*), Pauling (*13*) and Totter *et al.* (*14*).

Some meteorological comments. The carbon-14 measurements afford a remarkable opportunity to study stratospheric motions. The radiocarbon observations pre-date the stratospheric measurements of particulate fission products (*15*). In addition, carbon-14, in the form of gaseous carbon dioxide, is a better tracer than the particulate fission products because there is little likelihood of separation from its associated air mass by gravitational settling or other mechanisms. Only a few of the meteorological interpretations of the data are presented below.

Figure 4 depicts the distribution of artificial radiocarbon as a function of altitude and latitude as measured during the period from April through September 1955. The analysis for the atmosphere north of 45°N and south of 23°S is subjective. The period April–September 1955 was chosen for discussion because all significant stratospheric injections prior to this date occurred at 11°N. For all practical purposes they originated from only two test series, that in the fall of 1952 and that in the spring of 1954. The cross section for July 1955 (Fig. 4) is thus based on samples collected more than 1 year after the second and larger injection.

Two of the significant features of Fig. 4 are (i) the greater concentrations at the same altitudes at the San Angelo and Minneapolis locations than at the Canal Zone and (ii) the higher concentrations at San Angelo, Minneapolis, and the Canal Zone than at São Paulo, except for the 90,000-foot levels, for which there are very few measurements. There appear to be two possible explanations of the distribution shown in Fig. 4 from a source which originated closest in latitude to the Canal Zone.

The first explanation assumes that the transfer of matter in the stratosphere takes place only by diffusive mixing along the gradient of concentration. Since the Canal Zone is close to the latitude of the source, the highest concentration on such a theory must always be present at this observing point relative to the other three stations. But, since the data indicate otherwise—namely, that the Canal Zone has lower concentrations level for level than San Angelo and Minneapolis—it is necessary to locate the highest concentrations above the highest altitude of the measurements, that is, above 90,000 feet. From these hypothetical high concentrations over the Canal Zone the radiocarbon mixes laterally either horizontally or along surfaces which slope gradually downward toward the poles. If the former, then vertical mixing is assumed to become more intense at the latitudes of the other stations. To account for lower concentrations at São Paulo than at San Angelo or Minneapolis north-south mixing across the equator must be assumed to be slow. The diffusion explanation may be reasonable since several powerful nuclear explosions were known to have risen beyond 90,000 feet and, in general, higher concentrations are observed at increasing altitudes at all locations. It may be objected to on the grounds that the greatest lateral extent of the visible clouds, even for those which rose very high, may have been below 90,000 feet although the gases could be concentrated at the top of the mushroom. Further, high values at Minneapolis in December 1954 appear at 80,000 rather than at 90,000 feet. Finally, it is always unsatisfactory to explain something by unknown data just beyond the reach of observations.

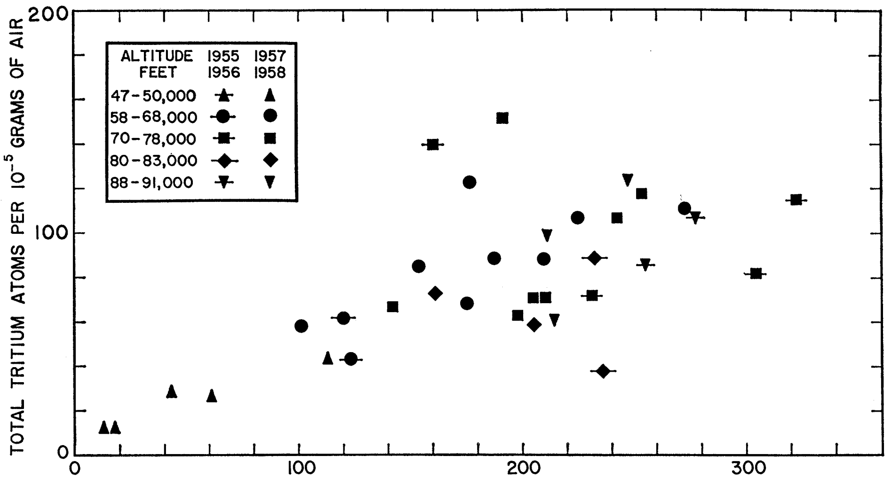


Fig. 5. Relationship between total tritium and excess carbon-14 in the stratosphere.

The alternate explanation follows the Brewer-Dobson (*16*) model, as interpreted by Stewart (*17*) and Machta (*18*). Air is presumed to rise very slowly into the equatorial stratosphere from the troposphere, then drift upward and poleward. Each hemispheric branch is approximately symmetrical with respect to the geographical equator. Later, air descends into the lower stratosphere in the temperate or polar zone, or both. In this picture, the radiocarbon injections can take place below 90,000 feet. During the year following the 1954 tests, the northward drift bodily carried peak concentrations poleward in the Northern Hemisphere, leaving much smaller concentrations at the Canal Zone. Turbulent mixing is present in the stratosphere, but the transport is dominated by the circulation. Horizontal mixing across the equator brought a small amount of radiocarbon into the Southern Hemisphere for transport to the upper levels over São Paulo by the Southern Hemisphere circulation. This explanation is supported by the continued decrease of concentrations at the higher stratospheric altitudes over the Canal Zone until the summer of 1956, when new equatorial injections took place.

Another meteorological item of current interest is the residence time of air masses in the stratosphere before mixing with the troposphere. It has often been assumed that first-order kinetics can be applied to this exchange mechanism—that is, that the amount of a tracer removed from the stratosphere is always a fixed proportion of the stratospheric inventory. It is implied in first-order kinetics that the reservoir (in this case the stratosphere) is thoroughly mixed on a time scale shorter than that of the removal process. The nonhomogeneous stratospheric distribution more than a year after the injection (Figs. 3 and 4) argues strongly against a hypothesis of fast mixing. However, there is interest in knowing what fraction of the carbon-14 came out of the stratosphere during the early years. It should be noted that this fraction need not be the same for later years or for the first few years following an injection into the stratosphere at other latitudes and altitudes. Most of the carbon-14 prior to 1957 was added at 11°N.

Calculation of the stratospheric removal rate from differences in the total stratospheric contents at successive times is unreliable because of the inaccuracies in the inventories and the uncertain magnitudes of the injections of carbon-14 occurring during the periods considered. Instead, two other methods will be used.

On 1 January 1957 the total artificial radiocarbon inventory was about 9.0×10^{27} carbon-14 atoms, of which 6.6×10^{27} were still in the stratosphere. The fraction of the total remaining in the stratosphere can be explained by using known times and relative magnitudes of stratospheric injections (taking into account local fallout) and an exponential removal rate corresponding to about 17 percent per year. The mean stratospheric residence time on this basis is about 6 years (a half time for removal of 4 years).

A mean residence time in the stratosphere can also be calculated from the average stratospheric carbon-14 inventory and Broecker and Walton's growth curve for the troposphere (*6*). The stratospheric inventory is constant, almost within experimental error, at 7.2×10^{27} carbon-14 atoms during the period 1 July 1955 to 1 July 1958 (see Table 5). The average rate of increase of carbon-14 in the troposphere during this period was 1.2×10^{27} atoms per year (*6*). A very rough estimate of the average rate of uptake of the artificial carbon-14 by the oceans during the same period is 0.3×10^{27} atoms per year. This is based on a mean tropospheric content of 2.1×10^{27} atoms and a removal rate of one-seventh per year (*11*). Thus, about 1.5×10^{27} artificially produced carbon-14 atoms were removed from the stratosphere each year, corresponding to a mean residence time of slightly less than 5 years.

These residence times, about 5 years, are shorter than Libby's earlier estimate (*19*) but are close to the recent computations reported by Shelton (*20*), Libby (*21*), and Machta and List (*22*).

The uncertainty in the residence time as calculated in this paper is directly proportional to the uncertainty of the stratospheric inventory and depends to a lesser extent on the rate of uptake by the oceans. Further, it is assumed that all of the injections enter the stratosphere. A small error is made because some radiocarbon from weapons tests is inserted directly into the troposphere.

Comparison of tritium and carbon-14 measurements. Figure 5 is a plot of the total tritium as a function of the excess carbon-14 concentration at Minneapolis. It is evident that there is a positive correlation between the tritium concentration and the excess radiocarbon. On the assumption of a constant tritium to carbon-14 ratio from contributing nuclear explosions, the data can be used to calculate the natural tritium content of the stratosphere. The tritium concentration corresponding to zero excess carbon-14 is the natural tritium background value. Although only the 1957–58 points near the 50,000-foot level yield a statistically significant background value, 7×10^5 ($\sigma = 3.5 \times 10^5$) tritium atoms per gram of air, the entire body of the 1957–58 data gives a value consistent with this. If this concentration exists throughout the stratosphere, then there are approximately 6×10^{26} naturally produced tritium atoms in the entire stratosphere. Assuming that the mean stratospheric residence time is 5 years for natural tritium and that two-thirds of the natural production takes place in the stratosphere (as would be the case for cosmic ray production), 6×10^{26} tritium atoms lead to an average world-wide natural production rate of 1 tritium atom per square centimeter per second, in good agreement with recent estimates summarized by Begemann (*23*).

The tritium concentrations expressed as atoms per gram of air can be converted to tritium atoms per 10^{18} hydrogen atoms by estimating the stratospheric moisture content. The frost-point in the stratosphere is believed to lie in the range of −65° to −80°C. Thus, the 50,000-foot background tritium concentration of 7×10^5 tritium atoms per gram of air would correspond to between 4×10^5 and 4×10^6 tritium atoms per 10^{18} hydrogen atoms. For comparison, rains in Chicago prior to 1954 contained 3 to 34 tritium atoms per 10^{18} hydrogen atoms (*24*).

The good correlation between the excess carbon-14 and tritium permits an estimate of the number of tritium atoms associated with each carbon-14 atom produced by weapon tests. Assuming that the ratio of tritium to carbon-14 atoms, 0.4, based on all of the 1957–58 data was the same at other locations than Minneapolis, the stratospheric inventory of excess tritium on 1 January 1958 was 2.4×10^{27} atoms, or about 4 times the natural content of tritium in the stratosphere. The rapid removal of tritium from the troposphere by precipitation puts most of nonstratospheric tritium in the oceans. Because of inadequate data on the tritium content of ocean waters, no attempt has been made to obtain a complete inventory of weapon-produced tritium.

References and Notes

1\. A report on the technical details, together with a further evaluation of the experimental uncertainties, data from later sampling, and a more extensive interpretation, is in preparation.

2\. The collection equipment was developed by General Mills, Inc. Samples were collected by General Mills during the early period of work. Later, sampling was carried out by the Headquarters Command, U.S. Air Force. We gratefully acknowledge the assistance of the following personnel of the mechanical division of General Mills, Inc.: J. W. Swisher, D. Crowe, W. Borgeson, R. Dungan, J. Smith, R. Mautner, D. Dietsche, J. Gravelle, H. Demerest, and R. Stuart.

3\. C. D. Keeling, *Geochim. et Cosmochim. Acta* 13, 322 (1958); *Trans. Am. Geophys. Union* 40, 57 (1959).

4\. The radiocarbon determinations were carried out at the Argonne National Laboratory, chemistry division. The assistance of A. S. Quattrochi and S. J. Rymas, who conducted the preparation and measurements of most of the samples, is gratefully acknowledged.

5\. K. Telegadas, in "Report of the Congress of the United States, Joint Committee on Atomic Energy, Special Subcommittee on Radiation, Fallout from Nuclear Weapons Tests" (Government Printing Office, Washington, D.C., in press).

6\. W. S. Broecker and A. Walton, *Science* **130**, 309 (1959).

7\. The contribution of Robert J. List of the U.S. Weather Bureau to the meteorological analysis of the data is acknowledged.

8\. K. O. Münnich and J. C. Vogel, *Naturwissenschaften* 14, 327 (1958).

9\. W. F. Libby, *Bull. Swiss Acad. Sci. No. 14* (1958), p. 309.

10\. *U.S. Atomic Energy Comm. Health and Safety Lab. Rept. No. 65* (1959), p. 193.

11\. H. Craig, *Tellus* 9, 1 (1957).

12\. O. I. Leipunsky, *Atomnaya Energ.* 3, 530 (1957).

13\. L. Pauling, *Science* 128, 1183 (1958).

→ J. L. Totter, M. R. Zelle, H. Hollister, *ibid.* 128, 1490 (1958).

15\. *U.S. Atomic Energy Comm. Health and Safety Lab. Rept. No. 42* (1958), p. 38.

→ A. W. Brewer, *Quart. J. Roy. Meteorol. Soc.* 75, 351 (1949); G. M. B. Dobson, *Proc. Roy. Soc.* (*London*) 236A, 187 (1956).

17\. N. G. Stewart, R. N. Crooks, E. M. Fisher, *Atomic Energy Research Establ.* (*G. Brit.*) *Rept. No. HP/R1701* (June 1955).

18\. L. Machta, "Transport in the stratosphere and through the tropopause," in *Advances in Geophysics* (Academic Press, New York, 1959), vol. 6, p. 273.

→ W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **42**, 365 (1956).

20\. F. Shelton, in "Report of the Congress of the United States, Joint Committee on Atomic Energy, Special Subcommittee on Radiation, Fallout from Nuclear Weapons Tests" (Government Printing Office, Washington, D.C., in press).

21\. W. F. Libby, talk given at the University of Washington, Seattle (March 1959).

22\. L. Machta and R. J. List, in *U.S. Atomic Energy Comm. Health and Safety Lab. Rept. No. 42* (1958), p. 282.

23\. F. Begemann, *Z. Naturforsch.* 14a, 334 (1959).

24\. H. von Buttlar and W. F. Libby, *J. Inorg. & Nuclear Chem.* 1, 75 (1955).

* * *