

Long-Term Environmental Monitoring of ^{14}C Levels in the Ottawa Region

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Abstract



Ambient levels of ^{14}C have been monitored near Ottawa, Canada by the Radiocarbon Dating Laboratory, Geological Survey of Canada, since 1961. The 32 years of data on maple leaves documents the rapid increase of bomb-generated ^{14}C in the mid-1960s and its exponential decline to the present. This bio-integrated data set indicates that the half-life of bomb-generated ^{14}C is 12 years as a result of dilution with prebomb ^{14}C from the decay of organic matter and exchange with the oceanic reservoir, and, of course, a dilution from fossil fuel combustion. Atmospheric ^{14}C activity will approach 10% of prebomb levels by the turn of the century, but it will be another century before the prebomb levels of atmospheric ^{14}C are approached.

Introduction

Carbon has one naturally occurring radioisotope, ^{14}C , which is continually produced in the upper atmosphere (stratosphere) by cosmic rays. This radioactive carbon is rapidly oxidized to carbon dioxide ($^{14}\text{CO}_2$) and mixes with the non-radioactive carbon dioxide in the lower atmosphere (troposphere). The breakdown of this naturally occurring ^{14}C allows the dating of carbonaceous materials. All radiocarbon dates are related to the 1950 (pre-bomb) activity of ^{14}C (Modern Carbon). Unfortunately bomb-generated ^{14}C undergoes the same fate as naturally occurring ^{14}C (Fig. 1) and is integrated into the global carbon cycle.

In 1959, W. Dyck established a radiocarbon dating laboratory at the Geological Survey of Canada. By the early 1960s, age determinations on materials as old as 40 000 years were routinely undertaken in support of research into the glacial history, rate of sea level change, and the identification of earthquake and landslide frequencies in Canada, as well as providing chronologies for events and rates of transition and development related to global change and human history. In 1961 a small project to monitor the variation in bomb-produced ^{14}C in the biosphere was initiated by W. Dyck to determine what effect bomb- ^{14}C would have on future radiocarbon age-determination programs. The sampling site selected is remote from both industry and municipal dwellings, and because it is in a park it had a good chance of maintaining its integrity over time. It is located about 20 km northwest of Ottawa on an escarpment about 210 m above the Ottawa Valley. The site is within the Gatineau Park near the Champlain Lookout (45°30' N, 75°54' W). From 1961 to 1964, material was collected only in mid-summer and initially was restricted to the sampling of maple leaves and grass, but the program was expanded in 1965 by J.A. Lowdon to include a variety of plants, and samples were taken from a number of regions in Canada (Lowdon and Dyck, 1974). In the early phase of this monitoring program samples from different regions of Canada, both east and west, and north and south did not show any appreciable difference in ^{14}C activity (Lowdon and Dyck, op. cit.), so the program was restricted to the Gatineau Park site. In 1965, monthly sampling of maple leaves during the growing season was initiated and has continued to the present.

The measurement techniques of the GSC Radiocarbon Dating Laboratory are presented in detail in Lowdon (1985). The 'high-precision' analytical technique used to measure the ^{14}C content of Gatineau Park maple leaves has remained uniform over the last 32 years and the data are therefore internally consistent. In 1990, the GSC Laboratory participated in a quality control program for radiocarbon laboratories developed by the International Atomic Energy Agency (IAEA, Geneva). One of the 5 samples measured was modern wood cellulose; the 'consensus' and GSC values for ^{14}C and the $\delta^{13}\text{C}$ are noted below:

<u>IAEA Material</u>		<u>Consensus Value</u>	<u>GSC Value</u>
modern	pMC	129.41±0.06%	130.20±0.48%
wood cellulose	$\delta^{13}\text{C}$	-25.4±0.2‰	-24.9±0.3‰

These results indicate that the GSC Laboratory is capable of accurately measuring the ^{14}C activity of modern materials within the precision of our gas proportion counting technique ($\pm 0.5\%$).

Results

Growing season averages of ^{14}C activity in maple leaves as per cent Modern Carbon (pMC) (**Fig. 2**) illustrates the rapid increase in bomb-generated ^{14}C in the early to mid-1960s. Since then there has been a steady decrease in ^{14}C activity as a result of dilution with pre-bomb ^{14}C from the decay of organic matter and exchange with the oceanic reservoir, and, of course, a dilution resulting from fossil fuel combustion (**Fig. 1**).

The levels documented by the GSC Laboratory are consistent with other observations worldwide and show a gradual decline from peak $\delta^{14}\text{C}$ values in 1964 ($\pm 850\%$) to present day levels of $\pm 150\%$. The decline from the peak levels observed in 1964 (**Fig. 3**) indicates a half-life of bomb-generated ^{14}C of about 12 years, with the atmospheric activity approaching 10% of pre-bomb levels by the turn of this century. It will be another century before we will approach the pre-bomb levels of ^{14}C . Although the sampling of the biosphere (e.g. maple leaves) generates intergrated data, unique events are still detectable. For example, the atmospheric bomb test in China in 1980 produced considerable ^{14}C , and is evident in both **Fig. 2** and **Fig. 3**. These data illustrate that there is a lag of a year or two between a bomb test and its appearance in the biosphere, presumably as a result of non-uniform stratosphere - troposphere coupling. **Fig. 2** also shows slightly enhanced values in the early 1970s related to testing in the late 1960s and early 1970s. The Chernobyl event did not produce sufficient ^{14}C to appear in our data, although atmospheric monitoring programs were able to detect it.

"By international agreement, conventional ^{14}C ages are calculated using the Libby half-life of 5570 ± 30 years and 0.95 times the activity of the NBS oxalic acid as the modern standard, normalized to $\delta^{13}\text{C} = -19\%$. Results which reflect the post AD 1955 rise in atmospheric ^{14}C (due to nuclear weapons testing) are reported as ratios of the modern standard value." (Kra, 1986). The GSC data are presented in their entirety as **Table 1**, which includes the count (cpm) data for the Oxalic Acid Standard and the monthly sample of maple leaves, the measured values of $\delta^{13}\text{C}$ (relative to the PDB standard), and the calculated values for $\delta^{14}\text{C}$, $\Delta^{14}\text{C}$, and pMC (per cent Modern Carbon). The calculations used for these parameters are noted below (cf. Stuiver and Polach, 1977 for additional details):

$$\delta^{13}\text{C} = \frac{(^{13}\text{C}/^{12}\text{C} \text{ sample}) - (^{13}\text{C}/^{12}\text{C} \text{ standard})}{(^{13}\text{C}/^{12}\text{C} \text{ standard})} \times 1000 \text{ per mil}$$

$$\delta^{14}\text{C} = \frac{(^{14}\text{C}/^{12}\text{C} \text{ sample}) - (^{14}\text{C}/^{12}\text{C} \text{ standard})}{(^{14}\text{C}/^{12}\text{C} \text{ standard})} \times 1000 \text{ per mil}$$

$$^{14}\text{C} = \Delta^{14}\text{C} - 2(\delta^{13}\text{C} + 25)(1 + \Delta^{14}\text{C}/1000) \text{ per mil}$$

$$\text{pMC} = ((\Delta^{14}\text{C}/1000) + 1) \times 100 \text{ per cent}$$

References

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Table 1. ^{14}C data on maple leaves from the Gatineau Park, Québec.

Year.Month	Sample cpm	Standard cpm	$\delta^{13}\text{C}$ ‰	$\delta^{14}\text{C}$ ‰	$\Delta^{14}\text{C}$ ‰	pMC ‰
61.07	23.825	19.700	-26.9	209	214	121.4
62.07	26.830	20.400	-27.0	315	320	132.0
63.07	31.940	19.700	-27.3	621	929	162.9
64.07	52.101	28.429	-27.5	833	842	184.2
65.05	52.805	28.790	-24.5	834	832	183.2
65.07	51.789	28.940	-27.3	790	798	179.8
65.08	51.665	29.063	-26.7	778	784	178.4
65.09	51.550	29.063	-25.4	774	775	177.5
66.05	50.817	28.938	-24.2	756	753	175.3
66.07	49.455	28.888	-28.1	712	723	172.3
66.08	49.736	28.776	-26.7	728	734	173.4
66.09	49.775	28.944	-27.1	720	727	172.7
67.05	49.060	29.103	-27.0	686	692	169.2
67.06	48.348	29.250	-27.0	653	660	166.0
67.07	48.348	29.250	-26.7	653	659	165.9
67.08	47.828	29.103	-27.0	643	650	165.0
67.09	47.642	28.953	-28.1	645	656	165.6
68.05	47.687	28.905	-25.2	650	650	165.0
68.06	46.569	28.905	-28.5	611	622	162.2
68.07	46.095	28.694	-24.8	606	606	160.6
68.08	46.378	28.694	-27.5	616	624	162.4
68.09	46.356	28.694	-26.3	616	620	162.0
68.10	46.719	28.898	-27.4	617	624	162.4
69.05	31.912	19.744	-26.0	616	620	162.0
69.06	45.417	28.488	-24.6	594	593	159.3
69.07	45.268	28.488	-25.2	589	590	159.0
69.08	45.235	28.412	-26.9	592	598	159.8
69.09	45.049	28.606	-26.8	575	580	158.0
70.05	45.104	28.823	-23.5	565	560	156.0
70.06	44.663	28.823	-26.4	550	554	155.4
70.07	44.293	28.889	-28.3	533	543	154.3
70.08	44.354	28.823	-28.3	539	549	154.9
70.09	44.184	28.708	-26.5	539	544	154.4
71.05	44.099	28.643	-25.7	540	542	154.2
71.06	44.271	28.643	-27.3	546	553	155.3
71.07	43.612	28.643	-27.2	523	529	152.9
71.08	44.086	28.556	-26.4	544	548	154.8
71.09	43.560	28.556	-26.1	525	529	152.9
71.10	43.588	28.358	-27.0	537	543	154.3
72.05	42.924	28.098	-24.0	528	525	152.5
72.07	42.544	28.551	-26.9	490	496	149.6
72.08	42.515	28.382	-27.0	498	504	150.4
72.09	42.568	28.376	-26.5	500	505	150.5
72.10	42.066	28.393	-27.9	482	490	149.0

Table 1 cont. ^{14}C data on maple leaves from the Gatineau Park, Québec.

Year.Month	Sample cpm	Standard cpm	$\delta^{13}\text{C}$ ‰	$\delta^{14}\text{C}$ ‰	$\Delta^{14}\text{C}$ ‰	pMC ‰
73.05	42.038	28.264	-27.6	487	495	149.5
73.06	41.841	28.448	-27.9	471	479	147.9
73.07	41.100	28.630	-28.7	436	446	144.6
73.08	41.563	28.501	-28.3	458	468	146.8
73.09	40.874	28.630	-28.3	428	437	143.7
74.05	41.231	28.537	-28.1	445	454	145.4
74.06	41.152	28.611	-28.8	438	449	144.9
74.07	40.965	28.611	-29.2	432	444	144.4
74.08	40.986	28.434	-28.4	441	451	145.1
74.09	40.454	28.382	-28.7	425	436	143.6
74.10	40.643	28.434	-28.4	429	439	143.9
75.05	40.381	28.241	-26.6	430	434	143.4
75.06	39.955	28.241	-28.3	415	424	142.4
75.07	40.441	28.376	-28.1	425	434	143.4
75.08	40.168	28.063	-28.3	431	441	144.1
75.09	39.426	28.063	-28.3	405	414	141.4
76.05	39.165	28.016	-26.8	398	403	140.3
76.06	39.255	28.102	-27.5	397	404	140.4
76.07	39.131	28.022	-28.4	396	406	140.6
76.08	38.919	28.022	-28.3	389	398	139.8
76.09	38.941	28.187	-28.3	382	391	139.1
76.10	38.803	28.208	-27.5	376	382	138.2
77.05	38.494	28.210	-26.4	365	368	136.8
77.06	38.188	28.210	-28.0	354	362	136.2
77.07	38.253	28.136	-28.6	360	369	136.9
77.08	38.069	28.197	-29.6	350	363	136.3
77.09	38.077	28.412	-29.1	340	351	135.1
78.05	37.608	27.783	-27.6	354	361	136.1
78.06	37.402	27.968	-28.1	337	346	134.6
78.07	37.400	27.972	-30.5	337	352	135.2
78.08	37.616	28.003	-28.7	343	353	135.3
78.09	37.439	27.953	-29.9	339	352	135.2
78.10	37.275	27.918	-29.8	335	348	134.8
79.05	37.254	27.965	-26.9	332	337	133.7
79.06	36.576	27.965	-28.2	308	316	131.6
79.07	36.645	27.903	-30.0	313	326	132.6
79.08	36.462	28.023	-30.2	301	315	131.5
79.09	36.371	27.805	-31.2	308	324	132.4
80.05	36.626	27.805	-27.7	317	324	132.4
80.06	36.239	28.095	-28.0	290	298	129.8
80.07	36.593	28.152	-28.6	300	309	130.9
80.08	36.190	27.857	-29.2	299	310	131.0
80.09	36.236	27.848	-28.8	301	311	131.1
81.05	36.436	28.063	-29.4	298	310	131.0
81.06	36.384	28.014	-29.3	299	310	131.0
81.07	36.285	27.802	-29.0	305	316	131.6
81.08	37.629	28.013	-29.1	343	354	135.4
81.09	36.336	28.013	-30.3	297	311	131.1
81.10	36.235	27.949	-29.9	296	309	130.9

Table 1 cont. ^{14}C data on maple leaves from the Gatineau Park, Québec.

Year.Month	Sample cpm	Standard cpm	$\delta^{13}\text{C}$ ‰	$\delta^{14}\text{C}$ ‰	$\Delta^{14}\text{C}$ ‰	pMC ‰
82.05	36.230	27.731	-28.2	306	315	131.5
82.06	36.484	27.837	-29.4	311	322	132.2
82.07	36.838	27.898	-29.4	320	332	133.2
82.08	36.395	27.686	-26.6	315	319	131.9
82.09	36.241	27.759	-30.0	306	319	131.9
82.10	36.565	27.811	-28.2	315	323	132.3
83.05	36.676	27.917	-28.6	314	323	132.3
83.06	34.892	27.917	-29.7	250	262	126.2
83.07	35.195	27.917	-29.0	261	271	127.1
83.08	35.181	27.767	-29.6	267	279	127.9
83.09	35.279	27.720	-29.4	273	284	128.4
83.10	35.334	27.720	-29.7	275	287	128.7
84.05	34.690	27.828	-27.8	247	254	125.4
84.06	34.548	27.828	-27.3	241	247	124.7
84.07	34.565	27.828	-29.4	242	253	125.3
84.08	34.253	27.736	-29.2	235	245	124.5
84.09	34.419	27.764	-29.5	240	251	125.1
84.10	34.368	27.764	-29.3	238	249	124.9
85.05	34.276	28.554	-28.2	200	208	120.8
85.06	33.963	27.970	-30.5	214	228	122.8
85.07	33.864	27.764	-30.1	220	232	123.2
85.08	33.484	27.764	-30.8	206	220	122.0
85.09	33.638	27.433	-30.1	226	239	123.9
85.10	33.426	27.433	-30.2	218	231	123.1
86.05	34.489	28.202	-27.2	223	228	122.8
86.06	33.906	28.301	-30.1	198	210	121.0
86.07	33.939	28.020	-30.2	211	224	122.4
86.08	33.749	27.986	-30.3	206	219	121.9
86.09	34.072	28.099	-30.3	213	225	122.5
87.05	33.908	28.320	-30.5	197	211	121.1
87.06	33.901	28.320	-31.3	197	212	121.2
87.07	33.759	28.320	-31.0	192	206	120.6
87.08	33.656	28.320	-31.0	188	203	120.3
87.09	33.665	28.320	-30.8	189	203	120.3
87.10	33.641	28.206	-31.9	193	209	120.9
88.05	33.536	28.206	-27.9	189	196	119.6
88.06	33.533	28.206	-31.3	189	204	120.4
88.07	33.205	28.206	-28.1	177	184	118.4
88.08	21.274	18.080	-29.6	177	187	118.7
88.09	33.449	28.148	-29.6	188	199	119.9

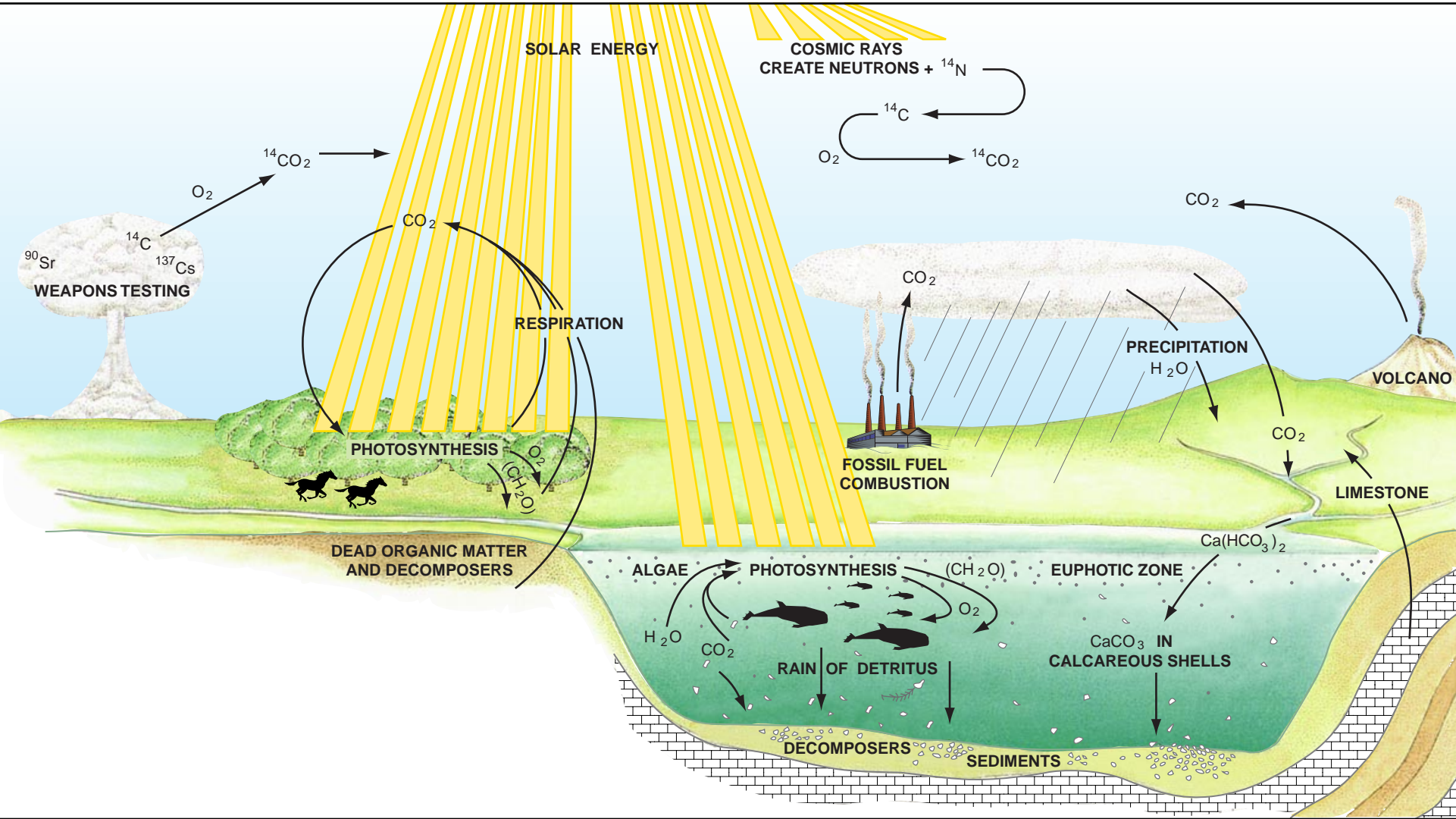


Fig. 1. Carbon pathways in the biosphere/geosphere

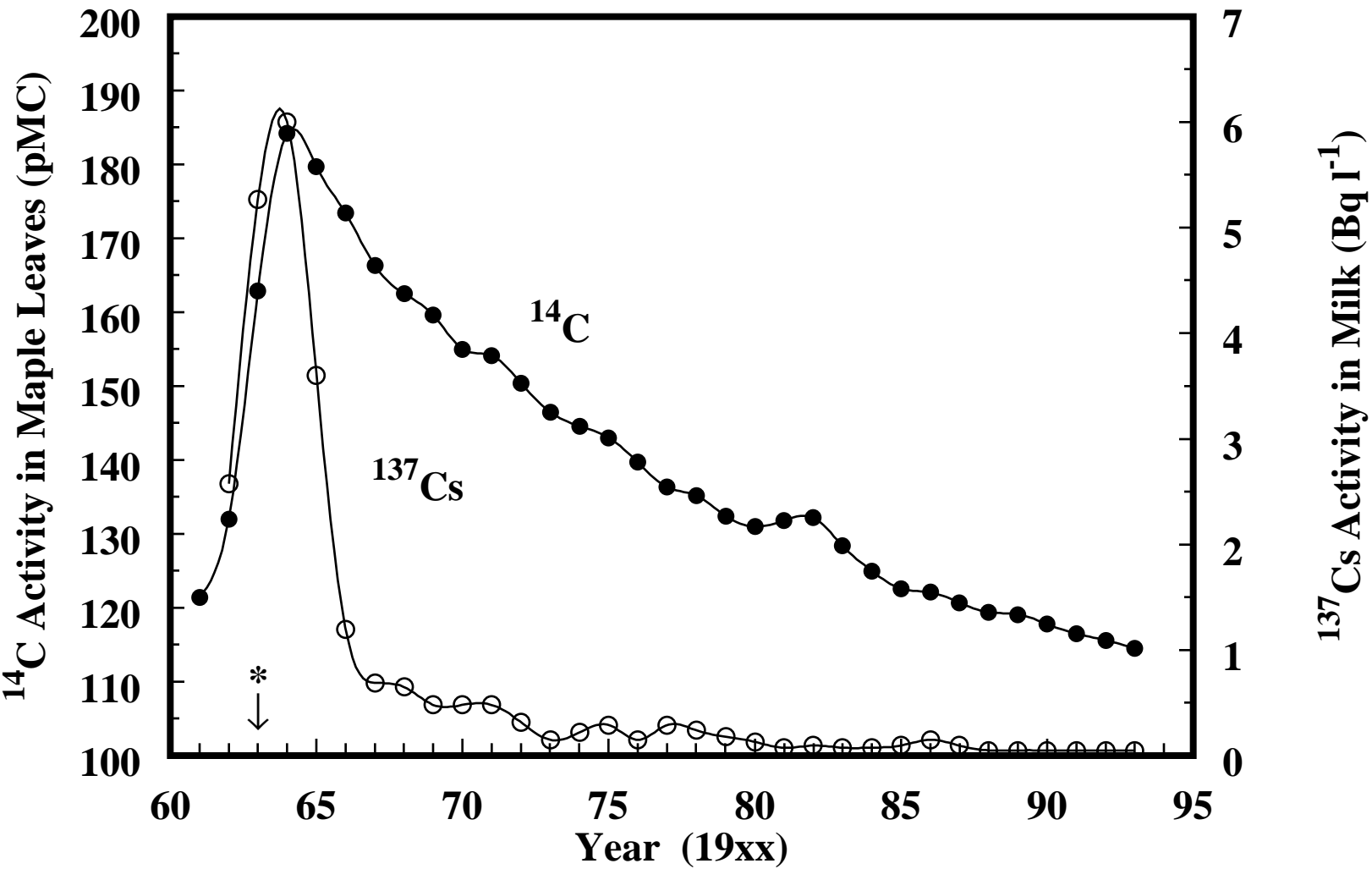


Figure 2. ^{14}C in Maple Leaves and ^{137}Cs in Milk in the Ottawa Region from 1961-1993.

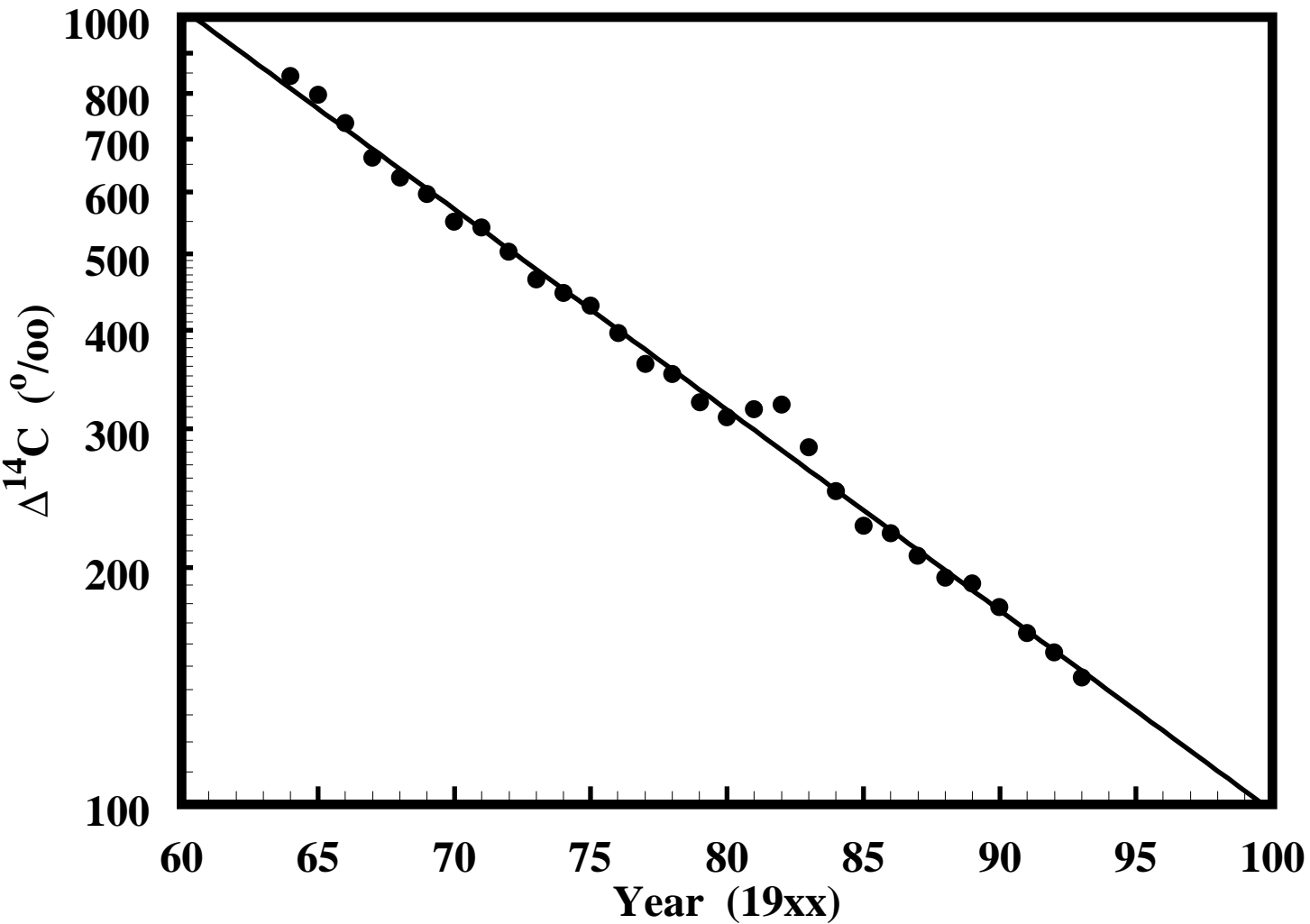


Figure 3. Decline of bomb ^{14}C activity in maple leaves, 1964-1993.