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VARIATIONS IN THE ATMOSPHERIC RADIOCARBON CONCENTRATION OVER THE PAST 1300 YEARS

E. H. WILLIS (Cambridge), H. TAUBER (Copenhagen) and

K. O. MÜNNICH (Heidelberg)

Considerable attention has been focussed in recent years upon the validity of the radiocarbon dating method by papers whose authors have considered that one or other of the fundamental principles might either be in error or require serious modification (Crowe, 1958; Milojčić, 1957; Elsasser, Ney, and Winkler, 1957; Daniel, 1959). It has even been suggested that errors as great as 800 years might arise between datings on the same sample made in different laboratories (Crowe, 1958). In the light of such criticism, it is clearly of the utmost importance to investigate, and, if possible, justify the basic assumptions on which the validity of the method rests.

The assumption which might be most open to doubt is that the specific activity of living organic material has been constant over a very long period. This supposition implies that the contemporary assay is universal and that the carbon-exchange reservoir should be in isotopic equilibrium. The other two assumptions, namely, that the biological materials assayed should have retained their true original composition, and that the half-life of radiocarbon shall be accurately determined, have already been discussed adequately (Tauber, 1958; Barker, 1958; Broecker and Olson, 1959). De Vries (1958) had previously made some precise measurements upon tree rings of known age, and had concluded from this preliminary investigation that the contemporary assay, and thus the atmospheric radiocarbon concentration, had varied in the order of 1% during the past 400 years. He was able to correlate these variations with advance and retreat of glaciers, thus tentatively linking up the variation in atmospheric radiocarbon concentration with climatic phenomena. The evidence also suggested that the contemporary assay had varied not only with time but with position on the Earth's surface.

The radiocarbon dating laboratories at Cambridge, Copenhagen, and Heidelberg have jointly undertaken a research project designed both to extend present knowledge of variations in atmospheric radiocarbon, demonstrated by de Vries, and to serve as an exercise in inter-laboratory cross-checking. The subject of the experiment was a section of a sequoia tree (*Sequoia gigantea*) contained in the museum collection of the Botany School, University of Cambridge. The section had been ring-counted soon after its felling, and was fully documented. Samples were taken at 50-year ring intervals through the section by a half-inch drill, and each sample was subdivided into three parts,

TABLE 1

The percentage variation of the activity of samples from a section of a giant sequoia tree, corrected for age, compared with the activity of the sample from the 1859 tree ring. (The standard deviation on the measurement is ± 0.6%.)

Series Sample Number	Year of Growth A.D.	Number of Rings Cut by Drill	Percentage Variation from the Activity of the 1859 Tree Ring					
			Cambridge		Copenhagen		Heidelberg	
			No.	Var.	No.	Var.	No.	Var.
1	659	7	Q323	–0.1	K619	+0.8	.....	.....
2	709	7	Q324	–1.6	K601	–1.3	.....	.....
3	764	11	.....	.....	.....	.....	H587	+0.6
4	809	12	Q326	–1.7	.....	.....	H663/727	+0.9
5	859	10	.....	.....	K602	–0.7	.....	.....
6	909	11	.....	.....	K616	+1.7	H591/726	+1.8
7	959	16	Q329	–2.3	.....	.....	H727	+0.1
8	1009	15	.....	.....	K603	–1.8	.....	.....
9	1059	16	Q331	+0.7	.....	.....	H616	+0.9
10	1109	15	Q332	–1.4	.....	.....	.....	.....
11	1159	13	.....	.....	K604	–0.9	.....	.....
12	1209	13	.....	.....	.....	.....	H622	–0.4
13	1259	14	Q335	+0.2	.....	.....	.....	.....
14	1309	11	.....	.....	K605	+0.4	.....	.....
15	1359	12	.....	.....	.....	.....	H670	+1.3
16	1409	15	Q338	+2.0	.....	.....	.....	.....
17	1459	13	.....	.....	K606	+1.3	.....	.....
18	1509	17	.....	.....	K617	+2.3	H714	+2.1
19	1559	12	Q341	+0.7	.....	.....	.....	.....
20	1609	16	.....	.....	K607	–0.2	H737	+2.2
21	1659	13	Q343	+1.1	K618	+0.1	.....	.....
22	1709	14	Q344	+1.8	.....	.....	.....	.....
23	1759	15	.....	.....	K608	–0.8	.....	.....
23	1809	11	.....	.....	.....	.....	H715	+0.1
25	1859	14	Q347	0.0	K609	0.0	H711	0.0

one going to each of the three laboratories. The drill invariably cut rings on either side of the selected ring, but errors due to this cause are thought to be small, and the number of rings cut by the drill is indicated in the results. The first sample was taken from the ring dated at A.D. 659, and the final one at A.D. 1859, making a series of 25 in all. In the first instance it was decided that in the interests of economy of effort each laboratory should measure every third sample, i. e., the Cambridge laboratory should measure numbers 1, 4, 7, . . . , Copenhagen 2, 5, 8, . . . , and Heidelberg 3, 6, 9, . . . , allowing cross-checks to be made by each laboratory upon measurements of the others after the first pattern of results had emerged. Since the radiocarbon uptake of only one tree is involved in the measurements, the possible variation due to geographic position is eliminated, and isotopic fractionation effects from sample to sample are reduced to a minimum (Craig, 1954).

The activities obtained for each sample have been extrapolated to A.D. 1859 by allowing for the radiocarbon decay, and the extrapolated activities have been compared with the activity obtained for the A.D. 1859 sample as a percentage variation from that activity. The choice of 1859 as the standard activity for this experiment allows industrial dilution effects to be ignored. The results given are at present relative values only; one of us (Münnich) has analyzed the statistical implications of cross-check samples both in this experiment and in the oxalic-acid and Heidelberg standards, and it is proposed to convert our relative scale to an absolute scale after further checks on the latter two standards. It seems unlikely that the broad conclusions derived in this paper will be materially affected.

The difficulties inherent in conducting an experiment so close to the limits of sensitivity of the technique will be readily appreciated, and certain anomalous measurements occur which are difficult to explain on purely statistical grounds. Nevertheless, certain trends in the pattern of results emerge from the graphical presentation in figure 1. It will be observed, for instance, that from A.D. 1200 to A.D. 1859 the mean values for 11 years appear above the level of the 1859 activity, whilst only one falls below it. Similarly, prior to A.D. 1200, the mean values of 4 years appear above the line and 8 below it. It might be inferred, therefore, that A.D. 1200 is in some way a cross-over point between a period whose average concentration values are higher than the 1859 value, namely, +1.0%, and a period of a slightly lower average value, –0.3%.

Although the line joining the points in figure 1 is intended as a visual aid only, it is interesting to note the general correspondence over the last 300 years with the concentration curve obtained previously by de Vries. These results appear to confirm the existence of short-term oscillations in the radiocarbon concentration, perhaps with a period of the order of 150 to 200 years, superimposed upon an oscillation having a longer period of the order of 1200 years. The underlying cause of the oscillations, however, remains obscure, as does their possible correlation with climatic phenomena. The atmospheric radiocarbon concentration could be affected either by variations in the cosmic-ray flux, perhaps in association with changes in the Earth's magnetic field, or by fluctuations in the isotopic equilibrium of the carbon-exchange reservoir. Perhaps the most promising line of inquiry might be to look for evidence of

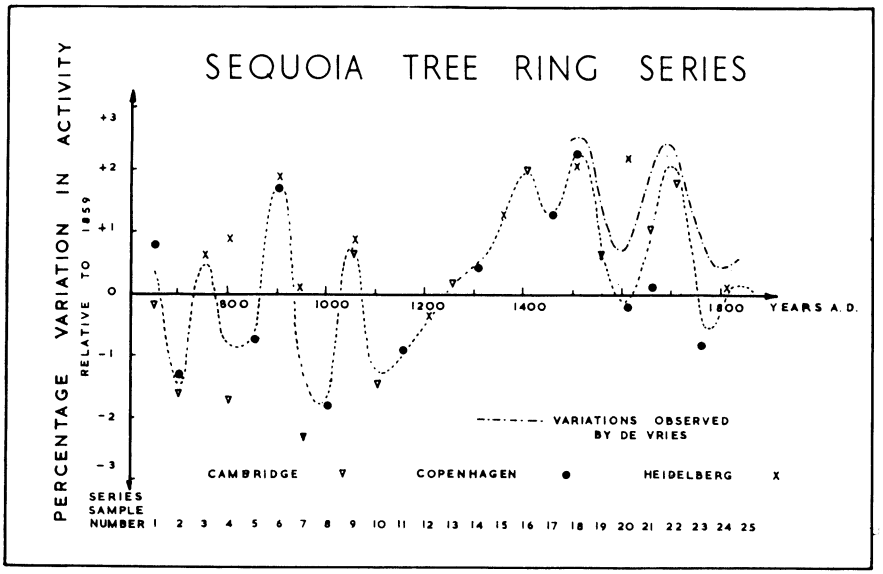

Fig. 1. The initial activities of each of the 25 samples taken at 50-year intervals from a giant sequoia, represented as a percentage variation from the activity of the wood from the 1859 tree ring. The dotted curve is intended as a visual aid only. The Copenhagen and Heidelberg measurements are both corrected for C^{13}/C^{12} variations, and the authors are greatly indebted to Dr. W. Dansgaard of Copenhagen and Dr. J. C. Vogel of Heidelberg for the measurements of these ratios.

appropriate climatic changes parallel with the observed shift in radiocarbon concentration since about A.D. 1200.

The experiment has served to demonstrate that over the past 1200 years the fundamental assumptions of the radiocarbon dating method are empirically correct to about 1.5%. Whereas the implications of an error of this magnitude might be disturbing for very recent samples, with older samples the effect might be expected to be of little significance. It is to be noted also that, although check samples between laboratories on occasion produced results outside statistical expectation, such instances were few.

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