## **Carbon-14** Trends in Subfossil Pine Stubs

Abstract. Subfossil pine stubs from a peat bog in the Netherlands were linked together dendrochronologically and sampled at intervals of 30 or 50 years for carbon-14 analysis. The results suggest that the carbon-14 content of the atmosphere was not changing between about 5400 and 5100 B.C., but that it was increasing slightly about 800 years earlier.

It has long been known that radiocarbon dates for the oldest historically datable period-that of 3rd millennium Egypt-appear to be several hundred years too young (1). The discrepancy remained unexplained until recently, when an exceptionally long dendrochronological sequence, reaching back to 5112 B.C., was established (2). The sequence is based on bristlecone pines (Pinus aristata) growing in the White Mountains of California and has been developed by Ferguson and co-workers at the Laboratory of Tree-Ring Research, University of Arizona, Tucson, Precisely dated wood samples were thus made available for calibrating the C14 time scale. This time-consuming task was undertaken by Suess (3), among others, who showed that the initial  $C^{14}$ content of tree rings, and therefore also of the atmospheric carbon dioxide, has increased more or less steadily from about 1 percent below normal 2000 years ago to 8 percent above normal at 4000 B.C. With the aid of these measurements radiocarbon dates can now be calibrated. The date of Dynasty I in Egypt, for instance, which used to be 2530 to 2400 B.C. in conventional radiocarbon years, now becomes 3290 to 3110 B.C. when converted to calendar years by means of the tree-ring calibration curve (4).

Several reasons have been proposed to explain the observed variations in the  $C^{14}$  content of the atmosphere in the past-variations in the cosmic ray intensity, activity of the sun, average global temperature, and the magnetic field of the earth. With regard to the influence of temperature it is important to ascertain whether the initial C<sup>14</sup> content continues to increase as the last ice age is approached, or whether the high value of about +8percent above normal is a maximum corresponding to the postglacial climatic optimum during the Atlantic period. For this reason the opportunity of obtaining a rather long "floating" tree-ring

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chronology (internally correct, year by year, but without calendar dates) from the numerous subfossil tree stubs preserved in a peat bog E of Emmen, Netherlands, was taken to determine the C<sup>14</sup> content of wood samples at close intervals. Provided a sufficiently long time span could be bridged by linking the stubs together dendrochronologically, it was felt that the trend in the  $C^{14}$ content prior to 4000 B.C. would be revealed. The measurements could thus provide evidence of the direction in which the initial C<sup>14</sup> content was changing at a time prior to the oldest measurements of Suess, although the magnitude of the deviations from the normal value cannot, of course, be determined.

The peat bog E of Emmen is part of the "Bourtanger Moor" (Fig. 1), the western (Dutch) arm of which was formed in an ice-marginal valley, 10 to 15 m deep, called the Hunzelaagte, which extends roughly from Emmen to Groningen. The southern part of the valley, covering approximately 100 km<sup>2</sup>, drains toward the northwest through a spillway in a west-east-oriented cover sand ridge, east of the village of Nieuw Dordrecht and along the deepest (western) part of the depression that forms the source of the rivulet Hunze. From the Late Glacial to the middle of the Atlantic period, fen peat, poor in iron, accumulated in this drainage area, 1 to 2 km broad. Because of the lowering of the water table to the south of the cover Table 1. Characteristics of the dendrochronological groups. Groups D, E, and  $E^{\circ}$  could be reassembled into a single sequence consisting of ten trees. The other groups could not be cross-dated.

Group	Years in group (No.)	Years synchro- nized (No.)	Av. age of trees (years)	Trees synchro- nized (No.)	
Α	112	92	74	4	
В	156	146	94	4	
C	135	86	97	7	
$\mathbf{D}^{\circ}$	330	264		3	
D	294	251	204	4	
E°	369	339	256	4	
Έ	193	183		2	

sand ridge, the fen peat dried out twice during the Atlantic, so that trees (Pinus sylvestris L.) could grow there over an area of about 10 km<sup>2</sup>, only to become drowned when waterlogging set in again (5). Consequently, the stumps of these pine trees have been preserved in the fen peat at two different levels. During the latter of the two dry periods especially, an extensive Pinus forest developed on the surface of the iron-poor fen peat. It is represented by numerous stumps, sometimes of astonishing size, in a wood peat layer, 10 to 70 cm thick, which preceded a period of ombrogenous peat growth.

At the beginning of the Atlantic phase, iron-rich seepage started east of the fen peat zone and ferruginous Hypnaceae peat began accumulating in



Fig. 1. Map showing the locality of the pine stubs in the peat bog. Locations A to E indicate where the different groups were collected. [Drawing by J. Klein]

Table 2. Carbon-14 analysis of tree-ring samples. Column 6, relative  $C^{13}/C^{12}$  ratio with respect to Peedee belemnite standard. Column 7,  $C^{14}$  activity with respect to 0.95 of that of the U.S. National Bureau of Standards oxalic acid standard. Column 8, conventional  $C^{14}$  age in years before 1950 (6). Column 9, deviation in age-corrected  $C^{14}$  activity from average for group.

Anal- ysis No. GrN- (1)	Tree (2)	Sam- ple (3)	Sampled ring- years of tree (4)	Av. age in group (years) (5)	δ C <sup>13</sup> (%) (6)	C <sup>14</sup> activity (% modern) (7)	Con- ventional C <sup>14</sup> age (years B.P.) (8)	Δ C <sup>14</sup> (%) (9)
				Group	DEE	)		
4913	E°4	-1	31-39	35	-25.8	$45.54 \pm 0.18$	$6325 \pm 32$	+10.8
4914		-2	61–68	65	-24.7	$44.89 \pm .17$	$6440 \pm 30$	- 7.3
4915		-3	90–100	95	-25.2	$45.05 \pm .18$	$6410 \pm 32$	- 7.1
4916			120-129	125	-25.0	$45.63 \pm .18$	$6310 \pm 32$	+ 2.0
4917		-5	150-160	155	24.7	$45.32 \pm .17$	$6365 \pm 30$	- 8.5
4918		6	180–190	185	-24.8	$46.49 \pm .17$	$6160 \pm 30$	+13.4
4279	E°1	-1	57-67	169	-24.9	$45.74 \pm .18$	$6290 \pm 32$	- 1.4
4277		-2	107-117	219	-24.8	$46.08 \pm .19$	$6230 \pm 33$	+ 0.3
4278		-3	157–167	269	-24.8	$46.11 \pm .18$	$6225 \pm 32$	- 5.1
4276		-4	207–217	319	-25.3	$46.52 \pm .17$	$6150 \pm 30$	- 2.2
4301	E 4	-1	22-32	169	-27.3	$46.25 \pm .37$	$6200 \pm 65$	+ 9.9
4302		-3	122-132	269	-24.2	$46.13 \pm .36$	$6220 \pm 63$	- 4.6
4303		4	172-182	319	-24.3	$46.98 \pm .37$	$6075 \pm 63$	+ 7.6
4314	D 2	-1	115-123	223	-24.6	$46.42 \pm .37$	$6170 \pm 64$	+ 7.2
				Gr	ade D°			
4263	D°1	-1	29-39	55	-26.4	$41.20 \pm .18$	$7130 \pm 35$	+ 2.4
4264		-3	84-94	110	-26.3	$40.99 \pm .16$	$7170 \pm 30$	- 9.3
4265		-4	134-144	160	-24.6	$41.74 \pm .16$	$7025 \pm 30$	+ 2.9
4266		5	184-194	210	-24.2	$41.84 \pm .16$	$7005 \pm 35$	- 0.9
4267		-6	234-244	260	-25.2	$42.34 \pm .16$	$6910 \pm 30$	+ 4.9
4274	<b>B</b> 2	1	1-10	5	-26.8	$47.16 \pm .33$	$6045 \pm 60$	
4275		5	100–109	105	-26.1	$48.06 \pm .38$	$5890 \pm 65$	
4313	C 4	1	74-89	128	-24.6	$54.04 \pm .28$	$4950\pm40$	
4312	<b>A</b> 2	1	30-40	52	-24.8	$53.77 \pm .39$	$4990\pm60$	



Fig. 2. Deviations of age-corrected C<sup>14</sup> content of tree-ring samples from the average for the group versus relative age in floating tree-ring chronology. Dashed line, best straight line through the points. Derived conventional C<sup>14</sup> age (6) of "year 0" is 6440 years B.P. for group D E E<sup> $\circ$ </sup> and 7200 years B.P. for group D<sup> $\circ$ </sup>.

that part of the valley (see Fig. 1). This deposit of moss peat came into being more or less independently of the fen peat in the drainage area. Toward the end of the Atlantic the seepage suddenly ceased and Pinus started to grow over the desiccated ferruginous peat area (15 km<sup>2</sup>), although a dense forest was not formed as in the fen peat area. Subsequently this region was also covered with ombrogenous Sphagnum peat. Since the pine woods appeared only after sudden desiccation of the peat bog, it can be assumed that on each occasion the trees began to grow more or less simultaneously and that it would be possible to link them dendrochronologically. For this purpose sections of some 52 stumps were collected at five different places, indicated by the letters A to E on the map (Fig. 1). With the exception of groups E and E°, which were still in situ, the sections were taken from stumps left behind by the peat diggers and lying more or less in their original position.

Preliminary radiocarbon dating revealed that the groups A and C had an age of about 5000 radiocarbon years (6); groups B, D, E, and E° about 6000 years; and group D° about 7000 years. Thus group D° could be assigned to an early Atlantic desiccation, while the bulk of the stumps (B, D, E, E°) belonged to a middle Atlantic pine forest, and groups A and C to the end of this period.

At each of the localities there was abundant material, but only a small number of the stumps was suitable for dendrochronological investigation (7). In groups A, B, and C the age of the trees was, in general, too low (less than 50 years) and the growth of the annual rings very irregular. In groups D°, D, E°, and E the trees had attained a considerable age (in some cases more than 200 years), but the wood was often in a bad state of preservation.

A total of 52 samples was analyzed. The methods used are those already applied successfully to living and subfossil Pinus sylvestris (8). For every specimen the thickness of each tree ring was plotted against its number, and the individual curves thus obtained for a group were matched against one another. In this manner a floating chronology was established for each group and an average "group curve" constructed. An attempt to cross-date the groups was then made by comparing both the individual curves and the group curves. The synchronizations were checked by deducing various

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qualitative and quantitative characteristics of the curves (coefficient of coincidence, analysis of characteristic years, and so forth). Those individual trees that could not be matched were rejected. The lengths of the group chronologies vary considerably, depending on the age of the specimens and the degree of overlap. The principal characteristics of each chronology are given in Table 1.

From the investigation it can be concluded that (i) at each locality several pine trees were at least partly coeval; (ii) at the edge of the bog the simultaneousness can be demonstrated over a relatively large area (D E E°), where the pine forest existed for more than three centuries; (iii) the large proportion of curves, which could not be cross-dated (24 out of 52), indicates irregular growth and cannot be considered as proof of differing ages; and (iv) in the center of the bog the groups cannot be cross-dated, undoubtedly because the sequences are not long enough and the growth too irregular. Of the two series that were relatively long (D°, 330 years, and D E E°, 369 years), samples that comprised not more that 10 ring-years each were selected at regular intervals of 50 or 30 years for radiocarbon analysis. Unfortunately, the complete range could not be spanned owing to the condition of the cores of stumps and the breadth of outermost rings. Of group  $D^{\circ}$ , five samples were analyzed from stump D°1, spanning 205 years; and of group D E E°, 14 samples, spanning 284 years, were analyzed (Table 2).

When all the results obtained for each group are used, the conventional radiocarbon dating (6) for group D° with 330 ring-years is 7200 to 6870 years before the present (B.P.), while that for group D E E° with 369 ring-years is 6440 to 6070 years B.P. The outermost rings of the younger group thus have approximately the same age as the oldest samples of the bristlecone pine chronological sequence which has been radiocarbon-dated to approximately 6130 years B.P. (conventional) (9). If the bristlecone pine chronology is accepted, the actual (solar) age of the youngest sample in group D E E° corresponding to 6120 conventional radiocarbon years, is about 5100 years B.C. The D° group should be approximately 800 years older.

The C<sup>14</sup> activity of each sample was corrected for variations in isotope fractionation by means of the  $C^{13}/C^{12}$ 28 NOVEMBER 1969

ratio (10) and reduced to the expected value at year 0 of the group chronology, with the use of a half-life of 5730 years. The deviation in activity from the average, a delta-equivalent, is given in the last column of Table 2 and plotted in Fig. 2.

The younger group shows no trend in the C14 activity over the 284-year span: the slope of the best straight line through the points is  $-0.6 \pm 2.4$  per mille per century. When this result is compared with the decrease in initial C<sup>14</sup> content over the period 4000 to 3000 B.C., of 4 per mille per century (3), we must conclude that this rapid decrease did not take place during the three centuries covered by the group. There is, however, an indication of a short-time fluctuation in C14 activity during this period: the sample  $E^{\circ}$  4-6 from the years 180 to 190 on the floating scale has 1.35 percent more  $C^{14}$  than the average, which causes it to seem 110 years too young. The older series, D°1, shows an increase in the initial C<sup>14</sup> content amounting to  $+2.5 \pm 3.7$  per mille (best straight line) over the two centuries covered.

Although the length of the two floating chronologies is admittedly too short to gain certainty about the trend in C<sup>14</sup> prior to 5000 B.C., the indications are that at about 6000 B.C. (solar years) the C<sup>14</sup> content was increasing; at about 5000 B.C. the level was more or less constant, to start decreasing again after 4000 B.C.

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## **References and Notes**

- H. de Vries, G. W. Barendsen, H. T. Waterbolk, Science 127, 129 (1958); H. de Vries and H. T. Waterbolk, *ibid.* 128, 1550 (1958).
   C. W. Ferguson, *ibid.* 159, 839 (1968).
   H. E. Suess, J. Geophys. Res. 70, 5937 (1965);
- H. E. Suess, J. Geophys. Res. 70, 5937 (1965); in Radioactive Dating and Methods of Low-Level Counting (Intern. Atomic Energy Agency, Vienna, 1967), p. 143.
   ——, Z. Phys. 202, 1 (1967); J. C. Vogel, Helinium 9, 3 (1969).
   A detailed investigation of the structure and concrete of this part hog is being conducted
- genesis of this peat bog is being conducted by W. A. Casparie.
- 6. Age calculated with a half-life for  $C^{14}$  of 5570 years and assuming the initial  $C^{14}$  content of the samples, after correction for variations in isotope fractionation, to have been equal to 0.95 of that of the U.S. National Bureau of Standards oxalic acid  $C^{14}$  standard. B.P. B.P., before the present, that is, before 1950.
- 7. B. Huber, Holz Roh Werkstoff 6, 263 (1943);

E. Schulman, Dendroclimatic Changes in Semiarid Arizona (Univ. of Arizona Press, Tucson, 1956).

- 8. A. V. (1966). V. Munaut, Agricultura 14, 193 and 361
- 1960). 9. E. K. Ralph, H. N. Michael, Archaeometry 10, 1 (1967).
- 10, 1 (1967).
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## Nonexistence of Large Mascons at Mare Marginis and Mare Orientale

Abstract. The analysis of line-ofsight residual accelerations from Lunar Orbiters 3 and 5 does not show any evidence for large mascons near the lunar limbs. Although unfavorable geometry reduces the acceleration effect due to any mascon near the limb, simulations show that large masses at Mare Orientale and Mare Marginis would produce substantial accelerations, in complete disagreement with the actual Doppler tracking data obtained from a Lunar Orbiter experiment.

The observation of large accelerations of Lunar Orbiter spacecraft above the ringed maria on the lunar nearside (1), and the association of these accelerations with mass concentrations (mascons) immediately beneath these maria (1), have prompted speculation that all ringed maria have associated mascons whose masses increase with increasing ring diameter. In particular, Campbell et al. have suggested that there are mascons at Mare Orientale and Mare Marginis which are 1.43 and 2.42 times the size of the Mare Imbrium mascon, respectively (2). We show here that typical Doppler tracking data, from Lunar Orbiter spacecraft passing near these features, are in complete disagreement with such an assignment of masses.

Figure 1 shows the Doppler residuals, after a least-squares estimation of the six parameters of spacecraft state at epoch, for an orbit of low inclination (21°). This orbit of Lunar Orbiter 3 traversed from west to east, across the nearside of the moon, passing over Mare Orientale and Mare Crisium and near Mare Marginis at the indicated times. For comparison, Fig. 1 also shows a simulation of the residuals which would result for a hypothetical spacecraft on the same trajectory over a spherical moon having eight mascons, with the sizes and locations postulated by Campbell et al. (2). The Doppler signatures at the regions of Mare Ori-