GEOPHYSICAL RESEARCH LETTERS, VOL. 25, NO. 9, PAGES 1321-1324, MAY 1, 1998

Temporal variation in the interhemispheric ^{14}C offset

F.G. McCormac[1], A.G. Hogg[2], T.F.G. Higham[2], J. Lynch-Stieglitz[3], W.S. Broecker[3], M.G.L. Baillie[1], J. Palmer[4], L. Xiong[4], J.R. Pilcher[1], D. Brown[1] and S.T. Hoper[1]

Abstract. Contemporaneous tree-ring dated wood, from trees in the northern and southern hemispheres, gives different ^{14}C dates. Previous studies [Vogel et al., 1986; 1993] using wood from South Africa and The Netherlands have shown depletion's of -4.56 ± 0.85‰ and -5.12 ± 0.62‰ respectively. This translates to age differences of 36 ± 7 and 41 ± 5 years (yrs) with the southern hemisphere giving the older dates. More recently, Stuiver and Braziunas [1998] have shown that an offset of 23 ± 4 yrs exists between combined 19th century wood measurements from Tasmania and Chile in the southern hemisphere and the west coast of the U.S. (Washington) in the northern hemisphere. In this study measurements on contemporaneous decadal samples of oak from the British Isles and cedar from New Zealand over the period 1725 to 1885 AD show a depletion of -3.4 ± 0.58‰ (27.2 ± 4.7 yrs). However, data after 1895 AD has a mean offset of 0.66 ± 1.06‰ (-5.3 ± 8.5 yrs) with increased variance compared to 19th century data. This, we believe, is attributable to anthropogenic fossil fuel, which, due to its long residence time in the earth, has long since lost any ^{14}C component and when burned preferentially depletes the northern hemisphere atmosphere of ^{14}C.

Introduction

The growth rings of trees provide an annually dated record of atmospheric conditions at the location of growth. However, contemporaneous wood from different locations within a given hemisphere does not always give identical $\Delta^{14}C$ measurements [Damon, 1995; McCormac et al., 1995; Stuiver and Braziunas, 1998]. Species to species fractionation effects can result in contemporaneous wood having a different specific activity but this is corrected using $\delta^{13}C$ measurements. Pre-treatment of the wood to α-cellulose ensures that the molecular composition of the fraction of wood dated is the same for the different species used [Stuiver et al., 1984; Hoper et al., 1997] and so variable lignin and/or other cellulose compounds cannot contribute to $\Delta^{14}C$ differences. Remaining offsets must therefore reflect real atmospheric differences in $\Delta^{14}C$ even if these are merely the result of early or late onset of tree growth in a given region with the tree sampling an annually varying $\Delta^{14}C$ level [Stuiver and Braziunas, 1998].

Rings from trees, that grew in the same year, in opposite hemispheres, give different carbon dates (southern hemisphere carbon dates of wood of the same age are older by about 40 yrs)[Lerman et al.,1970]. This is because the larger expanse of ocean and slightly higher wind speeds cause more ^{14}C depleted CO_2 from the ocean to enter the southern atmosphere than the northern [Cain and Suess, 1976]. This natural offset was thought to be constant through time, however, here we show that, small changes with time do occur and the burning of fossil fuels in the industrial northern hemisphere has resulted in a slow change in the natural isotopic balance of the atmosphere between the hemispheres.

Measurements

In this study we have used oak (*Quercus petraea*) from Sherwood Forest, England (53° 12'N 01° 04'W altitude 80m) (1725 - 1745 AD) and Shane's Castle, Co. Antrim, N. Ireland (54° 44'N 06° 16'W altitude 20m) (1755 - 1935 AD) and cedar (*Libocedrus bidwillii*) from Hihitahi State Forest Sanctuary in the North Island of New Zealand (39° 32'S 175° 44'E altitude 976m). Two trees, one cedar one oak, were tree-ring dated and blocks of 125g were separated. Samples were pre-treated to α-cellulose [Hoper et al., 1997] thereby removing all mobile fractions and the ^{14}C activities were determined by liquid scintillation counting of benzene [McCormac, 1992; McCormac et al., 1993]. Small differences in the ^{14}C activities of wood from different locations are extremely difficult to detect. If wood from different regions is measured in different laboratories then small systematic differences between laboratories can easily mask any atmospheric signal that may exist [Damon et al., 1989; McCormac et al., 1995]. We therefore measured both the oak and cedar in the same laboratory and then replicated the measurements between the laboratories at The Queen's University of Belfast, N. Ireland and The University of Waikato, New Zealand. This approach is vindicated because the data shows an offset of ~10 yrs between laboratories for identical samples. This does not in any way affect the measured relative differences between the two species of tree.

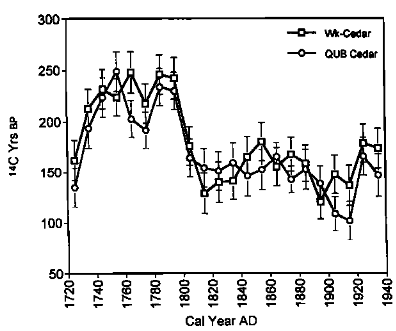

Figure 1. Independent ^{14}C measurements (yrs BP) on decadal samples of cedar from New Zealand (39°S) made in QUB and Waikato. Error bars for all figures are ± 1σ which includes the Poisson counting error and an error multiplier to account for overall laboratory reproducibility.

[1] Palaeoecology Centre, The Queen's University of Belfast, Belfast, Northern Ireland
[2] Radiocarbon Dating Laboratory, University of Waikato, Hamilton, New Zealand
[3] Lamont-Doherty Earth Observatory and Department of Earth and Environmental Sciences, Columbia University, Palisades, NY
[4] Plant Sciences Department, Lincoln University, Lincoln, New Zealand

Paper number 98GL01065.
0094-8534/98/98GL-01065$05.00

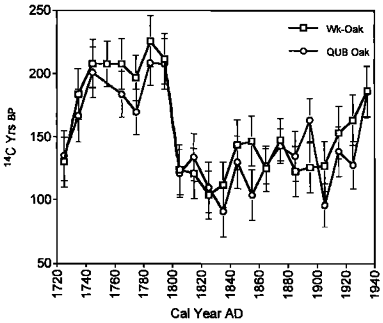

Figure 2. Independent ^{14}C measurements (yrs BP) on decadal samples of oak from the British Isles (53°N, Sherwood Forest, England and 54°N, Shane's Castle, N. Ireland) made in QUB and Waikato.

The replicate measurements are shown in Figures 1 and 2. The time structure of both the cedar and oak measurements is similar to previously published data from the same periods [Stuiver and Becker, 1993; Pearson et al., 1986].

Weighted differences of the cedar and oak measurements shows a consistent depletion in the southern hemisphere of -3.4 ± 0.58‰ until the decade centred on 1885. After that the results from both laboratories show a reversal such that the northern hemisphere is slightly depleted and thereafter the depletion appears to alternate between hemispheres. A similar change in hemispheric offset has been recently reported [Stuiver and Braziunas, 1998]. Table 1 gives our individual measurements on decadal samples of oak and cedar from both laboratories. Errors on the measurements include both the Poisson counting error and an error multiplier to account for overall laboratory reproducibility. Also given are the per mil. differences between cedar and oak determined independently in both laboratories.

Table 2 shows the error weighted mean of the differences between oak and cedar for selected periods. The offset between cedar and oak over the entire set of measurements is -2.46 ± 0.51‰ (19.7 ± 4.1 yrs). However Figure 3 shows that the offset changes considerably after 1895 and if data before and after this period are compared the offset is -3.40 ± 0.58‰ (27.2 ± 4.7 yrs) for the period up to 1885 and 0.66 ± 1.06‰ (-5.3 ± 8.5 yrs) for the post 1885 interval.

An error weighted running mean on the combined offset data is shown in Figure 4. Here sets of 3 decadal offset measurements from each laboratory have been averaged and the age difference plotted against the central date. This shows some variability with time of the hemispheric offset notably between the intervals 1725 - 1795 AD (offset 23.1 ± 7.0 yrs), 1805 - 1865 AD (offset 34.6 ± 7.1 yrs) and again the large change around 1895 AD thought to be related to the anthropogenic input of fossil fuel CO_2 to the northern hemisphere atmosphere. The pre-anthropogenic change in offset value for the two earlier intervals given above is 11.5 ± 10.0 yrs. Although not highly significant this result is suggestive of small temporal changes in the pre-anthropogenic hemispheric offset value. The changes in Δ^{14}C as a function of time are evident in Figure 5 which shows the combined oak measurements (circles) and combined cedar measurements (squares) from both laboratories.

Modelling

The relative depletion of ^{14}C in the southern hemisphere atmosphere before 1885 can be attributed to the differences in ocean circulation between the two hemispheres. In the Southern Ocean, surface ocean ^{14}C is low, reflecting the exposure of old, deep waters at the surface. While there is a similar but smaller depletion of ^{14}C in the North Pacific, the ^{14}C in North Atlantic surface waters remains high reflecting their origins in low latitudes. The ocean effect on atmospheric ^{14}C gradients has previously been shown in both a simple one-dimensional atmospheric model [Levin et al., 1987] as well as an atmospheric transport model from the GISS GCM [Braziunas et al., 1995]. Here, we estimate the magnitude of this pre-anthropogenic interhemispheric gradient by using a simple two box (northern and southern hemispheres) atmospheric

Table 1. ^{14}C Measurements (yrs BP) on Decadal Samples of Cedar and Oak and Hemispheric Differences in ^{14}C (‰) Determined Independently in Belfast (QUB) and Waikato (Wk)

Year AD	Cedar-Wk	Cedar-QUB	Oak-Wk	Oak-QUB	Diff.-Wk‰	Diff.-QUB‰
1725	162 ± 20	135 ± 19	130 ± 20	135 ± 20	-4.0 ± 3.5	0.0 ± 3.5
1735	213 ± 19	194 ± 20	184 ± 20	167 ± 21	-3.6 ± 3.5	-3.4 ± 3.6
1745	232 ± 19	224 ± 19	208 ± 19	201 ± 20	-3.0 ± 3.4	-2.9 ± 3.5
1755	224 ± 18	249 ± 19	208 ± 18		-2.0 ± 3.1	
1765	248 ± 20	203 ± 18	208 ± 20	184 ± 18	-5.0 ± 3.5	-2.4 ± 3.1
1775	218 ± 19	192 ± 18	197 ± 18	170 ± 19	-2.6 ± 3.2	-2.8 ± 3.1
1785	246 ± 19	234 ± 18	226 ± 20	209 ± 18	-2.5 ± 3.5	-3.1 ± 3.1
1795	242 ± 20	230 ± 18	212 ± 20	208 ± 20	-3.8 ± 3.5	-2.8 ± 3.4
1805	176 ± 19	164 ± 19	124 ± 20	121 ± 19	-6.5 ± 3.4	-5.4 ± 3.4
1815	129 ± 20	154 ± 19	121 ± 20	134 ± 19	-1.0 ± 3.5	-2.5 ± 3.4
1825	140 ± 20	151 ± 19	104 ± 19	110 ± 20	-4.5 ± 3.2	-5.1 ± 3.5
1835	141 ± 18	159 ± 20	112 ± 18	91 ± 21	-3.6 ± 3.1	-8.5 ± 3.5
1845	165 ± 20	146 ± 20	144 ± 20	130 ± 21	-2.6 ± 3.5	-2.0 ± 3.6
1855	180 ± 19	152 ± 20	147 ± 20	104 ± 20	-4.1 ± 3.5	-6.0 ± 3.5
1865	155 ± 19	165 ± 14	125 ± 18	127 ± 14	-3.8 ± 3.2	-4.8 ± 2.5
1875	167 ± 17	142 ± 13	148 ± 17	143 ± 22	-2.4 ± 3.0	0.1 ± 1.9
1885	158 ± 18	152 ± 20	123 ± 20	135 ± 20	-4.4 ± 3.1	-2.1 ± 3.5
1895	120 ± 17	138 ± 17	126 ± 20	164 ± 17	0.8 ± 3.0	3.2 ± 3.0
1905	147 ± 19	108 ± 17	127 ± 20	96 ± 17	-2.5 ± 3.5	-1.5 ± 3.0
1915	136 ± 20	101 ± 19	154 ± 21	139 ± 20	2.2 ± 3.5	4.8 ± 3.5
1925	178 ± 19	165 ± 19	164 ± 20	128 ± 19	-1.7 ± 3.4	-4.6 ± 3.4
1935	173 ± 20	146 ± 21	187 ± 20	186 ± 20	1.7 ± 3.1	5.0 ± 3.6

Table 2. Cedar Minus Oak Determined Independently in Both Laboratories (offsets given in yrs)

	1725-1935	1725 - 1885	1895 - 1935
Waikato	21.5 ± 5.8	27.7 ± 6.6	-0.4 ± 12.4
QUB	17.9 ± 5.8	26.8 ± 6.6	-9.6 ± 11.7
Combined	19.7 ± 4.1	27.2 ± 4.7	-5.3 ± 8.50

$x = \Sigma (x_i/\sigma_i^2) / \Sigma (1/\sigma_i^2)$ and $1/\sigma^2 = \Sigma (1/\sigma_i^2)$

model in which we assume that the concentration of atmospheric ^{14}C in each hemisphere reflects a balance between production, exchange with the ocean, and exchange between hemispheres (Table 3). Our "best guess" values for exchange terms and surface ocean values give a depletion in the southern hemisphere of -3.0 ‰. This value is sensitive to the interhemispheric mixing time, the accuracy of our reconstruction of pre-anthropogenic surface ocean $\Delta^{14}C$ and the production rate of ^{14}C. While we present results for our "best guess" values, reasonable adjustments in any of these poorly constrained parameters can bring the calculated average interhemispheric gradient into agreement with the tree ring measurements given above (-3.4‰) as well as the more complex models of Levin et al. [1987] (-4 ‰) and Braziunas et al. [1995] (-3.1‰). Also, while we present the difference between average values in each hemisphere, the more detailed study of Braziunas et al. [1995] shows that there is considerable structure in $\Delta^{14}C$ within each hemisphere, with the difference for the latitudes of the trees in this study approximately 30% higher than the average interhemispheric gradient

The switch to relative depletion of ^{14}C in the northern hemisphere is a consequence of the addition of fossil fuel carbon (no ^{14}C) primarily to the northern hemisphere atmosphere. We can calculate the expected change in the atmospheric $\Delta^{14}C$ gradient by calculating the increase in carbon in each hemisphere of our two box model due to the addition of fossil fuel carbon into the northern hemisphere and the uptake of part of this carbon by ocean and/or land biota. We add fossil fuel carbon to the northern hemisphere atmosphere using the fluxes from Keeling [1973] (before 1950) and Marland and Rotty [1984] (after 1950). It is assumed that 60% of the fossil fuel emissions remain in the atmosphere, consistent with ocean carbon cycle models [Siegenthaler and Joos, 1992]. While there is much debate [Tans et al., 1990; Heimann and Keeling, 1989; Broecker and Peng, 1982] as to where the excess carbon is actually absorbed into the ocean and atmosphere, we simply assume a null biosphere and add the carbon to the northern (41%) and southern (59%) hemisphere ocean in proportion to their areas and gas exchange rates. The magnitude of the calculated northern hemisphere depletion is consistent with the observations from the tree rings (Figure 4). The atmospheric gradient in $\Delta^{14}C$ induced by the input of fossil fuel carbon into the atmosphere is, like the preindustrial gradient, sensitive to the interhemispheric mixing time, but is also sensitive to the location of the uptake of fossil fuel carbon. If we put 80% of the fossil fuel that does not remain in the atmosphere into a northern hemisphere sink [Tans et al., 1990] the magnitude of the interhemispheric gradient drops only to 0.9 ‰ in 1950 vs. 0.2‰ when 41% of the carbon is absorbed in the northern hemisphere. While we show the $\Delta^{14}C$ gradient induced by the input of fossil fuel carbon into the atmosphere after 1952, in reality the initiation of atmospheric testing of atomic weapons at this time would once again produce higher atmospheric ^{14}C in the northern hemisphere.

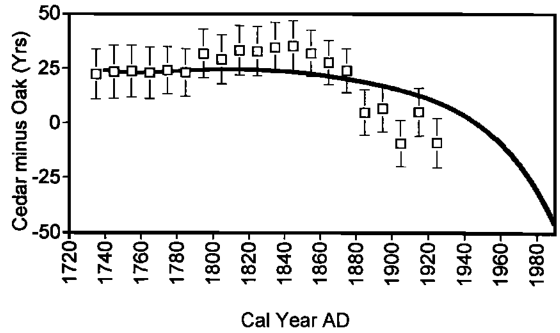

Figure 4. Three decade running mean of hemispheric differences in ^{14}C. Model results are superimposed (solid line).

Conclusions

^{14}C measurements made on contemporaneous decadal blocks of tree rings from the British Isles and New Zealand show a depletion of -3.4 ± 0.58 ‰ for the period 1725 to 1885 AD and a mean offset of 0.66 ± 1.06 ‰ for the period post 1895 AD. Calculations using a simple two box atmospheric model are consistent with this being caused by a depletion of the ^{14}C content of the atmosphere as a result of anthropogenic input of fossil fuel CO_2 into the northern hemisphere atmosphere since the time of the industrial revolution. A small change in the pre-anthropogenic offset beween the periods 1725 - 1795 AD and 1805 - 1865 AD is suggestive of a natural time varying component to the hemispheric offset.

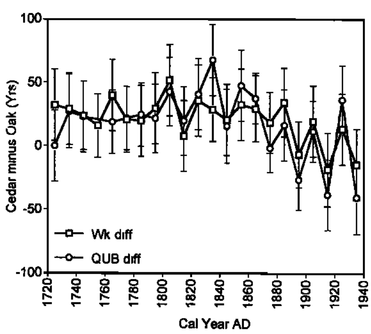

Figure 3. Differences in ^{14}C dates (yrs) of southern hemisphere (cedar) and northern hemisphere (oak) wood measured by QUB and Waikato (Wk).

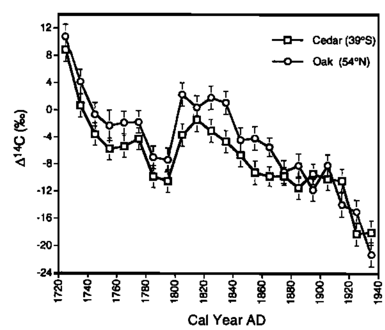

Figure 5. $\Delta^{14}C$ (‰) values for the combined cedar (southern hemisphere) and combined oak (northern hemisphere) measurements. The disappearance of the hemispheric offset around 1895 AD is evident.

Table 3. Parameters Used to Calculate Steady State ^{14}C Concentration in Northern and Southern Hemisphere Atmosphere. Air-sea Fluxes of Carbon and ^{14}C are Calculated for 10° Latitude Bands for Each Ocean

Time constant for interhemispheric exchange of CO_2[1]	1.2 yr
Northern Hemisphere	
CO_2 in atmosphere[2]	26 x 10^{15} moles C
^{14}C production rate[3]	230 moles ^{14}C/yr
Air-Sea exchange[4]	2.5 x 10^{15} moles C/yr
$\Delta^{14}C$ of CO_2 transfered from sea to air[5]	-55 ‰
Southern Hemisphere	
CO_2 in atmosphere[2]	26 x 10^{15} moles C
^{14}C production rate[3]	230 moles ^{14}C/yr
Air-Sea exchange[4]	4.2 x 10^{15} moles C/yr
$\Delta^{14}C$ of CO_2 transferred from sea to air[5]	-65 ‰

[1] Tans et al. [1978] [2] Broecker and Peng [1982] [3] Broecker and Peng [1974]

[4] Air-Sea flux is area weighted gross flux using wind-speed dependent exchange relationship in Tans et al. [1990], pCO_2 = 280. Sea-air exchange is area weighted gross flux using wind-speed dependent exchange relationship and surface ocean ΔpCO_2 in Tans et al. [1990], mean ocean pCO_2 = 280. Modern ΔpCO_2 are adjusted everywhere by 6 μatm for no net flux of CO_2 to the ocean.

[5] Surface ocean ^{14}C (pre-bomb) from Broecker and Peng [1982] corrected to pre-anthropogenic values by adding 10 ‰ to warm surface water values. The surface ocean $\Delta^{14}C$ is weighted by the CO_2 flux from the ocean for each 10° latitude band and each ocean.

Acknowledgements. This work was supported by a research grant from the Natural Environment Research Council (NERC), Grant No GR9/02597 to Dr F.G. McCormac at The Queen's University of Belfast and by a grant to Dr A.G. Hogg at The University of Waikato, by the New Zealand Foundation for Research, Science and Technology Grant number UOW-508.

References

Braziunas, T.F., I.Y. Fung, and M. Stuiver The pre-industrial atmospheric $^{14}CO_2$ latitudinal gradient as related to exchanges among atmospheric, oceanic and terrestrial reservoirs. *Global Biogeochemical Cycles 9*, 565-584, 1995.

Broecker, W.S., and T-H Peng *Tracers in the Sea*, Eldigio, Palisades, New York, 1982.

Broecker, W.S., and T-H Peng, Gas exchange rates between air and sea, *Tellus, 26.*, 21-35, 1974.

Cain, W.F., and H.E. Suess, Carbon-14 in tree-rings, *J. Geophys. Res.*, *82*, 3688-3694, 1976.

Damon, P.E., S. Cheng, and T.W. Linick, Fine and hyperfine structure in the spectrum of secular variations of atmospheric ^{14}C, *Radiocarbon 31*, 704-718, 1989.

Damon, P.E., A note concerning "location-dependent differences in the ^{14}C content of wood" by McCormac et al., *Radiocarbon, 37*, 829-830, 1995.

Heimann, M., and C.D. Keeling, A three-dimensional model of atmospheric CO_2 transport based on observed winds, 2, Model description and simulated tracer experiments, in *Aspects of Climate Variability in the Pacific and Western Americas*, edited by D.H. Peterson, pp. 237-275, Geophy. Monograph Series, 55, AGU, Washington DC, 1989.

Hoper, S.T., F.G. McCormac, A.G. Hogg, T.F.G. Higham, and J. Head, Evaluation of wood pre-treatment on oak and cedar, *Radiocarbon* (in press), Proceedings of the 16th International ^{14}C Conference, 1997.

Keeling, C.D., Industrial production of carbon dioxide from fossil fuels and limestone,*Tellus, 35*, 174-198, 1973.

Lerman, J.C., W.G. Mook, and J.C. Vogel, ^{14}C in tree rings from different localities, in *Radiocarbon Variations and Absolute Chronology*, edited by I.U. Olsson, Proceedings, XII Nobel Symposium, Wiley, New York, 275-301, 1970.

Levin, I., B. Kromer, D. Wagenbach, and K.O. Munnich, Carbon isotope measurements of atmospheric CO_2 at a coastal station in Antarctica, *Tellus, 39B*, 89-95, 1987.

Marland, G., and R.M. Rotty, Carbon dioxide emissions from fossil fuels: a procedure for estimation and results from 1950-1982,*Tellus, 36B*, 232-261, 1984.

McCormac, F.G., Liquid scintillation counter characterization, optimization and benzene purity correction, *Radiocarbon, 34*, 37-45, 1992.

McCormac, F.G., R.M., Kalin, and A. Long, Radiocarbon dating beyond 50,000 years by liquid scintillation counting, in *Liquid Scintillation Spectrometry* , edited by Noakes, J.E., Schonhofer, F., and Polach, H., pp. 125-133, Radiocarbon, Tucson, Arizona, 1993.

McCormac, F.G., M.G.L., Baillie, J.R., Pilcher, and R.M. Kalin, Location dependent differences in the ^{14}C content of wood, *Radiocarbon, 37*, 395-407, 1995.

Pearson, G.W., J.R., Pilcher, M.G.L., Baillie, D.M., Corbett, and F. Qua, High-precision ^{14}C measurement of Irish oaks to show the natural ^{14}C variations from AD 1840 to 5210 BC, *Radiocarbon, 28*, 911-934, 1986.

Siegenthaler, U., and F. Joos, Use of a simple model for studying the oceanic tracer distributions and the global carbon cycle,*Tellus, 44B*, 186-207, 1992.

Stuiver, M., R.L. Burk and, P.D. Quay, $^{13}C/^{12}C$ ratios in tree rings and the transfer of biospheric carbon to the atmosphere, *J. Geophys. Res.*, *89*, 11731-11748, 1984.

Stuiver, M., and B. Becker, High-precision decadal calibration of the radiocarbon timescale, AD 1950-6000 BC, *Radiocarbon, 35*, 35-65, 1993.

Stuiver, M., and T. Braziunas, Anthropogenic and solar components of hemispheric ^{14}C, *Geophys. Res. Lett.*, *25*, 329-332, 1998.

Tans, P.P., I.Y. Fung, and T. Takahashi, Observational constraints on the global atmospheric CO_2 budget, *Science, 247*, 1431-1438, 1990.

Vogel, J.C., A., Fuls, E. Visser, and B. Becker, Radiocarbon fluctuations during the third millennium BC, *Radiocarbon, 28*, 935-938, 1986.

Vogel, J.C., A., Fuls, E. Visser, and B. Becker, Pretoria calibration curve for short-lived samples, *Radiocarbon, 35*, 73-85, 1993.

F.G. McCormac, M.G.L. Baillie, J.R. Pilcher, D. Brown and S.T. Hoper, Palaeoecology Centre, School of Geosciences, The Queen's University of Belfast, Belfast BT7 1NN, Northern Ireland. (e-mail: f.mccormac@qub.ac.uk; m.baillie@qub.ac.uk; j.pilcher@qub.ac.uk; d.brown@qub.ac.uk; s.hoper@qub.ac.uk)

A.G. Hogg and T.F.G. Higham, Radiocarbon Dating Laboratory, University of Waikato, Private Bag 3105, Hamilton, New Zealand. (e-mail: ahogg@waikato.ac.nz; thigham@waikato.ac.nz)

J. Lynch-Stieglitz and W.S. Broecker, Lamont-Doherty Earth Observatory and Department of Earth and Environmental Sciences, Columbia University, Palisades, NY 10964 (e-mail: jean@ldgo.columbia.edu; broecker@ldgo.columbia.edu)

J. Palmer and L. Xiong, Plant Sciences Department, Lincoln University, Lincoln, New Zealand. (e-mail: palmer@kauri.lincoln.ac.nz)

(Received December 11, 1997; revised March 2, 1998; accepted March 11, 1998)