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INVESTIGATING THE INTERHEMISPHERIC ^{14}C OFFSET IN THE 1ST MILLENNIUM AD AND ASSESSMENT OF LABORATORY BIAS AND CALIBRATION ERRORS

Alan Hogg[1] • Jonathan Palmer[2] • Gretel Boswijk[3] • Paula Reimer[4] • David Brown[4]

ABSTRACT. Past measurements of the radiocarbon interhemispheric offset have been restricted to relatively young samples because of a lack of older dendrochronologically secure Southern Hemisphere tree-ring chronologies. The Southern Hemisphere calibration data set SHCal04 earlier than AD 950 utilizes a variable interhemispheric offset derived from measured 2nd millennium AD Southern Hemisphere/Northern Hemisphere sample pairs with the assumption of stable Holocene ocean/atmosphere interactions. This study extends the range of measured interhemispheric offset values with 20 decadal New Zealand kauri and Irish oak sample pairs from 3 selected time intervals in the 1st millennium AD and is part of a larger program to obtain high-precision Southern Hemisphere ^{14}C data continuously back to 200 BC. We found an average interhemispheric offset of 35 ± 6 yr, which although consistent with previously published 2nd millennium AD measurements, is lower than the offset of 55–58 yr utilized in SHCal04. We concur with McCormac et al. (2008) that the IntCal04 measurement for AD 775 may indeed be slightly too old but also suggest the McCormac results appear excessively young for the interval AD 755–785. In addition, we raise the issue of laboratory bias and calibration errors, and encourage all laboratories to check their consistency with appropriate calibration curves and invest more effort into improving the accuracy of those curves.

INTRODUCTION

Tree rings formed at the same time but in opposite hemispheres give different radiocarbon dates, with Southern Hemisphere samples being older by a few decades. This is thought to be due to the larger expanse of the Southern Hemisphere oceans and slightly higher wind speeds resulting in more ^{14}C-depleted CO_2 from the ocean entering the southern atmosphere than the northern (Cain and Suess 1976). This difference has been referred to as the interhemispheric offset.

Most previous measurements of this offset were derived from paired tree-ring samples from the 2 hemispheres, with mean interhemispheric offset values ranging from 23–41 yr (Table 1). This approach was necessarily confined to relatively young samples because of a lack of older dendrochronologically robust Southern Hemisphere data sets.

Table 1 Previously published Southern Hemisphere offset values.

Authors	Geographic location of sample pairs Southern Hemi./Northern Hemi.	Cal age of samples	Southern Hemi.–Northern Hemi. (^{14}C yr)
Lerman et al. 1970	Northern Patagonia/Europe-North America	~AD 1835	36 ± 8
Vogel et al. 1986	South Africa/Netherlands	AD 1840–1890	36 ± 7
Vogel et al. 1993	South Africa/Netherlands	AD 1835–1900	41 ± 5
McCormac et al. 1998	New Zealand/United Kingdom	AD 1725–1885	27 ± 5
Stuiver and Braziunas 1998	Chile/Western United States	19th century	23 ± 4
Hogg et al. 2002	New Zealand/United Kingdom	AD 950–1850	40 ± 13

[1]Radiocarbon Laboratory, University of Waikato, PB 3105, Hamilton 3240, New Zealand. Corresponding author. Email: alan.hogg@waikato.ac.nz.
[2]Gondwana Tree-Ring Laboratory, P.O. Box 14, Little River, Canterbury 7546, New Zealand.
[3]School of Environment, University of Auckland, PB 92019, Auckland, New Zealand.
[4]Centre for Climate, the Environment & Chronology (14CHRONO), School of Geography, Archaeology and Palaeoecology, Queen's University Belfast, Belfast, BT7 1NN, United Kingdom.

Some studies wiggle-matched older floating Southern Hemisphere tree-ring data sets against Northern Hemisphere curves, and did not find an interhemispheric offset (e.g. Barbetti et al. 1992, 1995; Sparks et al. 1995). However, Hogg et al. (2009) analyzed 9 floating Southern Hemisphere data sets using the Bayesian calibration program OxCal 4.1 (Bronk Ramsey 1995, 2001) by wiggle-matching them against the Northern Hemisphere calibration curve IntCal04 (Reimer et al. 2004), utilizing the reservoir offset function (Bronk Ramsey 2009) to allow for an offset. They found that 5 data sets were capable of providing reliable offset information, yielding an interhemispheric difference of approximately 50 yr between 2–10 kyr BP. The remaining data sets were not sufficiently precise to identify real geographic location differences in atmospheric $\Delta^{14}C$ of small magnitude. This finding reinforces the view of McCormac et al. (1998) who considered the interhemispheric offset could be determined accurately only by measuring $\Delta^{14}C$ in contemporaneous Southern Hemisphere and Northern Hemisphere sample pairs within a single laboratory.

The Southern Hemisphere calibration curve SHCal04 (McCormac et al. 2004) extends beyond the range of measured Southern Hemisphere data to 11.0 cal kyr BP, by using Northern Hemisphere data with an offset (55–58 yr) permitted to vary slowly over time, within the constraints of that observed from AD 950–1900.

The principle focus of this paper is to increase the range of ^{14}C measurements of Southern and Northern Hemisphere sample pairs into the 1st millennium AD to reduce uncertainties associated with Holocene interhemispheric offset levels. It is the first stage of a larger program to extend the high precision Southern Hemisphere ^{14}C data set from AD 950 to 200 BC.

METHODS

Experimental Design

We followed the approach of McCormac et al. (1998) and minimized laboratory bias by measuring $\Delta^{14}C$ in contemporaneous Southern Hemisphere and Northern Hemisphere sample pairs. This research focuses upon three 1st millennium AD time intervals, characterized by strong inflections in the Northern Hemisphere data set; AD 245–335 (10 decadal samples), AD 745–785 (5 decadal samples) and AD 895–935 (5 decadal samples). Remeasurement of Northern Hemisphere samples not only permits calculation of the offset but also adds to the Northern Hemisphere calibration data sets, and provides an element of quality assurance.

Dendrochronology

It is important that ^{14}C calibration studies utilize, wherever possible, wood from tree-ring chronologies that are well replicated and securely dated. Some previous studies have used single, non-cross-dated trees (e.g. Sparks et al. 1995), which can contain either missing and/or false rings (Fritts 1976).

Southern Hemisphere samples for this study have been derived from kauri (*Agathis australis*) securely cross-dated by the recently constructed New Zealand kauri chronology (Boswijk et al. 2006). The trees used for the measurements were obtained from Okapakapa Swamp in Northland; geographic coordinates are given in Table 2.

We obtained Irish oak (*Quercus petraea*) samples from the long-established Irish oak chronology (Pilcher et al. 1984) for the Northern Hemisphere measurements. Trees were obtained from various counties in Ireland. Details of site locations for the various time periods are also given in Table 2.

Table 2 Location and time intervals of samples analyzed in this research.

Species and location	Cal yr interval (midpoint of decade)	Dendro tree ID	Geographic location (latitude/longitude)
New Zealand Kauri	AD 245–335	Tree Har005	36°00′S/173°50′E
Hardings Farm, Okapakapa	10 decadal samples		
swamp, North Kaipara	AD 745–785	Tree Har010	
Peninsula, New Zealand	5 decadal samples		
	AD 895–935	Tree Har010	
	5 decadal samples		
Irish oak	AD 245–335	Tree Q9887	54°33′N/06°17′W
Ballinderry, Co. Antrim,	10 decadal samples		
Northern Ireland			
Irish oak	AD 745–785	Trees Q3691	52°40′N/06°17′W
Brabstown, Co. Kilkenny,	5 decadal samples	and Q3693	
Ireland			
Irish oak	AD 895–935	Tree Q9846	54°33′N/07°24′W
Ballinderry, Co. Antrim,	5 decadal samples		
Northern Ireland			

Wood Pretreatment

If wood $\Delta^{14}C$ values are to accurately reflect archived atmospheric ^{14}C levels, they should not be influenced by species differences or variable lignin fractions (McCormac et al. 1998). All samples in this study have been pretreated to α-cellulose, representing approximately 40–50% by weight of total wood components. The process involves grinding wood to pass a 20-mesh sieve, 3-stage refluxing using Soxhlet apparatus (chloroform/ethanol; ethanol; distilled water), bleaching with acidified $NaClO_2$, NaOH extraction and finally treatment with HCl (see Hoper et al. 1998 for full details). It should be noted that high-precision radiometric dating by liquid scintillation counting requires large sample sizes (initial weight of ~80 g per decadal sample) and pretreatments may therefore need to be more intensive than pretreatments routinely applied to comparatively small, high surface area, AMS samples.

Radiocarbon Analysis

^{14}C activities were determined by liquid scintillation counting (LSC) of benzene using PerkinElmer Wallac 1220 Quantulus spectrometers. Hogg et al. (2007) detailed the instrumental and laboratory procedures to optimize the LSC method for high-precision dating. These include:

- Utilization of a sample processing blank to accurately assess background levels.
- Generation of sample benzene in batches confined to wood of similar age to minimize the effects of sample cross-contamination.
- ^{14}C measurement in liquid scintillation spectrometers with high sensitivity and low and stable backgrounds (e.g. PerkinElmer Wallac 1220 Quantulus).
- Use of low-activity liquid scintillation vials with good physical properties (e.g. Waikato 10-mL synthetic silica liquid scintillation vials, Hogg 1993).
- High benzene weights for improved sensitivity (e.g. 7.5 g).
- Long counting times to reduce standard errors (e.g. 6–7 k min per sample).

The precision of each decadal measurement is approximately ±20 yr. A laboratory error multiplier of 1.2, derived from replicate measurements of the ANU sucrose standard, has been applied to all results.

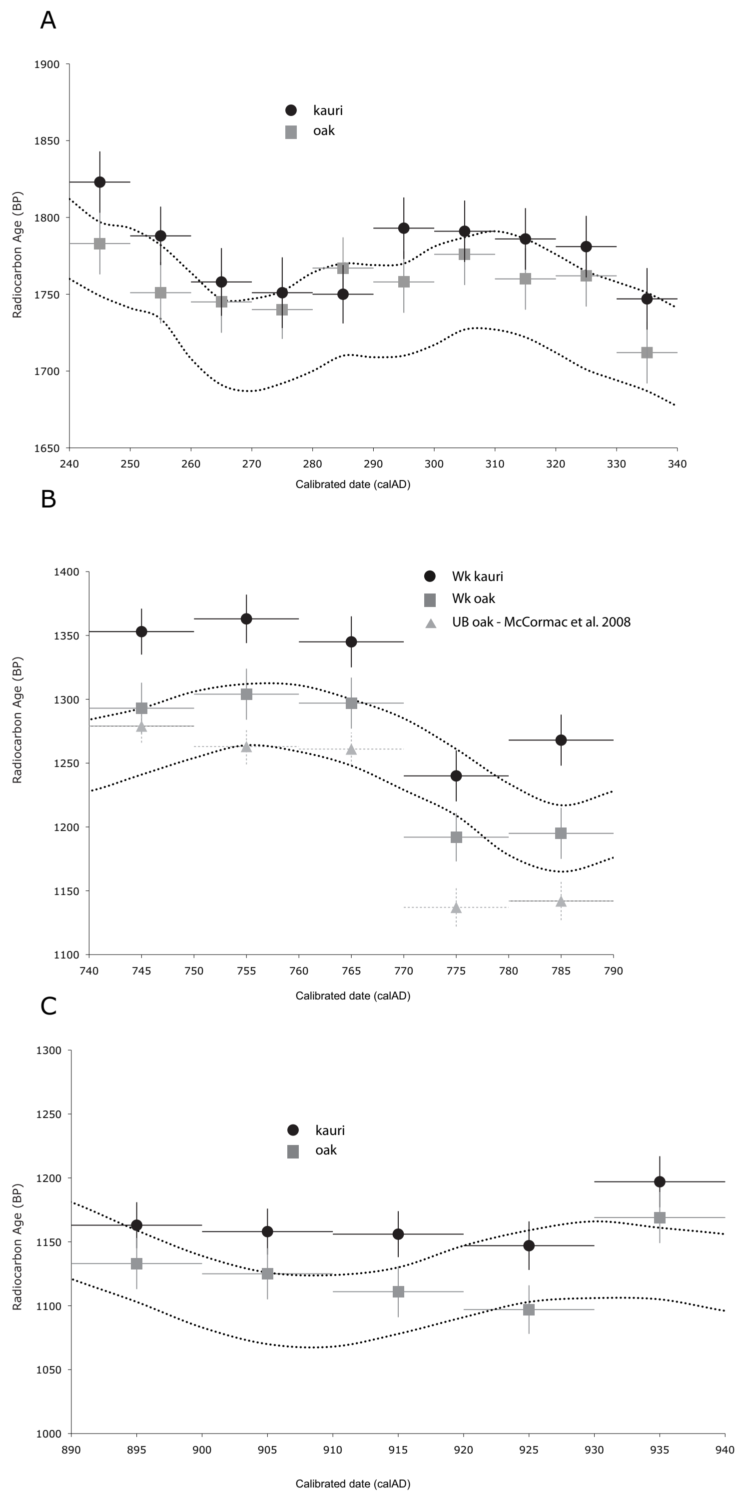


Figure 1 ^{14}C measurements (with 1-σ errors) from dendrochronologically dated wood from the Southern Hemisphere (New Zealand kauri) and Northern Hemisphere (Irish oak); overlain by the 95% probability envelope of IntCal04 (Reimer et al. 2004): A) AD 245–335; B) AD 745–785 (McCormac et al. 2008 Irish oak data added for comparison); C) AD 895–935.

We used ancient (>140 kyr) kauri with no original ^{14}C as a background blank to assess contamination either not removed by sample pretreatment, or added during α-cellulose extraction or benzene synthesis (Hogg et al. 2006). The ancient wood blank standards in this study were subjected to exactly the same pretreatment processes and benzene synthesis procedures as the unknown-age samples. The background blank level is 0.071 ± 0.005 pMC, equivalent to an apparent age of 58.2 ^{14}C kyr (Hogg et al. 2006).

RESULTS AND DISCUSSION

The results of the paired measurements on kauri and oak for the intervals AD 245–335, 745–785, and 895–935 are shown in Figure 1 (A, B, and C, respectively) and Table 3. The offsets between the kauri and oak are shown in Figure 2, with the error-weighted mean differences for the 3 intervals given in Table 4.

Table 3 ^{14}C measurements (yr BP) on decadal samples of kauri and oak and interhemispheric difference.

Yr AD	Lab ID	kauri	$\delta^{13}C$ (%)	Lab ID	oak	$\delta^{13}C$ (%)	kauri-oak
245	Wk-20327	1823 ± 20	–22.2	Wk-22429	1783 ± 20	–24.9	40 ± 28
255	Wk-20328	1788 ± 19	–22.1	Wk-22430	1751 ± 20	–24.8	37 ± 28
265	Wk-20329	1758 ± 22	–21.8	Wk-22431	1745 ± 20	–24.7	13 ± 30
275	Wk-20330	1751 ± 23	–21.3	Wk-22432	1740 ± 19	–25.2	11 ± 30
285	Wk-20331	1750 ± 19	–21.6	Wk-22433	1767 ± 20	–23.8	–17 ± 28
295	Wk-20332	1793 ± 20	–20.9	Wk-22434	1758 ± 20	–23.8	35 ± 28
305	Wk-20373	1791 ± 20	–21.5	Wk-22435	1776 ± 20	–24.1	15 ± 28
315	Wk-20374	1786 ± 20	–22.4	Wk-22436	1760 ± 20	–24.7	26 ± 28
325	Wk-20375	1781 ± 20	–22.3	Wk-22437	1762 ± 20	–24.6	19 ± 28
335	Wk-20376	1747 ± 20	–21.9	Wk-22438	1712 ± 20	–24.7	35 ± 28
745	Wk-22108	1353 ± 18	–22.5	Wk-22424	1293 ± 20	–24.8	60 ± 27
755	Wk-22109	1363 ± 19	–22.5	Wk-22425	1304 ± 20	–24.7	59 ± 28
765	Wk-22110	1345 ± 20	–22.5	Wk-22426	1297 ± 20	–24.2	48 ± 28
775	Wk-22111	1240 ± 20	–22.4	Wk-22427	1192 ± 20	–25.0	48 ± 28
785	Wk-22112	1268 ± 20	–22.0	Wk-22428	1195 ± 20	–24.7	73 ± 28
895	Wk-22165	1163 ± 18	–21.6	Wk-21167	1133 ± 18	–24.7	30 ± 25
905	Wk-22166	1158 ± 18	–22.2	Wk-21168	1125 ± 18	–24.4	33 ± 25
915	Wk-22167	1156 ± 18	–21.1	Wk-21169	1111 ± 18	–24.4	45 ± 25
925	Wk-22168	1147 ± 19	–21.3	Wk-21170	1097 ± 20	–24.6	50 ± 28
935	Wk-22169	1197 ± 20	–21.4	Wk-22445	1169 ± 20	–24.3	28 ± 28

The results show an average hemispheric offset for all measurements spanning the 3 time intervals, of 35 ± 6 yr. This value is consistent with the average offset of 40 ± 13 yr reported for the interval AD 950–1850 by Hogg et al. (2002). Temporal variation is apparent in the offset values, which range from 21 ± 9 to 58 ± 12 yr (Table 4)—χ^2 test: $df = 1$, $t = 5.55$ (5% 3.84).

Although the χ^2 test indicates individual Waikato (Wk) oak measurements are statistically consistent with decadal averages from IntCal04 (Reimer et al. 2004), our oak data are on average, slightly older (Figure 1). The error weighted mean differences from IntCal04 for the 3 intervals are shown in Table 4, and range from 13.7 yr for AD 245–335, 5.3 yr for AD 745–785, and 7.9 yr for AD 895–935 with an average for all 20 measurements of 10 ± 5 yr. For these time intervals, IntCal04 is constructed from 2 raw data sets: University of Washington (QL) decadal tree-ring data (Stuiver et

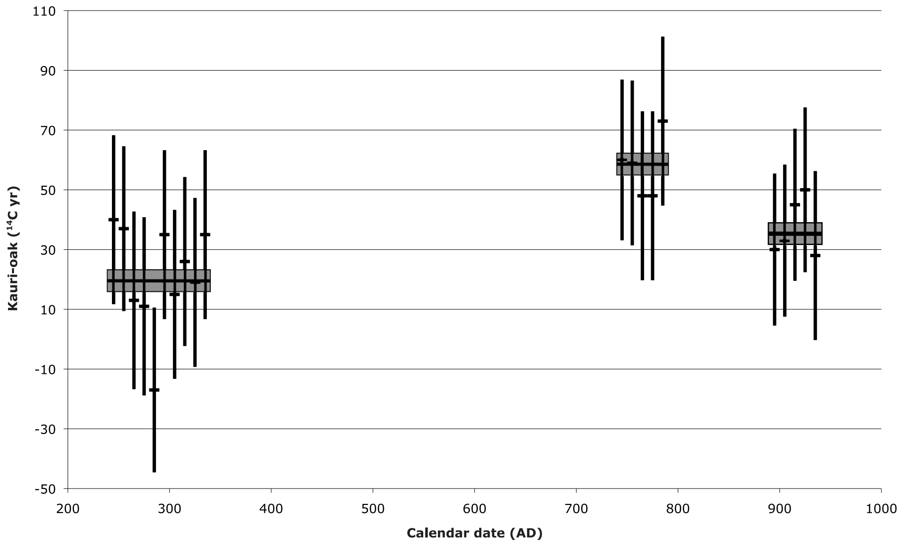


Figure 2 Hemispheric differences in ^{14}C years (with 1-σ errors) between individual decadal measurements of kauri from New Zealand (~36°S) and oak from Ireland (~54°N), for selected time intervals from AD 240–940. The gray band represents the mean difference ±1-σ errors.

Table 4 Kauri minus oak and Waikato offsets for selected periods (^{14}C yr).

Interval	AD 245–335	AD 745–785	AD 895–935
Kauri–oak	21.5 ± 9.0	57.7 ± 12.5	37.2 ± 11.8
Wk oak–IntCal04	13.7 ± 7.8	5.3 ± 10.6	7.9 ± 10.4
Wk oak–QL[a]	9.8 ± 8.5	4.8 ± 10.9	–2.9 ± 12.3
Wk oak–UB[a]	28.0 ± 12.5	9.5 ± 18.4	7.5 ± 16.4

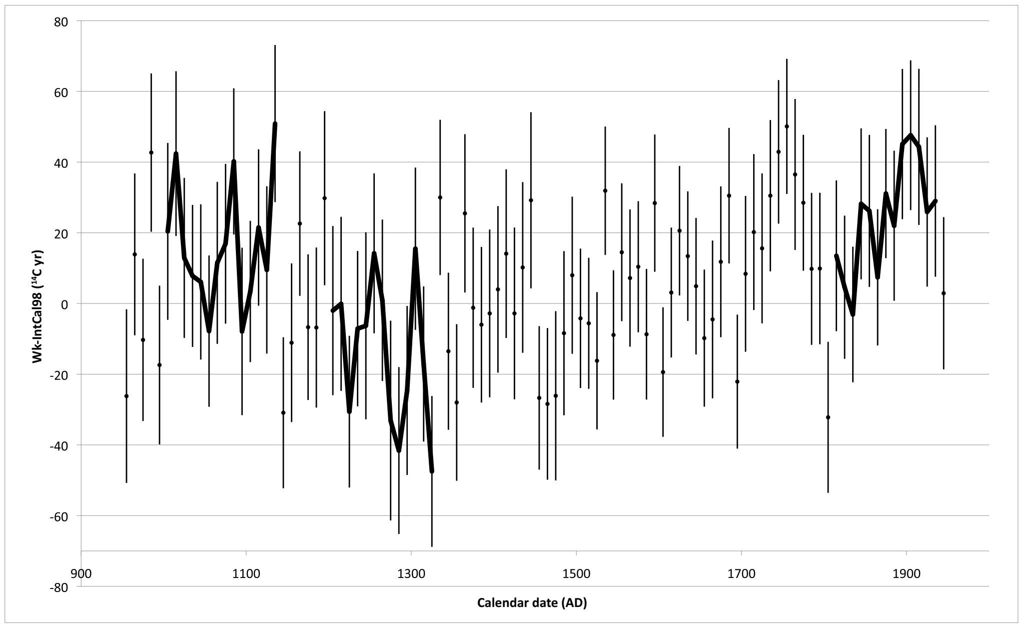
[a]QL and UB data sets obtained from the IntCal04 supplementary files (see http://www.radiocarbon.org/).

al. 1998b) and Queen's University Belfast (UB) bidecadal tree-ring data (Pearson et al. 1986). The error weighted mean differences from the QL and UB subsets for the 3 intervals are also shown in Table 4, with averages for all 20 measurements of 5 ± 6 yr (Wk-QL) and 18 ± 9 yr (Wk-UB). Our data is therefore more closely aligned with the Quaternary Isotope Lab (QL) component of IntCal04 than the Queen's University Belfast (UB) component.

McCormac et al. (2008) reported ^{14}C ages significantly younger than IntCal98 (Stuiver et al. 1998a) for the interval AD 775–785 (their data shown in Figure 1B and Table 6). Comparison of our oak data with that given in McCormac et al. (2008) shows 2 decades (AD 775–785) lacking consistency, with the Wk results in closer agreement with IntCal04, suggesting the McCormac et al. (2008) data may be too young. Indeed for the interval AD 755–785, the McCormac data is uniformly younger than IntCal04, with an error weighted mean offset of –43 ± 9 yr (Table 6).

The maximum offset for the Waikato data against IntCal04 is 13.7 yr (over a 100-yr interval, AD 245–335). In order to determine how common an offset of this magnitude is, we investigated typical offsets within IntCal04, by comparing the 2nd millennium AD Waikato and Queen's University Belfast component (given in Hogg et al. 2002) with the QL and UB data component (i.e. data in IntCal98, given in Stuiver et al. 1998a). To avoid circularity, the IntCal98 data set was used rather than IntCal04, which includes the Hogg et al. (2002) measurements. From AD 1945–955, the Wk

A



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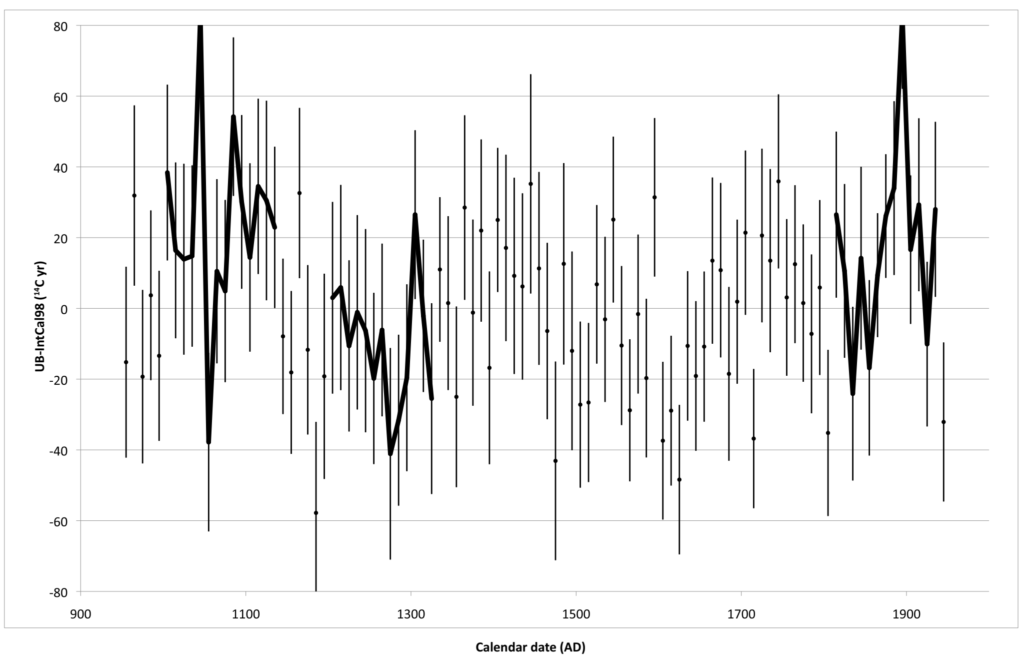

Figure 3 Difference between individual decadal measurements of A: Wk oak–IntCal98 and B: UB oak–IntCal98 calibration data (Stuiver et al. 1998a) for the time interval AD 1950–950 (data from Hogg et al. 2002). Black highlighting indicates periods where both Wk and UB data sets are systematically offset from IntCal98—see text for details).

Table 5 Difference in ^{14}C ages between oak data reported by Hogg et al. (2002) (Wk and UB data sets) and IntCal98 for 3 selected time intervals (^{14}C yr).

	AD 1005–1135 (140 yr)	AD 1205–1325 (130 yr)	AD 1815–1935 (130 yr)
Wk oak–IntCal98	16.1 ± 5.9	−14 ± 6.5	23.9 ± 5.7
UB oak–IntCal98	22.8 ± 25.1	−9.2 ± 7.1	18.9 ± 6.2

Table 6 Comparison of Wk and UB oak measurements (McCormac et al. 2008) for AD 745–785 (^{14}C yr).

Decade (mid-pt)	IntCal04	Wk	UB	χ^2 test (Wk/UB)	Wk-IntCal04	UB-IntCal04
AD 745	1267 ± 13	1293 ± 20	1279 ± 13	t = 0.34 (5% 3.84)	26 ± 24	12 ± 18
AD 755	1288 ± 12	1304 ± 20	1263 ± 14	t = 2.82 (5% 3.84)	16 ± 23	−25 ± 18
AD 765	1274 ± 13	1297 ± 20	1261 ± 13	t = 2.28 (5% 3.84)	23 ± 24	−13 ± 18
AD 775	1235 ± 13	1192 ± 20	1137 ± 15	t = 4.84 (5% 3.84)	−43 ± 24	−98 ± 20
AD 785	1191 ± 13	1195 ± 20	1142 ± 15	t = 4.49 (5% 3.84)	4 ± 24	−49 ± 20

and UB data is on average 7 ± 2 and 2 ± 2 yr, respectively, older than IntCal98. Although agreement between the 3 data sets is high over the 1000-yr interval, a more detailed analysis reveals significant, possibly systematic, shifts of up to 24 yr towards either older or younger ages, for time periods of up to 140 yr. Three such examples are shown in Figure 3 and Table 5. The Wk and UB data sets show consistent divergence from IntCal98 for these periods. This suggests offsets, such as the Waikato mean offset to IntCal04 of 10 ± 5 yr found in this research may be relatively common, with further analyses required to determine if they are systematic in nature.

The possibility of systematic offsets in IntCal98 and the Wk/IntCal04 and UB/IntCal04 offsets referred to above, highlights the potential issue of laboratory offsets to calibration curves and resulting calibration errors. International intercalibration exercises have generally focused upon pretreatment issues from variable sample types and ages, with less emphasis on calibration wood samples. As IntCal04 and SHCal04 are extensively used to obtain calibrated ages for atmospheric samples, we think intercalibration exercises should now focus more closely on sequences of calibration samples to quantify calibration errors associated with laboratory offsets. A batch of 10 successive decadal calibration wood samples for example, would not only provide a laboratory with information on their alignment with the calibration curve for that time interval, but would also provide reproducibility data and the effectiveness of their wood pretreatment procedures. Whilst the onus lies with individual laboratories to demonstrate alignment with the calibration curves, it is the responsibility of the international radiocarbon community to ensure those curves are as accurate as possible. The international research funding bodies should therefore be encouraged to support efforts to obtain supplementary calibration data to improve the reliability of existing calibration data sets.

CONCLUSIONS

Twenty pairs of high precision $\Delta^{14}C$ measurements on decadal samples of New Zealand kauri and Irish oak from 3 time intervals in the 1st millennium AD show an average hemispheric offset of 35 ± 6 yr. This value is consistent with measurements from the 2nd millennium AD of 40 ± 13 yr (Hogg et al. 2002) but is lower than the interhemispheric offset range of 55–58 yr used in the extension of SHCal04 beyond measured values (i.e. beyond AD 950). As this study is restricted to a mere 20% of the 1st millennium AD and the 55–58 yr offset was determined from the more extensive 950-yr 2nd millennium AD record, we do not believe a reduction in the offset is justified at this stage.

Extension of the Southern Hemisphere high precision data set from AD 950 to 200 BC in the second stage of this research program will indicate how appropriate a SHCal04 offset of 55–58 yr is for this time interval.

McCormac et al. (2008) reported anomalously young ^{14}C ages for 2 decadal samples, with midpoints AD 775 and 785. Our findings suggest that while IntCal04 may indeed be slightly too old for the first of these decades (AD 775), the McCormac results appear excessively young for both, with their data for the interval AD 755–785 offset by an average of –43 ± 9 yr.

The possibility of laboratory offsets to calibration curves with resulting calibration errors, should encourage all laboratories to check their alignment using sequences of calibration samples and the international carbon dating community to invest more effort into obtaining new calibration data to minimize the possibility of these curves containing systematic offsets.

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