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IN THIS ISSUE

The Radiocarbon Dating and Authentication of Iron Artifacts pp 717-732

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Cover image is an illustration from Biringuccio's *Pirotechnia* of 1540, showing an old tree bole being converted into charcoal for use as a metallurgical fuel (see page 720).

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FROM THE EDITOR

This volume is a little late in getting to you, but I trust it will be just as interesting. Our issue has a number of articles related to calibration. The first, by Hogg et al., studied differences in contemporaneous ^{14}C in tree rings from the Northern and Southern Hemispheres. We also have some more calibration work by McCormac and colleagues, as well as a preliminary report about INTCAL discussions. A refinement to the international radiocarbon calibration (INTCAL) will be discussed at the 18th International Radiocarbon Conference in New Zealand this coming September. We also have a paper about radiocarbon calibration software by Jones & Nichols, adding to the several computer programs already in existence for calibration.

Applications papers focus on studies of natural levels of carbon isotopes in lichens, radiocarbon dating of an interesting religious artifact from an Italian radiocarbon group, artifacts from Palau by Fitzpatrick and Boyle, and the hair of animals in the Altiplano of South America. To this diverse list, we can also add a summary of ideas about the radiocarbon dating of iron artifacts (Craddock et al.), archaeological studies in South America and discussion of isotope corrections. For discussion, Manning et al. take up the many issues raised by an earlier article by Keenan in this journal.

I hope everyone finds this issue as interesting as most, and I am sure many are anticipating the upcoming 18th International Radiocarbon Conference in New Zealand.

Best wishes,

A J Timothy Jull

Editor

QUATERNARY COMMUNITY LOSES GLENN GOODFRIEND

As scientists we each enjoy the genuine sense of discovery that comes from our daily work on earth systems and earth history. For most of us, our love of fieldwork is complemented by selfless hours of laboratory work and the camaraderie and inspiration that comes from collaborations with colleagues. The outgrowths of this process are the enduring publications that pass on our knowledge to others. On October 15, 2002, just before the national Geological Society of America annual meeting he rarely missed, Glenn A Goodfriend passed away prematurely at the age of 51—he was one who had so much more to discover and pass on. Following serious health problems that apparently developed late last spring, he died at George Washington University Hospital of pneumonia and serious complications from rhinocerebral mucormycosis, a systemic fungal disease.

Glenn was a scientist with many talents across the fields of Quaternary stratigraphy, paleontology, zoology, biogeochemistry, geochronology, and paleoclimate. He also had interests in geoarcheology and was most widely known for his specialties in land snail ecology and amino acid racemization studies.

Much like Louis Agassiz, Glenn came to geology via zoology. As a native of New Rochelle, New York, Glenn earned his undergraduate degree in zoology from the University of Rhode Island in 1973. He later earned a Masters degree in Evolutionary Biology from University of Chicago in 1978 and a PhD in zoology from the University of Florida in 1983. From 1983 to 1988 he worked as a post-doctoral fellow and research scientist in the Isotope Department at the Weizmann Institute of Science in Israel, a top-ranking multidisciplinary research institution. His position there changed to that of senior scientist in the then, renamed, Department of Environmental Sciences & Energy Research. After returning to the USA, he became a Senior Research Associate in the Geophysical Laboratory of the Carnegie Institution of Washington from 1990 to 1998 while also serving from 1993–1995 as an Adjunct faculty member at Johns Hopkins in the Environmental Earth Sciences and Policy Program. In 1998, Glenn then joined the Department of Earth and Environmental Sciences at George Washington University as a Research Professor until his death.

Glenn had published more than 40 scientific papers, not counting the ones he likely left unfinished on his computer due to health problems in recent months. As a broad thinker in his work, he had collaborations with scientists across a range of subdisciplines from taphonomy and paleontology to biogeochemistry. He is pictured below with Steven Jay Gould just a month or so before Gould died last year.

Glenn Goodfriend (at left) with Steven Jay Gould about one month before Gould died last year (photo courtesy of Bob Hazen, Carnegie Institute).

Glenn most recently was the lead editor of a major volume on *Perspectives in Amino Acid and Protein Geochemistry* (Goodfriend GA, Collins MJ, Fogel ML, Macko SA, Wehmiller JF, editors. 2000. New York: Oxford University Press). At his untimely death, he had nearly $400,000 in funded grants from the National Science Foundation including work on:

- "Geochronology and paleoecology of eggshells in central Africa in the context of human evolution"
- "Age mixing and taphonomy in Holocene shelf deposits, coastal Brazil"
- "Evolution and ecology of Cerion, a land snail from the Bahamas" (in collaboration with Steven Jay Gould)
- "Measurement of absorbed amino acids on mineral surfaces and the selectivity of absorption of amino-acid enantiomers".

According to John Wehmiller, University of Delaware, the current plans are for several of Glenn's colleagues, including John, to carry on Glenn's work and finish out the research he had started, something everyone is sure Glenn would have wanted. What I will remember most about Glenn was his gift of sharing and his attention to scientific detail. He allowed an eager senior honors student and I from the University of Massachusetts to pick his brain about lab procedures for a few days in his Carnegie lab. He then took a few more days out of his industrious schedule to visit me at the University of Massachusetts months later to follow up and help me troubleshoot a new lab instrument.

One of Glenn's favorite hobbies included the tasting of fine wines. Having been more interested in quantity rather than quality most of my life (a constant joke I had with him), I will never forget the night when Glenn, as a house guest, presented my husband and I with a bottle of wine so classy that you actually had to filter it before you drank it. On behalf of all of his many colleagues, let me summarize by saying that Glenn and his scientific creativity and contributions will be sorely missed. Like a fine wine, he is gone too soon.

Glenn is survived by his father and stepmother, Morton and Teresa Goodfriend of Huntley, Ill.

Julie Brigham-Grette

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Department of Geosciences
University of Massachusetts

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HIGH-PRECISION RADIOCARBON MEASUREMENTS OF CONTEMPORANEOUS TREE-RING DATED WOOD FROM THE BRITISH ISLES AND NEW ZEALAND: AD 1850–950

A G Hogg[1] • F G McCormac[2] • T F G Higham[3] • P J Reimer[4] • M G L Baillie[2] • J G Palmer[2]

ABSTRACT. The University of Waikato, Hamilton, New Zealand and The Queen's University of Belfast, Northern Ireland radiocarbon dating laboratories have undertaken a series of high-precision measurements on decadal samples of dendrochronologically dated oak (*Quercus petraea*) from Great Britain and cedar (*Libocedrus bidwillii*) and silver pine (*Lagarostrobos colensoi*) from New Zealand. The results show an average hemispheric offset over the 900 yr of measurement of 40 ± 13 yr. This value is not constant but varies with a periodicity of about 130 yr. The Northern Hemisphere measurements confirm the validity of the Pearson et al. (1986) calibration dataset.

INTRODUCTION

The radiocarbon ages of decadal (10 yr) samples of dendrochronologically dated wood from both hemispheres spanning the AD 1850–950 interval have been measured to high-precision in the Waikato and Belfast ^{14}C laboratories. The decadal blocks of dendrochronologically dated oak (*Quercus petraea*) from the British Isles and New Zealand cedar (*Libocedrus bidwillii*) and silver pine (*Lagarostrobos colensoi)* from New Zealand were used to provide comparative measurements of the ^{14}C content of the atmosphere in both hemispheres over a 900-yr period. This data also forms the basis for a companion paper which recommends its use for calibration of Southern Hemisphere ^{14}C measurements (McCormac et al., this issue). The blocks of wood were pretreated to α-cellulose (Hoper et al. 1997) thereby removing all mobile fractions, and the ^{14}C dates for each were determined by liquid scintillation counting of benzene (Hogg et al. 1987; McCormac 1992; McCormac et al. 1993; Higham and Hogg 1997). Given that the difference in Δ^{14}C between the hemispheres was expected to be small, it was felt necessary, in experimental design, to replicate the oak, cedar, and pine measurements in both laboratories, thereby creating 2 independent measurements of the offset and, thus, negating the effects of individual laboratory bias on the value determined for the interhemispheric offset. The results show a consistent ^{14}C depletion or older ^{14}C ages in the Southern Hemisphere over the period AD 950–1850. The results given here extend the Northern/Southern hemisphere data sets from AD 1940 to 1720 presented in McCormac et al. (1998a, 1998b).

LABORATORY OFFSET

The 900 yr of data at decadal intervals allows us to determine the offset between the Waikato and Belfast laboratories with some accuracy (Table 1). The Waikato and Belfast data sets show excellent agreement with the Belfast–Waikato offset, being –4.5 yr for the British Isles oak series and –3.9 yr for the New Zealand cedar/pine series. This offset compares very favorably with previous studies, which resulted in offsets ranging from 10 to 21 yr (Table 2).

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Table 1 Offsets between Belfast and Waikato measurements for the interval AD 955–1945. σ_1 is the average standard deviation based on quoted laboratory errors and σ_2 is the observed standard deviation in the age difference. The error multiplier k = σ_2/σ_1.

Samples	Offset	s_1	s_2	k	N
Cedar/pine	–3.9 ± 2.5	25.3	23.6	0.9	100
Oak	–4.5 ± 2.6	25.9	24.3	0.9	100

Table 2 Laboratory ^{14}C offsets on identical wood

Laboratories[a]	Cal yr interval	Trees	N	Offset ± σ_{meas} (^{14}C yr)
Belfast–Seattle[a]	BC 7750–5260	German oak	181	10 ± 2
Pretoria/Groningen–Seattle[a]	BC 3910–1930	German oak	194	17 ± 2
Heidelberg–Seattle[a]	BC 7720–4080	German oak	128	21 ± 3
Heidelberg–Seattle[a]	BC 9670–8000	German pine	102	16 ± 4
Belfast–Waikato	AD 955–1945	British Isles oak	50	–4.5 ± 2.6
Belfast–Waikato	AD 955–1945	New Zealand cedar/pine	50	–3.9 ± 2.5

[a] Stuiver et al. 1998. *Radiocarbon* 40(3):1041–83, p 1045

NORTHERN HEMISPHERE MEASUREMENTS

Figure 1 shows the individual measurements from Waikato (Wk) and Belfast (UB) on decadal samples of oak from the British Isles for the period AD 950–1850 (see also Table 3). These measurements essentially repeat, at higher temporal resolution, the Pearson et al. (1986, 1993) data published in the ^{14}C special calibration issues. The evolution of the Pearson data was a result of repeated corrections, which were questioned by McCormac et al. (1995). Thus, the new data offer the opportunity to test which corrections are appropriate in light of changes made to the calibration data published by Stuiver et al. (1998) and commonly known as INTCAL98.

Decadal values of the new oak measurements were combined to form bi-decadal averages centered on the intervals used by Pearson et al. (1986, 1993). The difference between the Pearson et al. (1986) data and the new suite of 72 measurements made in Belfast is –7.5 ± 2.8 yr with the new measurements being slightly older. The Pearson et al. (1993) data, which is a corrected version of the Pearson et al. (1986) measurements, is 7.3 ± 2.7 yr different from the new measurements. Although these results are not conclusive, other evidence supports the consistency of the Pearson et al. (1986) data (van der Plicht and McCormac 1995; van der Plicht et al. 1995).

A separate set of 7 sample pairs analyzed by the Belfast lab covering the period AD 610–730 gave differences between the Pearson et al. (1986, 1993) data of –3.8 ± 7.6 and 18.5 ± 7.3, respectively. In addition, a set of 8 University of Washington measurements of Irish oak from 505 to 655 BC resulted in a 3.6 ± 5.2 yr difference from the Pearson et al. 1986 results and –10.7 ± 5.2 yr difference from the 1993 dataset (Stuiver, personal communication 2002). We would therefore suggest that future calibration data for the Northern Hemisphere incorporates Pearson et al. (1986) data for the intervals beyond the range of data presented here.

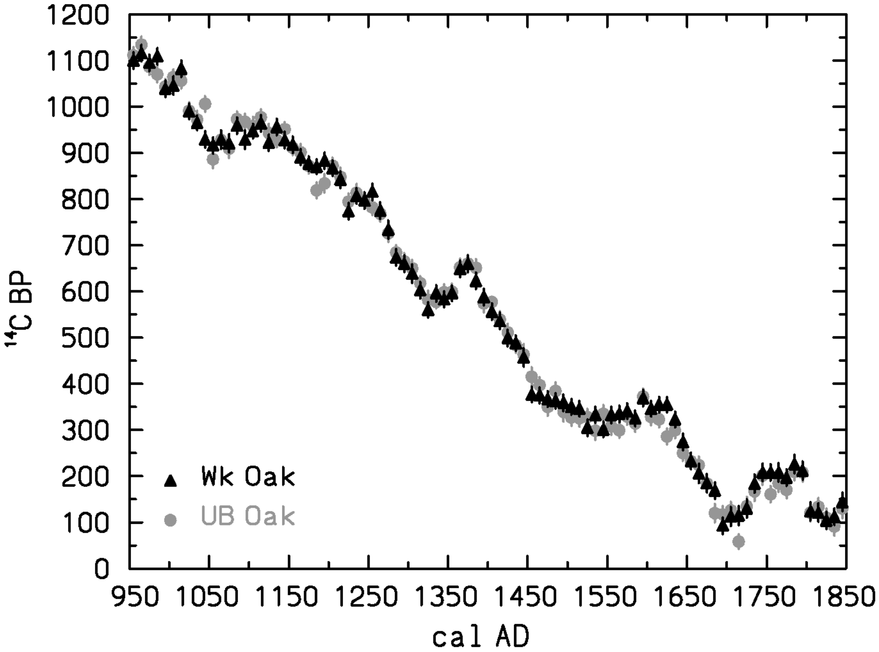

Figure 1 ^{14}C measurements on decadal samples of oak made in Queen's University of Belfast and the University of Waikato, New Zealand (AD 950–1850). AD 1855–1755 (Shane's Castle, N. Ireland); AD 1745–1505 (Sherwood Forest, England); AD 1495–1445 (Hillsborough Fort, N. Ireland); AD 1435–1325 (Toome, N. Ireland); AD 1315–1195 (Blackwater, N. Ireland); AD 1185–995 (Trim Castle, Ireland); AD 985–955 (Ballinderry, N. Ireland)

SOUTHERN HEMISPHERE MEASUREMENTS

Figure 2 shows the individual measurements from Waikato and Belfast on decadal samples of cedar and silver pine covering the period AD 950–1850. These follow closely the temporal variations of the oak measurements (Figure 1) and show again a very high degree of agreement between the measurements made in the 2 laboratories. Because of the limited age range of available cedar, it was necessary to change species to silver pine at AD 1405. The cedar trees grew in 2 sites in the middle of the North Island of New Zealand (rings AD 1401–1720, Takapari Forest Park, 40°04′S, 175°59′E; rings AD 1721–1850, Hihitahi Forest Park, 39°32′S, 175°44′E), while the silver pines grew on the west coast of the South Island (Oroko Swamp, 43°14′S, 170°17′E). Knox and McFadgen (2001) claim there is a statistical difference between the North Island cedar data given in McCormac et al. (1998a) and the South Island matai data given in Sparks et al. (1995) and, furthermore, suggest that the differences might be the result of either geographic location or proximity to the intermittently active volcano, Ruapehu. We consider it unlikely that volcanic emissions have affected the ^{14}C content of the cedar wood (Rubin et al. 1987; Bruns et al. 1980), as Hihitahi Forest Park is 32 km away, and Takapari Forest Park is 94 km away from the volcano. We have checked the consistency in Δ^{14}C between silver pine and Takapari Forest Park cedar by dating 5 wood samples of the same dendrochronological age from both species. The results are shown in Table 4. The weighted mean difference between the 2 species is 9.4 ± 7.6 yr based on 10 sample pairs. Using the student-t test for paired samples, there is no difference between the measurements for cedar and silver pine at the 95% confidence level.

Table 3 Measurements on decadal samples of wood from the British Isles and New Zealand. Uncertainties include both counting statistics and sample preparation. Cedar and pine measurements (C/P) have been averaged over the interval AD 1405–1445. The SH offset is calculated from the average of the difference in the New Zealand and British Isles measurements for each laboratory.

Year AD	Wk C/P (^{14}C BP)	δ^{13}C (‰)	Wk oak (^{14}C BP)	δ^{13}C (‰)	Qub C/P (^{14}C BP)	δ^{13}C (‰)	Qub oak (^{14}C BP)	δ^{13}C (‰)	SH offset (^{14}C yr)
955	1162 ± 18	–21.5	1100 ± 18	–25.8	1191 ± 20	–22.1	1111 ± 18	–25.9	71 ± 19
965	1166 ± 18	–21.9	1116 ± 18	–25.1	1169 ± 19	–21.9	1134 ± 18	–26.0	43 ± 18
975	1157 ± 18	–22.0	1095 ± 18	–25.3	1149 ± 17	–22.3	1086 ± 17	–25.6	63 ± 18
985	1165 ± 18	–21.7	1109 ± 18	–25.6	1119 ± 17	–21.9	1070 ± 17	–26.3	52 ± 18
995	1051 ± 18	–22.1	1039 ± 18	–23.5	1105 ± 19	–22.1	1043 ± 17	–24.3	37 ± 18
1005	1073 ± 20	–21.5	1046 ± 20	–23.2	1090 ± 17	–21.7	1064 ± 17	–24.4	26 ± 19
1015	1098 ± 18	–22.0	1082 ± 18	–22.9	1104 ± 17	–22.3	1056 ± 17	–24.6	33 ± 18
1025	1024 ± 17	–21.7	990 ± 17	–23.3	1072 ± 19	–22.1	991 ± 19	–23.2	55 ± 18
1035	1008 ± 16	–21.9	965 ± 16	–23.3	1018 ± 19	–22.1	972 ± 19	–23.6	44 ± 18
1045	1001 ± 17	–21.9	929 ± 18	–23.4	1017 ± 17	–22.2	1006 ± 17	–23.8	41 ± 17
1055	964 ± 20	–21.9	916 ± 18	–23.9	919 ± 19	–22.4	886 ± 19	–24.4	40 ± 19
1065	971 ± 20	–21.8	929 ± 20	–23.9	983 ± 20	–22.1	928 ± 20	–24.6	49 ± 20
1075	949 ± 20	–21.8	921 ± 20	–23.7	956 ± 20	–22.1	909 ± 20	–23.8	38 ± 20
1085	996 ± 20	–22.2	959 ± 18	–23.9	1001 ± 17	–22.1	973 ± 17	–23.8	32 ± 18
1095	1003 ± 20	–21.9	929 ± 20	–23.8	1019 ± 18	–22.4	967 ± 18	–24.1	62 ± 19
1105	1001 ± 15	–22.1	947 ± 16	–23.3	1000 ± 18	–22.2	958 ± 20	–24.1	49 ± 17
1115	977 ± 18	–21.9	964 ± 18	–23.3	1001 ± 19	–21.9	977 ± 18	–24.1	18 ± 18
1125	982 ± 20	–21.6	922 ± 18	–23.4	1022 ± 18	–21.9	943 ± 20	–23.6	70 ± 19
1135	971 ± 18	–21.5	956 ± 18	–25.0	1008 ± 16	–21.7	928 ± 16	–25.0	51 ± 17
1145	966 ± 18	–21.8	928 ± 18	–25.4	996 ± 16	–22.1	951 ± 16	–25.8	42 ± 17
1155	961 ± 18	–22.0	918 ± 18	–25.5	937 ± 16	–22.8	911 ± 16	–25.5	34 ± 17
1165	927 ± 17	–22.1	891 ± 17	–26.0	906 ± 18	–22.6	901 ± 18	–25.6	21 ± 18
1175	931 ± 16	–22.0	877 ± 16	–25.4	950 ± 17	–22.4	872 ± 17	–25.5	65 ± 17
1185	912 ± 16	–22.0	870 ± 16	–24.1	887 ± 17	–22.2	819 ± 17	–25.0	54 ± 17
1195	930 ± 18	–22.2	883 ± 18	–26.7	867 ± 20	–22.6	834 ± 20	–27.1	41 ± 19
1205	913 ± 17	–22.0	867 ± 17	–26.5	896 ± 18	–22.7	872 ± 18	–26.7	36 ± 18
1215	883 ± 18	–21.9	842 ± 18	–26.7	883 ± 20	–22.4	848 ± 20	–26.6	38 ± 19
1225	843 ± 18	–21.7	774 ± 18	–26.1	846 ± 18	–22.1	794 ± 18	–26.6	61 ± 18
1235	820 ± 17	–21.5	807 ± 17	–26.9	818 ± 20	–22.1	813 ± 20	–26.9	10 ± 19
1245	849 ± 17	–22.0	797 ± 18	–26.3	795 ± 19	–22.4	797 ± 18	–26.8	26 ± 18
1255	848 ± 18	–21.3	816 ± 18	–26.0	838 ± 17	–21.6	782 ± 17	–26.5	45 ± 18
1265	811 ± 18	–21.5	775 ± 18	–26.1	850 ± 17	–21.8	768 ± 17	–26.7	60 ± 18
1275	781 ± 20	–21.7	734 ± 20	–26.3	809 ± 19	–21.7	726 ± 19	–26.2	66 ± 20
1285	755 ± 18	–21.8	674 ± 18	–26.4	753 ± 20	–21.6	684 ± 16	–26.8	75 ± 18
1295	722 ± 18	–21.8	660 ± 18	–26.4	722 ± 18	–21.7	665 ± 18	–26.5	60 ± 18
1305	707 ± 18	–21.8	639 ± 20	–26.0	678 ± 18	–21.6	650 ± 18	–26.2	47 ± 19
1315	693 ± 18	–21.6	603 ± 18	–26.4	695 ± 14	–21.8	618 ± 15	–26.3	82 ± 16
1325	632 ± 18	–21.5	560 ± 17	–26.2	646 ± 18	–22.0	582 ± 20	–26.4	68 ± 18
1335	632 ± 18	–21.1	596 ± 18	–26.1	649 ± 18	–21.4	577 ± 14	–27.0	56 ± 17
1345	658 ± 17	–21.6	583 ± 17	–26.6	638 ± 17	–21.8	598 ± 17	–26.4	58 ± 17
1355	675 ± 17	–21.2	596 ± 17	–26.6	686 ± 17	–21.2	599 ± 18	–26.3	83 ± 17
1365	707 ± 18	–21.3	649 ± 18	–26.6	708 ± 18	–21.2	652 ± 19	–26.5	57 ± 18
1375	710 ± 18	–21.8	660 ± 18	–26.4	722 ± 19	–21.4	660 ± 19	–26.2	56 ± 19
1385	676 ± 18	–21.3	623 ± 18	–26.3	669 ± 19	–21.5	651 ± 19	–26.3	36 ± 19
1395	624 ± 18	–21.4	588 ± 18	–26.4	629 ± 21	–21.9	574 ± 19	–26.4	44 ± 19
1405	592 ± 13	–22.1/–21.3	556 ± 18	–26.0	583 ± 13	–22.2/–21.1	577 ± 12	–25.1	18 ± 14
1415	552 ± 13	–22.1/–21.4	536 ± 18	–25.9	574 ± 12	–22.1/–21.5	539 ± 18	–26.0	26 ± 16

Table 3 Measurements on decadal samples of wood from the British Isles and New Zealand. Uncertainties include both counting statistics and sample preparation. Cedar and pine measurements (C/P) have been averaged over the interval AD 1405–1445. The SH offset is calculated from the average of the difference in the New Zealand and British Isles measurements for each laboratory. (*Continued*)

Year AD	Wk C/P (^{14}C BP)	δ^{13}C (‰)	Wk oak (^{14}C BP)	δ^{13}C (‰)	Qub C/P (^{14}C BP)	δ^{13}C (‰)	Qub oak (^{14}C BP)	δ^{13}C (‰)	SH offset (^{14}C yr)
1425	543 ± 13	–22.0/–21.4	499 ± 18	–25.7	547 ± 13	–22.3/–21.7	511 ± 19	–26.0	40 ± 16
1435	524 ± 13	–22.2/–21.3	488 ± 17	–26.0	526 ± 12	–22.2/–21.6	484 ± 17	–26.2	39 ± 15
1445	492 ± 13	–22.0/–21.1	457 ± 20	–26.6	506 ± 13	–22.0/21.6	463 ± 23	–26.2	38 ± 18
1455	455 ± 17	–21.9	377 ± 17	–26.1	455 ± 17	–23.0	415 ± 21	–25.8	61 ± 18
1465	408 ± 17	–22.0	375 ± 17	–25.7	409 ± 17	–22.2	397 ± 18	–25.8	23 ± 17
1475	435 ± 18	–21.5	367 ± 17	–25.3	424 ± 20	–23.1	350 ± 19	–25.1	71 ± 19
1485	405 ± 17	–21.8	363 ± 17	–25.0	402 ± 17	–22.3	384 ± 20	–25.4	31 ± 18
1495	401 ± 18	–22.1	359 ± 18	–25.4	417 ± 18	–22.7	339 ± 21	–25.6	59 ± 19
1505	380 ± 17	–22.0	350 ± 17	–24.6	374 ± 18	–22.3	327 ± 18	–24.7	38 ± 18
1515	338 ± 17	–21.9	346 ± 17	–25.0	349 ± 18	–22.1	325 ± 18	–24.9	7 ± 18
1525	352 ± 17	–21.7	305 ± 18	–24.9	336 ± 18	–22.1	328 ± 18	–25.3	28 ± 18
1535	362 ± 17	–21.5	333 ± 17	–24.8	317 ± 19	–22.1	298 ± 19	–24.8	25 ± 18
1545	359 ± 18	–21.5	301 ± 17	–24.6	344 ± 19	–22.4	335 ± 19	–25.3	36 ± 18
1555	361 ± 18	–21.4	333 ± 18	–24.8	368 ± 18	–21.6	308 ± 18	–25.4	44 ± 18
1565	348 ± 18	–21.0	335 ± 18	–24.8	355 ± 16	–21.1	299 ± 16	–25.1	37 ± 17
1575	381 ± 17	–20.8	341 ± 17	–25.0	335 ± 18	–21.0	329 ± 18	–25.2	24 ± 18
1585	380 ± 17	–20.8	325 ± 17	–24.5	344 ± 19	–20.8	314 ± 18	–24.9	44 ± 18
1595	397 ± 17	–20.7	369 ± 18	–24.7	377 ± 19	–21.1	372 ± 18	–24.5	17 ± 18
1605	414 ± 17	–20.5	346 ± 17	–24.8	391 ± 18	–21.1	328 ± 18	–24.9	66 ± 18
1615	391 ± 17	–20.8	355 ± 17	–24.9	378 ± 18	–21.0	323 ± 17	–25.1	45 ± 17
1625	369 ± 17	–20.7	355 ± 17	–25.3	323 ± 17	–21.1	286 ± 17	–25.3	26 ± 17
1635	350 ± 17	–20.5	323 ± 17	–25.1	303 ± 17	–20.8	299 ± 17	–25.2	16 ± 17
1645	311 ± 18	–20.8	274 ± 18	–24.9	284 ± 17	–20.8	250 ± 17	–25.6	35 ± 18
1655	260 ± 18	–20.8	233 ± 18	–24.8	238 ± 16	–21.0	232 ± 17	–25.3	16 ± 17
1665	237 ± 18	–20.2	206 ± 21	–24.7	261 ± 16	–20.6	224 ± 19	–24.9	34 ± 19
1675	212 ± 18	–20.0	185 ± 20	–24.6	205 ± 17	–20.7	184 ± 20	–25.7	24 ± 19
1685	198 ± 18	–20.0	169 ± 18	–25.0	188 ± 21	–20.4	120 ± 20	–25.7	46 ± 19
1695	147 ± 18	–20.1	93 ± 18	–25.7	167 ± 20	–20.8	117 ± 19	–25.8	52 ± 19
1705	155 ± 21	–19.9	113 ± 21	–24.9	136 ± 20	–20.4	126 ± 19	–25.4	25 ± 20
1715	122 ± 17	–20.5	115 ± 21	–24.6	129 ± 16	–21.0	58 ± 16	–25.1	45 ± 18
1725	162 ± 20	–19.7	130 ± 20	–24.5	135 ± 19	–20.7	135 ± 20	–25.2	16 ± 20
1735	213 ± 19	–19.7	184 ± 20	–24.5	194 ± 20	–20.2	167 ± 21	–25.8	28 ± 20
1745	232 ± 19	–19.8	208 ± 19	–24.5	224 ± 19	–20.4	201 ± 20	–25.2	24 ± 19
1755	224 ± 18	–19.8	208 ± 18	–23.8	249 ± 19	–20.5	161 ± 18	–24.6	51 ± 18
1765	248 ± 20	–19.8	208 ± 20	–23.6	203 ± 18	–20.5	184 ± 18	–23.8	28 ± 19
1775	218 ± 19	–19.5	197 ± 18	–23.5	192 ± 18	–20.3	170 ± 18	–24.2	22 ± 18
1785	246 ± 19	–19.8	226 ± 20	–23.3	234 ± 18	–20.2	209 ± 18	–23.6	23 ± 19
1795	242 ± 20	–20.0	212 ± 20	–23.6	230 ± 18	–20.5	208 ± 20	–23.7	26 ± 20
1805	176 ± 19	–19.7	124 ± 20	–24.0	164 ± 19	–20.2	121 ± 19	–23.8	47 ± 19
1815	129 ± 20	–19.9	121 ± 20	–23.2	154 ± 19	–20.3	134 ± 19	–23.7	14 ± 20
1825	140 ± 20	–20.1	104 ± 19	–23.4	151 ± 19	–20.9	110 ± 20	–21.9	39 ± 20
1835	141 ± 18	–19.7	112 ± 18	–23.5	159 ± 20	–20.4	91 ± 20	–23.9	46 ± 19
1845	165 ± 20	–19.8	144 ± 20	–23.9	146 ± 20	–20.2	130 ± 21	–24.5	19 ± 20

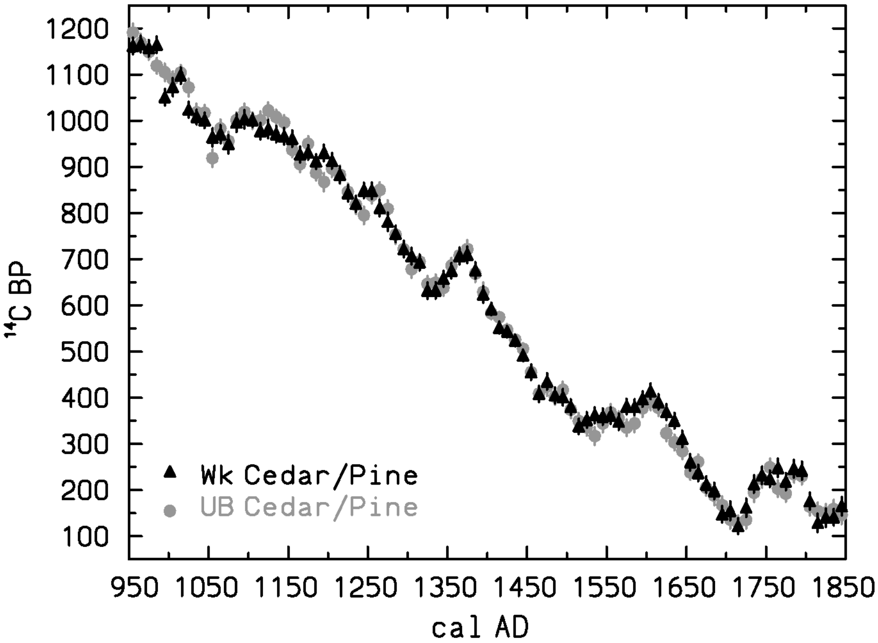

Figure 2 ^{14}C measurements on decadal samples of cedar and silver pine made in Queen's University of Belfast and the University of Waikato, New Zealand; (cedar AD 1405–1855; Hihitahi & Takapari Forest Parks, North Island, New Zealand; silver pine AD 955–1455, Oroko Swamp, South Island, New Zealand)

Table 4 ^{14}C measurements on the overlap between cedar and silver pine (AD 1405–1455)

Year	Lab	Cedar ^{14}C BP	Silver pine ^{14}C BP	Difference
1405	Wk	584 ± 16	602 ± 16	18 ± 22.6
1415	Wk	550 ± 16	554 ± 16	4 ± 22.6
1425	Wk	549 ± 16	537 ± 16	−12 ± 22.6
1435	Wk	515 ± 16	533 ± 16	18 ± 22.6
1445	Wk	477 ± 16	506 ± 16	29 ± 22.6
1405	UB	577 ± 21	587 ± 17	10 ± 27.0
1415	UB	569 ± 17	580 ± 18	11 ± 24.8
1425	UB	553 ± 17	538 ± 20	−15 ± 26.2
1435	UB	519 ± 17	533 ± 17	14 ± 24.0
1445	UB	500 ± 17	513 ± 20	13 ± 26.2

The combined measurements from the 2 laboratories on wood from the Southern and Northern Hemispheres are shown in Figure 3. The fact that the Southern Hemisphere measurements give older dates is clearly visible. The average value for the hemispheric offset over the 900 yr of measurement is 40 ± 13 yr. However, careful analysis shows that this value is not constant through time, but varies with a periodicity of about 130 yr. McCormac et al. (this issue), deals more thoroughly with this and make specific recommendations for the use of the data to calibrate ^{14}C ages.

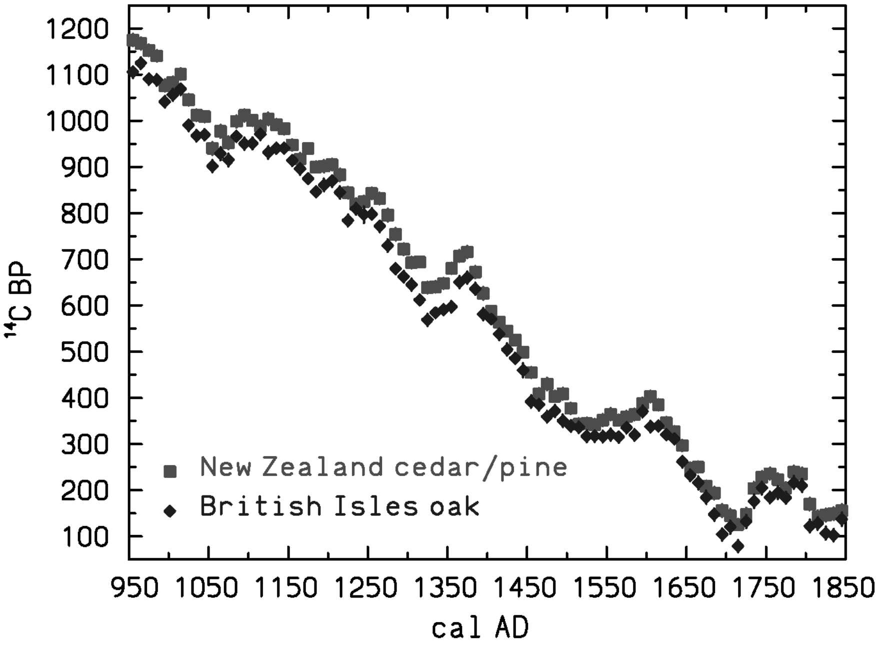

Figure 3 Comparison of combined ^{14}C dates of Southern and Northern Hemisphere wood measured by QUB and Waikato

CONCLUSIONS

Two sets of ^{14}C measurements on decadal samples of oak, cedar, and silver pine are presented covering the period AD 950–1850. The offset between the Waikato and Belfast laboratories is significantly lower than corresponding offsets presented in other studies. The Northern Hemisphere measurements confirm the validity of the Pearson et al. (1986) data and suggest its use in future calibration work.

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CALIBRATION OF THE RADIOCARBON TIME SCALE FOR THE SOUTHERN HEMISPHERE: AD 1850–950

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ABSTRACT. We have conducted a series of radiocarbon measurements on decadal samples of dendrochronologically dated wood from both hemispheres, spanning 1000 years (McCormac et al. 1998; Hogg et al. this issue). Using the data presented in Hogg et al., we show that during the period AD 950–1850 the ^{14}C offset between the hemispheres is not constant, but varies periodically (~130 yr periodicity) with amplitudes varying between 1 and 10‰ (i.e. 8–80 yr), with a consequent effect on the ^{14}C calibration of material from the Southern Hemisphere. A large increase in the offset occurs between AD 1245 and 1355. In this paper, we present a Southern Hemisphere high-precision calibration data set (SHCal02) that comprises measurements from New Zealand, Chile, and South Africa. This data, and a new value of 41 ± 14 yr for correction of the IntCal98 data for the period outside the range given here, is proposed for use in calibrating Southern Hemisphere ^{14}C dates.

INTRODUCTION

Regional radiocarbon offsets have been recognized in the past. Lerman et al. (1970), using wood from about AD 1835, showed that trees from the Southern Hemisphere dated older by approximately 30 (^{14}C) yr compared to identically aged Northern Hemisphere trees. More recently, Vogel et al. (1993) measured a 40-yr offset between hemispheres comparing data from the Netherlands and South Africa (wood spanning the yr AD 1835–1890). McCormac et al. (1998) measured hemispheric differences of 27 yr (British Isles/New Zealand; 18th to 19th century) and Stuiver and Braziunas (1998) measured differences of 23 yr (Western US/Chile; 19th century). Accepted theory attributes the difference to the influence of the larger expanse of ocean in the Southern Hemisphere and the atmosphere-ocean CO_2 exchange. Thus, ^{14}C is not uniformly distributed throughout the troposphere but exhibits hemispheric differences as observed in terrigenous organic material. The long-term question has been, do these differences vary temporally?

Small differences in the dates 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 signal that may exist (McCormac et al. 1995). For this reason, we have measured replicates of the oak (*Quercus petraea*), cedar (*Libocedrus bidwillii*), and silver pine (*Lagarostrobos colensoi)* at the Queen's University of Belfast and The University of Waikato, New Zealand ^{14}C dating laboratories Hogg et al. (this issue). Weighted means of the differences in the wood measurements show consistent ^{14}C depletion or older ^{14}C ages in the Southern Hemisphere over the period AD 950–1850. After these dates, i.e. in the period AD 1895–1935, the results from both laboratories show a reversal such that the Northern Hemisphere is slightly depleted. This reversal has been previously attributed to the burning of fossil fuel in the industrialized Northern Hemisphere (McCormac et al. 1998; Stuiver and Braziunas 1998).

In Figure 1, the mean differences of the data from Hogg et al. (SH offset) are plotted along with a 3-point moving average. A 120–130 yr periodicity with varying amplitude is clearly visible in the 3-

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point moving average. This periodicity is well above the red noise 95% confidence limit in the Fourier spectral analysis (Mitchell et al. 1966) (Figure 2) and was confirmed as one of the principle components of variation in the NS offset using singular spectrum analysis (Dettinger et al. 1995; Vautard et al. 1992). Spectral analysis was carried out on the data excluding the period before AD 1400 and the 120–130 yr periodicity persisted. A large increase in the SH offset is seen between AD 1245 and 1355.

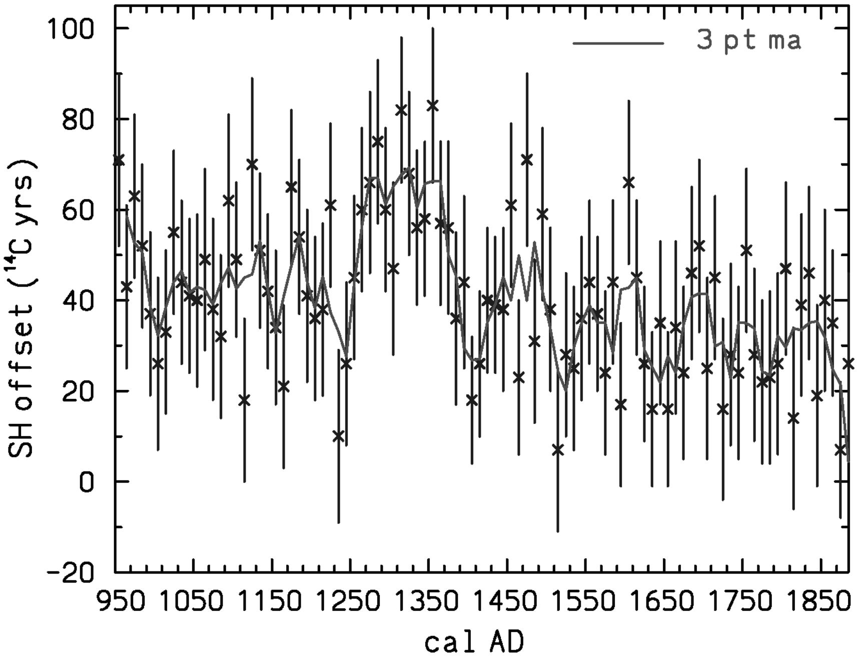

Figure 1 Difference between combined Southern and Northern Hemisphere measurements with a 3-point moving average superimposed

Previous work to establish the Southern Hemispheric offset has produced several high-precision datasets (Vogel et al. 1993; Sparks et al. 1995; Stuiver and Braziunas 1998). Although some of these datasets are based on ring-counted sequences rather than precise dendrochronological dates, we compared these results from the high-precision laboratories at the Universities of Washington (QL), Pretoria and Groningen (Pta/Grn), and the Rafter Radiocarbon Laboratory (NZ) with the combined measurements from Belfast and Waikato. The University of Washington data from Chile (previously published in graphical form in Stuiver and Braziunas 1998) is given in Table 1. The other data has been published previously. The offsets between the laboratories are shown in Table 2. There is a high level of agreement between our dataset and the QL and Pta/Grn measurements in terms of both the mean offset values and the spread of measurements. The mean offset value with the NZ measurements is larger especially for the period from AD 1335–1445. The NZ measurements were therefore not included in the Southern Hemisphere calibration dataset.

The combined Southern Hemisphere data are given in Table 3. Data have been combined by the method used for IntCal98 and described in Stuiver et al. (1998). The error multiplier (*k*) is a convenient measure of the degree to which the errors in measurement have been accounted for (Stuiver and Pearson 1986) with a value of 1.0 indicating that all sources of error in the measurements have

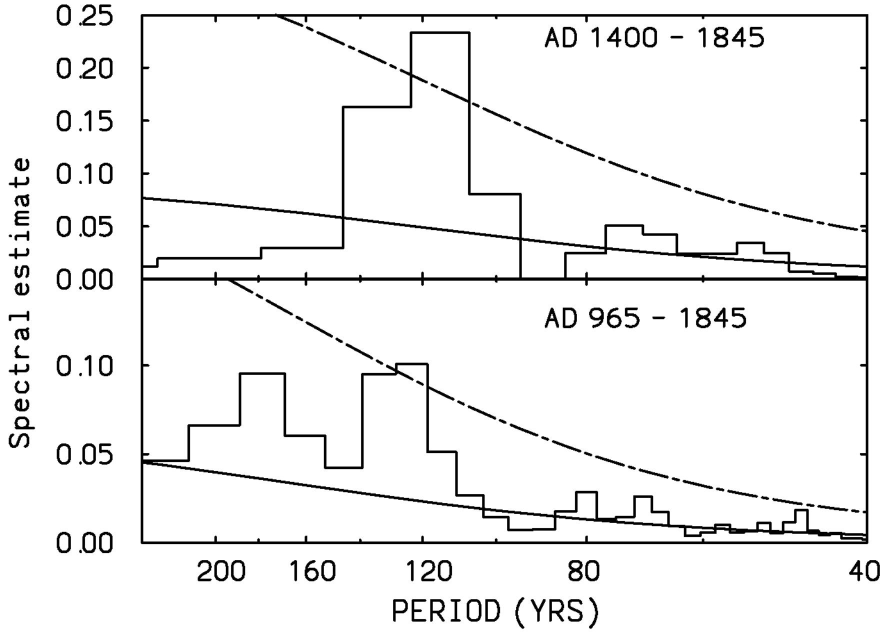

Figure 2 Spectral analysis of the difference between Northern and Southern Hemisphere measurements. The difference was detrended using a 20-point moving average and smoothed with a 3-point moving average. The 95% red noise confidence level is indicated by the dashed line.

been included, whereas values below 1.0 indicate an overestimate of the error. An error multiplier of 1.2 was used for the combined data from 1665 to 1955 based on the largest *k* value obtained for the differences between labs (Table 2). No error multiplier was applied to the data prior to 1665 as the *k* values indicate overestimates of the uncertainty were included in the Belfast and Waikato datasets. Taking the SH offset determined by subtracting the data in Table 3 from IntCal98 (Figure 3), we obtain a mean offset of 41 ± 1 yr with a standard deviation based on the spread in the differences of ±14. This value is similar to that found by Vogel et al. (1993), but encompasses a much longer time span. The offset may be used as a correction for Southern Hemisphere ^{14}C measurements beyond the range of the new data; however, past changes in the ocean-atmosphere regime could and will almost certainly have altered the inter-hemispheric difference in ^{14}C reservoirs especially beyond the Holocene. We therefore would suggest using this offset for Holocene samples only and recommend the use of the standard deviation of ±14 to encompass the time variation we observe over the last millennium.

DISCUSSION

The ocean effect on atmospheric ^{14}C gradients has previously been modelled (Stuiver and Braziunas 1998; Levin et al. 1987; Braziunas et al. 1995). The relative depletion of ^{14}C in the Southern Hemisphere atmosphere before 1885 can be attributed to the differences in ocean circulation and CO_2 exchange between ocean and atmosphere in the 2 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 and longer exposure time to the atmosphere. We used a 2-hemisphere ocean-atmosphere box diffusion model with our Northern Hemispheric ^{14}C record as input to identify possible causes of the increased SH offset for this period. We found that, by using the original parameters of Stuiver and Braziunas (1998), the model Southern Hemisphere atmosphere fit the data exceeding well from AD 1400 to 1840, but was less depleted than the ^{14}C values of the preceding period. Increases in the air-sea CO_2 exchange rate and the eddy diffusivity did not significantly alter the model output. Addition of an advection to the deep ocean and the subsequent replacement by upwelling in the Southern Hemisphere or an increase in the tropospheric exchange rate for the period AD 1200–1400 improved the fit considerably. This modelling work and discussion of the periodicity in the SH offset is expanded in the forthcoming paper by Reimer et al.

Table 1 Measurements on Chilean wood samples (*Notofagus dombeyi*) at the University of Washington. Uncertainties are based on counting statistics. ^{14}C ages are not given for samples later than AD 1958, after which ^{14}C from nuclear testing makes ^{14}C ages meaningless.

Year AD (midpoint)	Number of rings	^{14}C (BP)	Δ^{14}C (‰)	δ^{13}C (‰)
1665	10	271 ± 17	0.8 ± 2.1	–24.6
1671.5	3	205 ± 20	8.3 ± 2.5	–24.0
1674.5	3	219 ± 13	6.1 ± 1.7	–24.0
1677	2	200 ± 10	8.2 ± 1.3	–23.9
1679	2	192 ± 13	9.0 ± 1.6	–23.6
1681	2	178 ± 13	10.5 ± 1.7	–23.4
1683	2	177 ± 13	10.4 ± 1.6	–23.3
1685	2	171 ± 15	10.8 ± 1.9	–23.2
1687	2	196 ± 13	7.5 ± 1.6	–23.9
1688.5	5	163 ± 15	11.5 ± 1.9	–25.0
1689	2	172 ± 15	10.3 ± 1.9	–24.1
1691	2	165 ± 15	10.9 ± 1.9	–23.7
1692.5	1	173 ± 20	9.8 ± 2.6	–23.7
1693.5	1	185 ± 10	8.1 ± 1.2	–23.4
1694.5	1	135 ± 17	14.2 ± 2.1	–23.0
1695.5	1	188 ± 14	7.4 ± 1.8	–23.3
1696.5	1	159 ± 9	11.0 ± 1.2	–23.5
1697.5	1	161 ± 13	10.7 ± 1.6	–23.3
1698.5	1	177 ± 13	8.5 ± 1.6	–23.2
1698.5	5	154 ± 10	11.3 ± 1.3	–24.4
1699.5	1	156 ± 14	11.0 ± 1.8	–23.3
1700.5	1	149 ± 9	11.8 ± 1.2	–23.4
1701.5	1	154 ± 13	11.1 ± 1.6	–23.4
1702.5	1	151 ± 14	11.2 ± 1.8	–23.2
1703.5	1	152 ± 13	11.0 ± 1.6	–22.8
1703.5	5	141 ± 11	12.5 ± 1.4	–24.2
1705	2	167 ± 9	8.9 ± 1.1	–23.0
1706.5	1	138 ± 9	12.4 ± 1.1	–22.9
1707.5	1	149 ± 13	10.9 ± 1.7	–22.2
1708.5	1	178 ± 10	7.2 ± 1.3	–22.7
1708.5	5	146 ± 17	11.1 ± 2.1	–24.0
1709.5	1	154 ± 10	10.0 ± 1.2	–23.1

Table 1 Measurements on Chilean wood samples (*Notofagus dombeyi*) at the University of Washington. Uncertainties are based on counting statistics. ^{14}C ages are not given for samples later than AD 1958, after which ^{14}C from nuclear testing makes ^{14}C ages meaningless. (*Continued*)

Year AD (midpoint)	Number of rings	^{14}C (BP)			Δ^{14}C (‰)			δ^{13}C (‰)
1710.5	1	145	±	13	11.1	±	1.6	–22.8
1711.5	1	144	±	15	11.1	±	1.8	–22.8
1712.5	1	140	±	13	11.4	±	1.6	–22.8
1713.5	1	149	±	15	10.1	±	1.8	–23.0
1713.5	5	149	±	15	10.2	±	1.9	–24.3
1714.5	1	136	±	13	11.7	±	1.6	–22.9
1715.5	1	127	±	7	12.7	±	0.9	–23.1
1716.5	1	144	±	8	10.5	±	1.1	–23.0
1717.5	1	137	±	14	11.2	±	1.7	–23.1
1718.5	1	145	±	14	10.0	±	1.7	–22.7
1719.5	1	134	±	14	11.3	±	1.8	–22.3
1720.5	1	155	±	14	8.5	±	1.7	–21.6
1723	4	169	±	14	6.5	±	1.7	–22.3
1727.5	5	169	±	11	6.0	±	1.4	–22.8
1732.5	5	203	±	15	1.1	±	1.8	–22.0
1737.5	5	202	±	14	0.6	±	1.7	–21.7
1742.5	5	215	±	14	–1.7	±	1.7	–22.0
1747.5	5	215	±	14	–2.1	±	1.8	–21.8
1752.5	5	236	±	14	–5.4	±	1.7	–21.9
1757.5	5	190	±	14	–0.3	±	1.8	–22.0
1762.5	5	213	±	14	–3.7	±	1.7	–22.3
1767.5	5	235	±	14	–7.1	±	1.7	–21.8
1772.5	5	243	±	14	–8.7	±	1.7	–22.0
1773	5	206	±	12	–4.1	±	1.5	–22.0
1776.5	3	207	±	10	–4.7	±	1.3	–21.5
1779	2	212	±	9	–5.6	±	1.1	–21.7
1781	2	224	±	14	–7.4	±	1.7	–21.5
1783	2	223	±	10		±		–21.6
1785	2	208	±	15	–5.9	±	1.8	–21.2
1787	2	242	±	12	–10.3	±	1.4	–20.9
1789	2	226	±	15	–8.6	±	1.8	–21.1
1791	2	253	±	14	–12.2	±	1.8	–21.1
1793	2	267	±	15	–14.1	±	1.8	–21.3
1795	2	224	±	10	–9.1	±	1.2	–21.3
1797	2	240	±	15	–11.3	±	1.8	–20.8
1799	2	198	±	11	–6.3	±	1.4	–20.6
1801	2	193	±	10	–6.0	±	1.2	–21.1
1803	2	165	±	15	–2.7	±	1.8	–21.5
1804.5	1	170	±	14	–3.5	±	1.7	–21.5
1805.5	1	173	±	14	–4.0	±	1.8	–21.3
1807	2	185	±	14	–5.7	±	1.7	–21.6

Table 1 Measurements on Chilean wood samples (*Notofagus dombeyi*) at the University of Washington. Uncertainties are based on counting statistics. ^{14}C ages are not given for samples later than AD 1958, after which ^{14}C from nuclear testing makes ^{14}C ages meaningless. (*Continued*)

Year AD (midpoint)	Number of rings	^{14}C (BP)	Δ^{14}C (‰)	δ^{13}C (‰)
1809	2	160 ± 12	−2.8 ± 1.4	−20.9
1810.5	1	149 ± 16	−1.6 ± 2.0	−21.1
1811.5	1	136 ± 14	−0.1 ± 1.7	−21.1
1812.5	1	134 ± 14	0.0 ± 1.8	−21.2
1814	2	156 ± 13	−2.9 ± 1.7	−20.6
1815.5	1	142 ± 14	−1.3 ± 1.7	−20.8
1816.5	1	131 ± 14	−0.1 ± 1.7	−20.7
1817.5	1	120 ± 13	1.2 ± 1.7	−20.7
1819.5	3	97 ± 14	3.8 ± 1.7	−20.9
1824	2	104 ± 13	2.3 ± 1.6	−21.6
1828	5	107 ± 14	1.5 ± 1.8	−21.8
1833	5	143 ± 11	−3.6 ± 1.3	−21.5
1838	5	156 ± 13	−5.8 ± 1.6	−21.2
1843	5	164 ± 10	−7.3 ± 1.2	−21.5
1847.5	5	170 ± 14	−8.6 ± 1.7	−21.8
1850.5	3	148 ± 9	−6.4 ± 1.1	−22.2
1853.5	3	132 ± 15	−4.6 ± 1.9	−22.6
1856.5	3	142 ± 15	−6.3 ± 1.8	−22.2
1862.5	3	110 ± 14	−3.0 ± 1.8	−22.6
1865.5	3	129 ± 11	−5.8 ± 1.3	−22.7
1868.5	3	119 ± 14	−4.9 ± 1.8	−22.7
1871.5	3	144 ± 14	−8.4 ± 1.7	−22.6
1874.5	3	169 ± 10	−11.7 ± 1.2	−22.5
1877.5	3	136 ± 15	−8.1 ± 1.8	−22.8
1880.5	3	134 ± 14	−8.1 ± 1.8	−23.1
1883.5	3	135 ± 15	−8.7 ± 1.8	−22.9
1886.5	3	115 ± 15	−6.5 ± 1.8	−22.9
1889.5	3	121 ± 15	−7.7 ± 1.8	−23.0
1898	5	110 ± 10	−7.4 ± 1.3	−21.4
1903	5	146 ± 9	−12.4 ± 1.0	−21.2
1908	5	133 ± 10	−11.4 ± 1.2	−21.1
1913	5	147 ± 8	−13.7 ± 1.0	−20.6
1918	5	125 ± 14	−11.6 ± 1.8	−20.7
1923	5	133 ± 14	−13.2 ± 1.8	−20.0
1928	5	123 ± 14	−12.6 ± 1.8	−20.2
1933	5	158 ± 14	−17.5 ± 1.8	−20.7
1938	5	146 ± 14	−16.6 ± 1.7	−20.4
1943	5	162 ± 14	−19.1 ± 1.7	−20.6
1948	5	164 ± 15	−20.0 ± 1.8	−21.3
1953	5	177 ± 15	−22.1 ± 1.8	−20.8
1958	5	187 ± 15	−24.0 ± 1.8	−20.9

Table 1 Measurements on Chilean wood samples (*Notofagus dombeyi*) at the University of Washington. Uncertainties are based on counting statistics. ^{14}C ages are not given for samples later than AD 1958, after which ^{14}C from nuclear testing makes ^{14}C ages meaningless. (*Continued*)

Year AD (midpoint)	Number of rings	^{14}C (BP)		$\Delta^{14}C$ (‰)			$\delta^{13}C$ (‰)
1963	5	–	–	107.6	±	2.0	–21.3
1968	5	–	–	402.6	±	2.0	–21.3
1973	5	–	–	552.7	±	2.6	–21.4
1978	7	–	–	431.9	±	3.5	–20.8

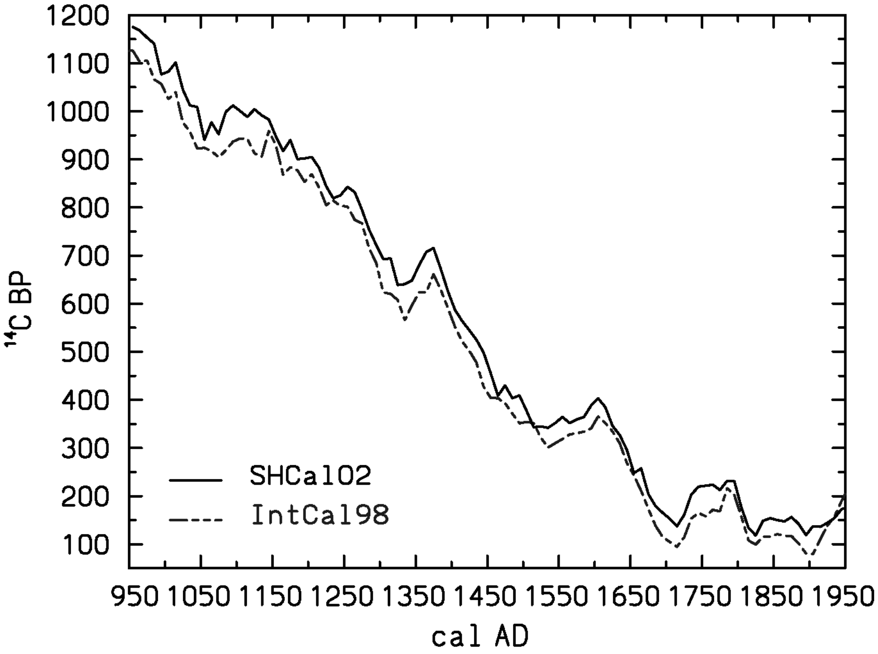

Figure 3 Comparison of the combined Southern Hemisphere datasets with IntCal

Table 2 Offsets between high-precision Southern Hemispheric datasets. σ_1 is the average standard deviation based on quoted laboratory errors and σ_2 is the observed standard deviation in the age difference. The error multiplier k = σ_2/σ_1.

Laboratories	Offset	s_1	s_2	k	N	Interval
UB – Wk	–3.9 ± 2.5	25.3	23.6	0.9	100	AD 955–1945
UB/Wk average – UW	5.5 ± 2.8	15.5	18.3	1.2	29	AD 1665–1945
UB/Wk average – Pta/Grn	1.4 ± 5.7	15.4	12.1	0.8	7	AD 1835–1895
UB/Wk average – NZ	16.2 ± 3.5	23.1	31.3	1.4	41	AD 1335–1745
UB/Wk average – NZ	39.2 ± 6.6	23.1	26.8	1.2	12	AD 1335–1445

Table 3 Combined Southern Hemisphere data from New Zealand, Chile, and South Africa (SHCal02) covering the period AD 955–1955. An error multiplier of 1.2 was applied to the uncertainties from 1665–1955, whereas no additional error term was included prior to this.

Year AD	Δ^{14}C (‰)			^{14}C BP		
955	−25.6	±	1.7	1175	±	13
965	−25.8	±	1.6	1167	±	13
975	−25.2	±	1.5	1153	±	12
985	−24.9	±	1.5	1141	±	12
995	−18.3	±	1.6	1077	±	13
1005	−20.3	±	1.6	1083	±	13
1015	−23.7	±	1.5	1101	±	12
1025	−18.1	±	1.6	1045	±	13
1035	−15.2	±	1.6	1012	±	12
1045	−16.0	±	1.5	1009	±	12
1055	−8.8	±	1.7	940	±	14
1065	−14.5	±	1.8	977	±	14
1075	−12.6	±	1.8	953	±	14
1085	−19.5	±	1.6	999	±	13
1095	−22.3	±	1.7	1012	±	14
1105	−22.1	±	1.5	1001	±	12
1115	−21.8	±	1.6	988	±	13
1125	−24.9	±	1.7	1004	±	14
1135	−24.6	±	1.5	992	±	12
1145	−24.7	±	1.5	983	±	12
1155	−21.6	±	1.5	948	±	12
1165	−19.0	±	1.5	917	±	12
1175	−23.0	±	1.5	940	±	12
1185	−19.3	±	1.5	900	±	12
1195	−20.7	±	1.7	902	±	14
1205	−22.3	±	1.5	905	±	12
1215	−20.8	±	1.7	883	±	14
1225	−17.3	±	1.6	845	±	13
1235	−15.4	±	1.6	819	±	13
1245	−17.3	±	1.6	825	±	13
1255	−20.6	±	1.5	843	±	12
1265	−20.4	±	1.5	832	±	12
1275	−17.3	±	1.7	796	±	14
1285	−13.3	±	1.7	754	±	14
1295	−10.6	±	1.6	722	±	13
1305	−8.1	±	1.6	693	±	13
1315	−9.6	±	1.4	694	±	11
1325	−3.9	±	1.6	639	±	13
1335	−5.3	±	1.6	641	±	13
1345	−7.5	±	1.5	648	±	12

Table 3 Combined Southern Hemisphere data from New Zealand, Chile, and South Africa (SHCal02) covering the period AD 955–1955. An error multiplier of 1.2 was applied to the uncertainties from 1665–1955, whereas no additional error term was included prior to this. (*Continued*)

Year AD	Δ^{14}C (‰)			^{14}C BP		
1355	−12.7	±	1.5	681	±	12
1365	−17.2	±	1.6	708	±	13
1375	−19.3	±	1.6	716	±	13
1385	−15.3	±	1.6	673	±	13
1395	−10.7	±	1.7	626	±	14
1405	−7.2	±	1.2	587	±	9
1415	−5.5	±	1.1	564	±	9
1425	−4.3	±	1.1	545	±	9
1435	−3.1	±	1.1	525	±	9
1445	−0.9	±	1.1	498	±	9
1455	3.2	±	1.5	455	±	12
1465	7.8	±	1.5	409	±	12
1475	3.9	±	1.7	430	±	14
1485	6.0	±	1.5	404	±	12
1495	4.1	±	1.6	409	±	13
1505	6.9	±	1.5	377	±	12
1515	9.9	±	1.5	343	±	12
1525	8.6	±	1.5	345	±	12
1535	7.7	±	1.6	342	±	13
1545	5.2	±	1.6	352	±	13
1555	2.4	±	1.6	365	±	13
1565	2.8	±	1.5	352	±	12
1575	0.6	±	1.5	359	±	12
1585	−1.2	±	1.6	364	±	13
1595	−5.4	±	1.6	388	±	13
1605	−8.4	±	1.5	403	±	12
1615	−7.4	±	1.5	385	±	12
1625	−3.8	±	1.5	346	±	12
1635	−2.5	±	1.5	327	±	12
1645	0.0	±	1.5	297	±	12
1655	4.9	±	1.5	248	±	12
1665	2.5	±	1.5	257	±	12
1675	7.8	±	0.9	204	±	7
1685	9.7	±	0.8	179	±	6
1695	10.3	±	0.6	165	±	5
1705	10.7	±	0.5	153	±	4
1715	11.4	±	0.5	138	±	4
1725	7.3	±	1.0	161	±	8
1735	0.8	±	1.2	203	±	10
1745	−2.5	±	1.2	220	±	10

Table 3 Combined Southern Hemisphere data from New Zealand, Chile, and South Africa (SHCal02) covering the period AD 955–1955. An error multiplier of 1.2 was applied to the uncertainties from 1665–1955, whereas no additional error term was included prior to this. (*Continued*)

Year AD	Δ^{14}C (‰)			^{14}C BP		
1755	–4.0	±	1.2	222	±	10
1765	–5.4	±	1.2	224	±	10
1775	–5.3	±	0.7	212	±	6
1785	–8.8	±	0.8	232	±	6
1795	–10.0	±	0.8	231	±	6
1805	–4.2	±	0.7	175	±	6
1815	–0.2	±	0.7	134	±	6
1825	0.3	±	1.2	118	±	9
1835	–4.6	±	0.9	148	±	7
1845	–6.5	±	0.9	154	±	7
1855	–7.1	±	0.7	150	±	6
1865	–8.2	±	0.8	148	±	6
1875	–10.4	±	0.6	156	±	5
1885	–9.9	±	0.7	142	±	6
1895	–8.4	±	0.9	119	±	7
1905	–11.6	±	0.8	137	±	7
1915	–12.6	±	0.9	136	±	8
1925	–14.8	±	1.2	144	±	10
1935	–17.3	±	1.2	155	±	10
1945	–20.7	±	1.2	173	±	10
1955	–22.1	±	2.2	177	±	17

CONCLUSIONS

A comparison between 2 sets of ^{14}C measurements on decadal samples of oak, cedar, and silver pine is presented covering the period AD 955–1845. The results show a periodicity in the offset of about 130 yr with a large change in amplitude centered around AD 1300.

We propose the use of the Southern Hemisphere data presented in Table 3 for calibrating samples from the Southern Hemisphere over the 2nd millennium AD. On the basis of the difference between this and IntCal98, we suggest that the Southern Hemisphere offset value that should be used for calibration prior to AD 955 is 41 ± 14. The ^{14}C calibration program CALIB at www.calib.org includes the new Southern Hemisphere data set and provides the option to calibrate Holocene Southern Hemisphere data before AD 955 using the new recommended offset.

While our model study is still in the preliminary stages, it does point out that increased upwelling in the Southern Ocean, caused by either increased deep water formation there or in the North Atlantic, could be responsible for the increased difference in atmospheric ^{14}C around AD 1300–1400. Whatever the cause of the variation in the inter-hemispheric difference, correction of Southern Hemispheric ^{14}C ages with a constant offset could result in an error of up to 50 yr.

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PRELIMINARY REPORT OF THE FIRST WORKSHOP OF THE INTCAL04 RADIOCARBON CALIBRATION/COMPARISON WORKING GROUP

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ABSTRACT. The first meeting of the IntCal04 working group took place at Queen's University Belfast from April 15 to 17, 2002. The participants are listed as co-authors of this report. The meeting considered criteria for the acceptance of data into the next official calibration dataset, the importance of including reliable estimates of uncertainty in both the radiocarbon ages and the cal ages, and potential methods for combining datasets. This preliminary report summarizes the criteria that were discussed, but does not yet give specific recommendations for inclusion or exclusion of individual datasets.

INTRODUCTION

Calibrated radiocarbon ages are the basis for comparison between many records in paleoclimatological, geophysical, and archaeological studies. It is, therefore, necessary to have a standard ^{14}C calibration dataset for intercomparisons to be valid. The ^{14}C community has recognized this need and hence has adopted an international standard for calibration, most recently IntCal98 (Stuiver et al. 1998a), which was ratified at the 16th International Radiocarbon Conference in Groningen (van der Plicht and Mook 1998). The importance of the quality of the dataset used for calibration cannot be overstated. As new data become available, careful consideration must be given to whether or not they should be included in the official calibration dataset. A balance must be maintained between accepting only true "calibration" datasets (i.e. those where the age on the cal axis is known very precisely such as dendrochronological dated tree rings and U/Th dated pristine corals) and incorporating all available datasets. A conservative approach has been followed in the past to avoid inclusion of datasets with potential errors in the cal ages or other underlying assumptions. While this means that some information about the details of the calibration dataset may be unavailable, the conservative approach assures that general trends are reliable. Therefore, we are advocating such a conservative approach for IntCal in the future.

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CRITERIA FOR ACCEPTANCE FOR CALIBRATION AND COMPARISON DATASETS

In general, for acceptance into the IntCal compilation, an individual dataset must pass certain criteria that depend on the type of record and measurements made. In all cases, uncertainties in both the ^{14}C age and the cal age must be quantified. A review of the datasets included in the IntCal98 calibration dataset and discussion of some problems with various "comparison" datasets are given by van der Plicht (2000). While some otherwise good datasets may not meet all criteria for inclusion into the calibration dataset, they may still be valuable for comparisons and for checking the magnitude of atmospheric excursions and/or marine reservoir corrections.

Datasets with large analytical errors in either the cal age or the ^{14}C age will only add noise and will be excluded. New data must be as good as extant IntCal98 for the time period in question. We would like input on what analytical errors would be considered acceptable for older samples where we know it is difficult to obtain highly precise and accurate measurements. It is not likely that we will develop a calibration record with ±30 yr ^{14}C errors to 50,000 yr!

Tree-Ring Criteria

Tree rings should be dendrochronologically dated and cross-checked where possible. In some cases, trees with a known felling age may be used by ring counting, if rings are well developed and single annual ring production in the species is known to be robust. For instance, the Douglas-firs (*Pseudotsuga menziesii*) from the Pacific Northwest were ring counted back to AD 1320 (Stuiver 1982). Uncertainty in single ring cal ages for dendrochronologically dated wood is on the order of 1 yr for highly replicated and crosschecked chronologies. An additional small uncertainty in cal age exists for ^{14}C dates on multiple yr blocks of wood, because of an unequal amount of carbon in each ring. In extreme cases, this could result in an uncertainty of several ^{14}C yr only. Floating tree-chronologies have been used in the past with a wiggle-matched connection to the dendrochronological secured chronology (Stuiver et al. 1998a). While this approach could be problematic, we note that the German pine wiggle-match ~10,000 cal BP used in IntCal98 was only 8 yr different from the dendrochronological result as detailed in a later section.

Coral Criteria

X-ray diffraction measurements should be performed and should show ≤1% calcite as a check against secondary calcite. The [U] of fossil corals should be within the range of living/modern samples taking into account differences between coral species and the natural variability expected from sea surface temperature (SST) changes. In order to increase our confidence that the corals have not been altered by diagenesis, $\delta^{234}U_{\text{initial}}$ of fossil corals should be within ±5‰ of the accepted modern seawater value. This criterion is based on current understanding that δ^{234}U in seawater is constant over the last ~30,000 yr. Accuracy and precision should be checked by measuring and reporting the δ^{234}U of recent corals and/or seawater (Delanghe et al. 2002). An alternative demonstration of accuracy and precision is to report δ^{234}U of an international standard with δ^{234}U similar to natural uranium (e.g. SRM-U960 or –U010) (Bard et al. 1998; Edwards et al. 1993). External replicates of different parts of the same coral sample should be measured as often as possible to derive an estimate of external reproducibility, which should be reported with the results. This demonstrates the overall reproducibility of U-Th ages and is also a necessary requirement suggesting closed system behavior. Interlaboratory comparisons of standards and coral samples are to be encouraged. New datasets should be encouraged to include protactinium measurement in particular for samples that have been subaerially exposed in the past. While it is impractical to measure protactinium for all previous coral calibration datasets, if discrepancies arise, we would encourage reanalysis of a sub-set of the original samples. AMS-^{14}C coral samples need to be physically separated and subjected to partial dissolution on the order of 30–50% with the degree of leaching necessary to be determined by experiment. Background correction must be applied and background errors included in the error analysis. External replicates should be done wherever possible to derive a more accurate external reproducibility.

The error bar of U-Th ages should be on the order or less than that obtained by ^{14}C measurement on the same sample. Consequently, samples counted by alpha spectrometry do not have sufficient precision to be included in IntCal. Very precise measurements do not necessarily mean accurate—we expect that there will be instances where there will be results that we cannot explain even if they meet our geochemical criteria. In part, this is a consequence of the subtleties of diagenesis and it will more likely impact those samples that have been subaerially exposed or have been recovered from fast uplifting regions.

A site-specific reservoir correction should be estimated with a "reasonable" error, if at all possible, although this may prove to be untenable for time periods of rapidly changing atmospheric ^{14}C. There is a preference for future records to be developed from oceanographically "simple" regions to minimize reservoir age uncertainty. Coral datasets should have a well-described and published stratigraphy. The ages should be distributed in a logical manner along the stratigraphic sequence, taking into account rates of sea-level changes, vertical movements (subsidence or uplift), the paleobathymetry of the coral species and the topography of the reef substratum.

Non-Coral Carbonates Criteria

Records of non-coral carbonates, such as flowstones, stalagmites, and aragonite deposits, have additional uncertainties in both the U/Th ages and ^{14}C ages. This is the result of possible incorporation of detrital Th and old or dead carbon from carbonate dissolution processes (e.g. water-rock interaction) or other sources of old carbon in lacustrine/marine environments. The initial conditions of the carbon budget need to be confirmed by independent environmental indicators (e.g. δ^{13}C). The U/Th age model should be verified by layer counting or ^{14}C measurements of terrestrial macrofossils, where possible. Appropriate corrections to measured values need to be applied. It is important to evaluate these conditions for various growth/deposition periods as the relative contribution of detrital Th and dead/old carbon may significantly change through time as a result of changes in the climate regime (e.g. ocean circulation, precipitation, temperature, vegetation). As with corals, partial dissolution of carbonates may be necessary to remove contamination, but the degree of leaching required should be determined for each particular material.

Non-Dendro Layer Counting Criteria (Terrestrial and Marine)

Multiple-core chronologies are critical in order to confirm that no sections are missing (e.g. from core-breaks, erosional scour, etc.). A "multiple proxy" approach in counting should be used. Discussion of errors should include uncertainty about the stability of the system producing layers through climatic changes, the likelihood of unrecognized depositional hiatuses, and any ambiguities in layer interpretation that are not resolvable by even "perfect" physical archive recovery (e.g. could lakes have frozen over during colder climates?). It is not sufficient to report only counting statistics. For both laminated sediments and ice cores, it is important that counting be replicated, whenever possible, by experienced individuals in independent laboratories (Hicks et al. 1991). Confirmation of the counting should also be made with independent chronologies and/or tie-points. Ideally, individual varves would be correlated across cores (Ralska-Jasiewiczowa et al. 1998). We acknowledge that there will be some sets of data that might not have the possibility of physical replication via multiple cores and in this instance independent chronologies and/or tie-points are increasingly important. An obvious example of this would be geochemical identification of precisely dated ash horizons (Gronvold et al. 1995).

There should be a minimal level of acceptable scatter in macrofossil ^{14}C ages due to reworking or delayed transport of old organic material, although this should be evaluated "piecewise". This means that a particular varved record could still be useful even if it includes a section with relatively high scatter. The type of macrofossils used will be undoubtedly vary, but should be reported for each sample, as it influences the interpretation of how long it takes the macrofossil from "creation" to deposition in the sediment. Where possible, multiple macrofossils should be analyzed. For marine sequences, the reservoir correction and the uncertainty in reservoir changes over time must be estimated.

Reconstructed Time Scales

Records with a calendar timescale based on correlation with signals in annually counted ice cores, or sediments with assumed constant ^{14}C time/depth relationship and absolute time markers have been used to reconstruct "comparison" curves. Many of these are marine records, and criteria for minimum acceptable boundary conditions are needed. Hydrographically complex locations, such as inside the polar front or in upwelling regions where we expect large variations in local reservoir age, are not ideal. A site should have simple ^{14}C systematics not requiring complex circulation or ventilation dynamics (i.e. to explain disparate benthic/planktonic data). Defendable and reasonable estimates of minimal bioturbation should be made—for example using ash shard counts. As the chronology of many of these records is based on correlation of one or more proxies to ice core properties (GISP2 and GRIP), it is critical to identify the physical mechanism relating the proxy to the ice core property, as well as the timescale of the linking mechanism. In addition, the errors in the ice core timescales must be considered. We encourage the continued investigation of differences between GISP2 and GRIP (and eventually NGRIP) timescales, as well as identification of radiometrically dated tie-points (e.g. Laschamps geomagnetic excursion; Dansgaard-Oeschger climate events).

Geomagnetic and Cosmogenic Isotope Based Models

Models based on paleomagnetic intensity or ^{10}Be and the global carbon cycle are useful correlation tools and can be important indicators of the causes of ^{14}C variations. However, at present, our understanding of cosmogenic production and the global carbon cycle, as well as ^{10}Be transport, deposition, and post-depositional processes, is not sufficient to make such records suitable for ^{14}C calibration.

PROJECTIONS FOR INTCAL04

Refinements to IntCal98 for the period 0–12 ka BP were readily agreed upon by workshop participants and are presented below. These changes should result in only minor differences from IntCal98 through the Holocene. Between 12 and 15 ka BP, there is good agreement between datasets, several of which meet the criteria laid out in the earlier section, therefore, a "calibration" curve can be given with reasonable certainty, although there are some details which remain to be worked out. From 15 to 25 ka BP there are important differences between some of the existing datasets with only coral data truly meeting the established criteria. However, some additional detailed coral data should be available for this time period (Cutler et al. forthcoming). These new data, together with an alternative statistical technique to the spline fit of coral data used for IntCal98, will hopefully remove the very large uncertainty at around 15 ka BP, which results in the feature known as the "pig-in-the-python" (Figure 1).

Although the working group is not yet ready to make specific recommendations for IntCal04 beyond 25 ka BP, there was much discussion about the need to provide some guidelines, if not calibration datasets, for the entire ^{14}C timescale.

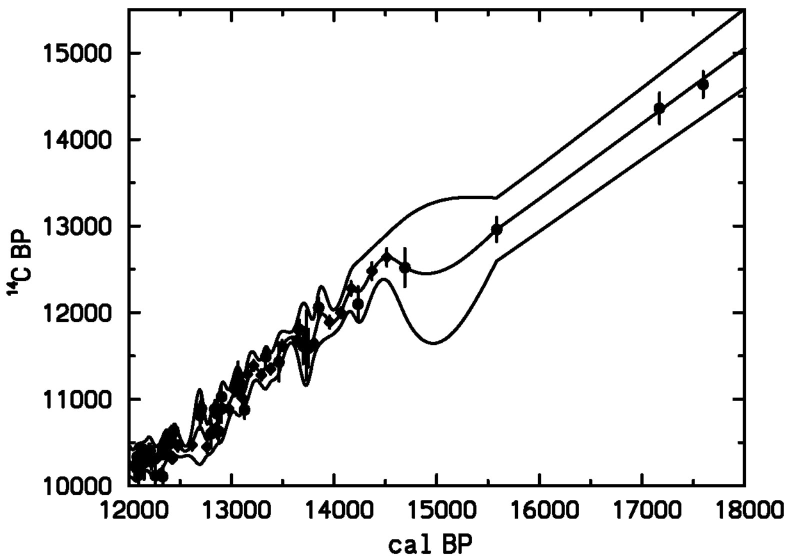

Figure 1 IntCal98 calibration curve and two-sigma error envelope. Individual data points from coral and foraminifera (varved sediment) have errors and reservoir corrections as reported in Stuiver et al. (1998a).

0–12 ka BP

The Holocene part of the ^{14}C calibration is based on several millennia-long tree-ring chronologies providing an annual, absolute time frame, which was rigorously tested by internal replication of many overlapping sections. Whenever possible, they were crosschecked with independently established chronologies of adjacent regions. The German and Irish oak chronologies were cross-dated until back into the 3rd millennium BC (Pilcher et al. 1984), and the German oak chronologies from the Main River, built independently in the Göttingen and Hohenheim tree-ring laboratories, cross-date back to 9147 cal BP (Spurk et al. 1998). The North American trees that form part of the ^{14}C calibration curve prior to AD 1320 were cross-dated with either the *Sequoiadendron* master chronology or with a Pacific Northwest Douglas-fir chronology (Stuiver 1982).

Before 9147 cal BP, we have to rely on single oak (back to 10,430 cal BP) and pine chronologies of the Hohenheim laboratory, which are not replicated externally. However, for the earliest oak the internal replication is high, and the trees cross-date with high statistical significance. Compared to the state entered into IntCal98, the German pine chronology has undergone several corrections:

1. The ^{14}C wiggle-match of the floating pine to the absolute oak chronology is superseded by a true dendro-synchronization of the 2 species, leading to a 8-yr shift of the pine with respect to the ^{14}C match (which had a ±20 yr confidence interval) (Friedrich et al. 1999).
2. The earliest centuries (prior to 11,200 cal BP) in the pine chronology, marked as tentative in Spurk et al. (1998), suffered severely from missing rings and a weak dendro-match. Using ample new finds, this part of the chronology is now securely cross-dated and sufficiently replicated. The tentative dendro-link as documented in Spurk et al. (1998) in the interval 11,200 to 11,370 cal BP is replaced by a statistically reliable match. The older part of the chronology therefore was shifted 70 yr to older ages. The former start of the German pine at 9922 cal BC (11,871 cal BP) (Spurk et al. 1998) is now shifted to 9992 cal BC (11,941 cal BP).

3. Including additional new finds the German pine is prolonged into the Younger Dryas. It now starts at 11,993 cal BP. A 20-tree pine chronology from Lake Neufchatel extends it further back to 12,057 cal BP.

Thus, while most of the ^{14}C calibration data as based on the German pine is affected only marginally by the corrections (+ 8 yr), in the first century of the Preboreal, and in the final five centuries of Younger Dryas the dendro-dates are shifted by 70 yr to older ages, compared to IntCal98. This interval happens to fall on a gently sloping ^{14}C age plateau (at 10,000–10,150 ^{14}C BP); hence the length of the plateau and the range of calibrated ages increase accordingly.

Because of periodic narrow rings caused by cockchafer beetles some German oak samples were excluded from IntCal98. Analysis of these tree-rings, with an understanding of the response of trees to the cockchafer damage, may allow some of these measurements to be re-instated in the chronology. Wood will be made available for measurement for any remaining gaps in the tree-ring ^{14}C record resulting from previous shifts.

Recent measurements of Belfast Irish oak over the past 1000 yr (Hogg et al. 2002) will be included to increase precision during this time period. Comparisons of these and other measurements with the 1986 and 1993 Irish oak datasets resulted in the acceptance of the 1986 datasets over the 1993 datasets. A Southern Hemisphere specific dendro-calibrated record will be included using the measurements of the New Zealand trees over the past 1000 yr (Hogg et al. 2002) and other Southern Hemisphere datasets (McCormac et al. 2002). An offset of 41 ± 14 ^{14}C yr BP from the IntCal98 dataset from AD 950 to 1850 will be used to extend the record through the Holocene. The offset will be recalculated for use with the new IntCal dataset. While this Southern Hemisphere ^{14}C offset may not be representative of the entire Holocene, we note that measurements for the early Holocene are of the same order of magnitude (Kromer et al. 1998).

The Arizona Bristlecone pine chronology (Linick et al. 1986), which was not included in IntCal98, was also discussed. Wiggle-matching the bristlecone pine dataset to IntCal98 suggests that there is not a problem with the dendrochronology but there is an average offset of approximately 35 yr in the ^{14}C ages with the bristlecone pine older. Part of this offset may be due to laboratory differences as the offset between 15 decades of bristlecone pine measured in both Seattle and Arizona is 25 ± 8 yr with Arizona measurements being older. However, comparisons made in several laboratories of bristlecone pine to German oak and Irish oak find the bristlecone pine consistently older. Furthermore, measurements in Arizona of single yr rings of sequoia from the Sequoia National Forest averaged as decades from AD 1065 to 1145 resulted in a difference of only –1.0 ± 2.1 ^{14}C yr BP compared to Seattle measurements of Douglas-fir from the Pacific Northwest. Therefore, while a regional or growing season difference during some time periods at least seems possible, more work is needed to resolve the issue. The treatment of offsets between datasets, and the inclusion of the Arizona Bristlecone pine measurements, will be discussed further at the next working group meeting. Additional tree-ring records will be considered for inclusion if available by the time of the next workshop.

The marine model used to produce the IntCal98 marine dataset from 0 to 8800 cal BP (Stuiver et al. 1998b) will be further tested by checking the model response to the nuclear-testing ^{14}C spike and to centennial-scale variations. The variability in the marine reservoir observed in recent records will be considered to better represent the uncertainty in the marine dataset for this period. Coral data (Bard et al. 1998; Burr et al. 1998; Edwards et al. 1993) and the Cariaco Basin varve dataset will provide the marine calibration from 8.8 to 12 ka BP. The high resolution Cariaco Basin dataset (Hughen et al. 2000) will replace the older varve dataset in IntCal98. The cal time scale will be optimized using appropriate coral tie-points within the error of the varve counting. As in previous versions of the marine calibration dataset, the time-dependent difference, ΔR(t), between the reservoir age of the region where samples are derived and the "global" calibration dataset, will need to be considered when calibrating marine samples.

12–15 ka BP

The coral datasets, including any new records meeting the IntCal criteria, and the Cariaco Basin varve dataset will provide the marine calibration for this period. The varved records from Lake Suigetsu in Japan (Kitagawa and van der Plicht 2000) and Lake Gościąż in Poland (Goslar et al. 2000) agree well with the "site-specific" reservoir corrected corals (T Goslar, personal communication 2000) and the new Cariaco Basin dataset but are not as high resolution as the Cariaco Basin and exhibit larger variability. It is anticipated that the atmospheric dataset for this portion of the curve will be based on the marine dataset with "site-specific" reservoir corrections and estimated uncertainties. The extent to which the reservoir corrections may vary with time will be estimated by comparison to lake varve macrofossil data, floating tree-rings, and other suitable paleorecords.

15–25 ka BP

Coral data provide the only secure calibration for this portion of the timescale. Although there is reasonable agreement between parts of the Bahamas speleothems (Beck et al. 2001) and the Lake Suigetsu varve record for this time period, the ^{14}C ages differ by up to 1200 yr at 20 ka BP suggesting that there is an error in either the Lake Suigestsu varve ages or the assumption of a constant dead carbon fraction in the Bahamas speleothem or a combination of both. At present, other available paleorecords either have cal age uncertainty that is too large or difficult to quantify, or require untested assumptions about the stability of the system producing the records. If other records or additional support for current records become available, they will be reconsidered at the next IntCal workshop.

Beyond 25 ka BP

Great strides have recently been made towards generating ^{14}C datasets beyond 25 ka BP with U/Th measured cal ages (Beck et al. 2001; Kitagawa and van der Plicht 2000; Schramm et al. 2000; Yokoyama et al. 2000) and with matches to the ice core timescales (van Kreveld et al. 2000; Voelker et al. 2000). However, these records are not sufficiently coherent that we can recommend a single calibration for this time period. Individually, many of these records reveal large and intriguing structure; however, differences of more than 5000 yr are observed amongst these records in some time frames. The choice of a particular dataset could result in vastly different "calibrated" age ranges (Bard 2001). The sources of these discrepancies are not currently understood, but may be due to some combination of artifacts relating to varve counting, ^{230}Th dating, uncertainties in the GISP2 chronology or correlation with GISP2 δ^{18}O, unaccounted for variability in reservoir correction or dead carbon fraction, or undetected overprinting from secondary alteration or authigenic mineral growth. Combining these various records together into a composite calibration would result in such a large error envelope as to make the calibration useless. Still, leaving the ^{14}C user-community to use "ad hoc" mixtures of ^{14}C records without proper consideration of potential errors in timescale or reservoir corrections is inadvisable, and could result in substantial confusion and misinterpretation. No decision was made at the Belfast IntCal04 working group meeting regarding how to resolve this problem, though it will be again addressed at the next workshop at Woods Hole Oceanographic Institution (WHOI). At the very least, we can advise the user community of the danger of ad-hoc calibration without proper consideration of all sources of uncertainty. Alternatively, we could attempt to provide a set of comparison curves that includes all quantifiable uncertainties in both the cal age and the ^{14}C age for these records, though these parameters may be difficult to assess for some cases.

Ideally, we would provide tools for the statistical estimation of cal age ranges given the types of information available to us. In any case, we recommend that no endorsement of a calibration in this time range be made at the present.

Post-Bomb Data

It was suggested that IntCal provide a compilation of post-nuclear testing atmospheric records. However, as the records are latitude dependent and many are not from clean air sites, it may be impractical to combine datasets. It would however be useful to provide access to all known post-bomb records in a single location with appropriate errors included. While some of this is already done at the CDIAC (Carbon Dioxide Information Analysis Center) as well as the NGDC "Paleo" database, we will attempt to provide an updated list of where post-bomb records can be found. We encourage individual investigators to digitally archive their respective datasets at the appropriate site, and this information should be provided in any original manuscripts.

Methods of Combining Data

In quantifying and reporting the error on the cal scale for the first time, we will need to reconsider the methods by which the data are combined to form the calibration curve. Thus, as well as conventional statistical methods of summarization, we will investigate the use of stochastic models for building calibration curves.

PRESENTATION OF RESULTS

It is difficult for the working group to predict what uses will be made of the data once they are released. As a result, and to provide as much flexibility as possible, the group agreed that it would be important to make the raw calibration data available in a suitable form for use in spectral analysis and curve modeling as well as to provide a recommended curve for immediate calibration purposes. It is anticipated that the IntCal04 calibration dataset will be presented for ratification at the 18th International Radiocarbon Conference in Wellington, New Zealand in 2003 prior to publication in early 2004.

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NEW RADIOCARBON CALIBRATION SOFTWARE

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ABSTRACT. We have developed a software utility, "DateLab", for conventional radiocarbon age (CRA) calibration and Bayesian analysis of CRAs. The current version has a smaller range of applicability than other similar utilities such as Bcal, Oxcal, and Mexcal. However, it enables analysis of some common types of CRA datesets. The main advantages of DateLab are its high quality sampling algorithm, the possibility of carrying out model comparison and hypothesis testing in a straightforward way, and the unbiased character of the summary statistics on which the analysis depends.

INTRODUCTION

A number of statistical packages for radiocarbon date calibration are freely available (see http://www.radiocarbon.org/). Those in most widespread use include Calib, described in Stuiver and Reimer (1993), which computes the likelihood[1] for each calibrated date, and OxCal, described in Ramsey (1995), which implements sample based Bayesian inference. We present another package, DateLab, implementing sample-based Bayesian inference. DateLab has, in some respects, a smaller range of applicability than existing packages. However, where it can be used DateLab offers high-quality, unbiased statistics and implements model comparison. This allows basic questions such as "Do these dates observe superposition?" to be quantified and statistically evaluated. As it is almost always the case that alternative models exist for Bayesian calibration this facility is a particularly useful feature in DateLab.

Calibration of independent CRAs is now a well-established process and a number of software packages (in particular Calib; Stuiver and Reimer 1993) exist to perform this calculation. However, in some cases it is useful to be able to calibrate CRAs that are related, in a manner that incorporates prior chronometric information into the calibration process. This is more complex than calibration by itself, and is typically performed via Bayesian inference (see Buck et al. 1996 and Christen 1994a for an overview of this type of application). Available packages include OxCal (Ramsey 1995), Bcal (Buck et al. 1999), and MexCal (Buck et al. 1999), all of which are designed to perform Bayesian calibration of CRAs. These tools are more than adequate for the job at hand, if used correctly. However, it is distressingly easy for users who are not experienced statisticians to inadvertently bias their calibration analyses. By restricting the class of models that can be applied and using more reliable algorithms to analyze those models, we hope to steer the user away from some uninteresting technical statistical problems that can otherwise invalidate their analysis. Here, we review the basic ideas of Bayesian inference, and then explain what those "uninteresting technical statistical problems" might be!

Bayesian Calibration

Bayesian methods for ^{14}C date calibration are now in widespread use (e.g. Naylor and Smith 1988; Buck et al. 1991, 1992, 1994, 1996; Christen 1994a, 1994b; Christen and Buck 1998; Christen and Litton 1995; Christen et al. 1995; Litton and Leese 1991; Nicholls and Jones 1998, 2001; Zeidler et al.1998). These statistical methods are attractive, as they allow associated chronometric information

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[1] The likelihood is equivalent to the calibrated distribution of a CRA.

to be taken into account, in an explicit way, in the chronometric analysis. For example, where artifacts are found in primary context, stratigraphic constraints can be applied to the calibrated dates. Information of this kind is made explicit in the analysis via a probability distribution, called the *prior*, which weights the calibrated dates toward values in line with our prior expectations. On the other hand, the data act through a distribution called the *likelihood*. A calibrated value that makes the observed CRA a likely outcome of the ^{14}C observation process has a high likelihood. The prior and likelihood distributions together determine a new probability distribution known as the *posterior*. Sets of calibrated dates agreeing with the data, and at the same time plausible in the light of prior information, yield a large posterior probability. In Bayesian calibration this posterior distribution is our analysis result.

Formally speaking, the un-normalized posterior distribution is given by:

$$\text{posterior} = \text{likelihood} \times \text{prior} \tag{1}$$

The likelihood is determined by the CRA data and the standard ^{14}C observation model (e.g. Buck et al. 1991). How do we summarize prior beliefs? An ideal approach is to try to specify a general "neutral", or non-informative prior probability distribution, that is, a type of prior that assigns equal probability to any set of calibrated dates. However, a single neutral set of prior beliefs cannot exist in archaeology or any other domain. Prior beliefs that are non-informative with respect to any one hypothesis must be informative with respect to some other hypothesis. We must accept a prior that is in some respects informative and check that the information is representative of knowledge available in the problem at hand. So, for example, Nicholls and Jones (1998, 2001) write a simple prior model and check that it is non-informative for many (but not all!) situations of practical interest in ^{14}C dating. Finally, when conflicting models are proposed, we simply ask, "Which prior model does the data support?". DateLab implements statistical tools that answer such questions.

The posterior distribution defined in Equation 1 is the joint probability of a number of event date parameters. We summarize this multidimensional distribution by considering the distribution of some meaningful statistic of direct interest (the span of calibrated ages might be one such salient statistic). We take the original joint posterior distribution of all parameters and integrate out the uninteresting parameters, in order to compute the "marginal distribution" for the remaining salient statistic. It is most often necessary to carry out the integration numerically, using a computer, as the integrals involved cannot usually be done by hand.

Several software packages designed to perform Bayesian calibration already exist. DateLab differs from these in terms of both functionality and in some aspects of the underlying mathematical framework. In particular, the default prior probability distribution for calibrated dates used in DateLab follows that given in Nicholls and Jones (1998, 2001). This prior avoids an undesirable bias toward more widely spread calibrated dates, which is imposed by the priors in common use[2]. A further difference is the manner in which the marginal distributions are integrated out of the posterior. These are typically calculated via some form of sampling procedure, which is usually based around Markov chain Monte Carlo (MCMC) techniques. MCMC samples require careful checking to ensure they really do represent the posterior distribution. It cannot be assumed that the typical user of Bayesian calibration software is versed in this highly technical subject. DateLab offers an

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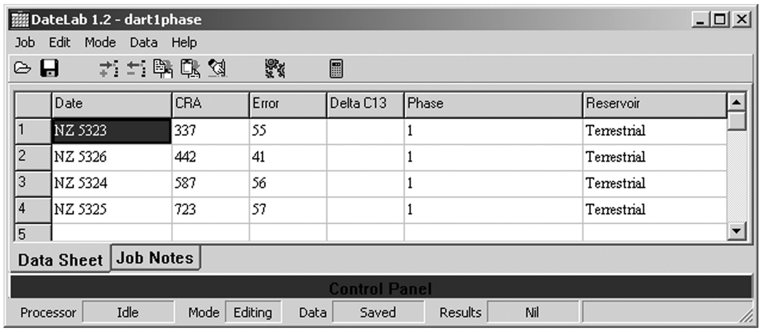
[2] This prior is now also applied in OxCal. However, it should be noted that the prior outlined by Nicholls and Jones (2001) has been explicitly developed for the limited phase model described in this paper. Application of their prior to more complex phase models can give rise to biased posterior distributions.

alternative to the MCMC sampling algorithm called the rejection sampling algorithm. Samples generated by the rejection algorithm can be relied on to represent the posterior distribution.

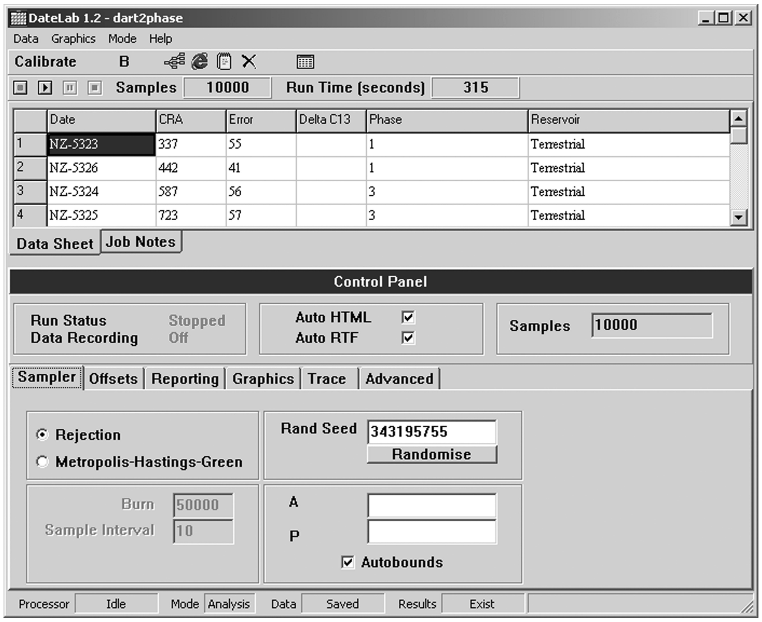
In the following, we outline the functionality of DateLab and detail the mathematical framework underlying the analysis process.



	Date	CRA	Error	Delta C13	Phase	Reservoir
1	NZ 5323	337	55		1	Terrestrial
2	NZ 5326	442	41		1	Terrestrial
3	NZ 5324	587	56		1	Terrestrial
4	NZ 5325	723	57		1	Terrestrial
5						

Processor Idle Mode Editing Data Saved Results Nil

Figure 1 DateLab data entry screen



Samples 10000 Run Time (seconds) 315

	Date	CRA	Error	Delta C13	Phase	Reservoir
1	NZ-5323	337	55		1	Terrestrial
2	NZ-5326	442	41		1	Terrestrial
3	NZ-5324	587	56		3	Terrestrial
4	NZ-5325	723	57		3	Terrestrial

Run Status Stopped
Data Recording Off
Auto HTML ☑
Auto RTF ☑
Samples 10000

● Rejection
○ Metropolis-Hastings-Green
Rand Seed 343195755
Burn 50000
Sample Interval 10
A
P
☑ Autobounds

Processor Idle Mode Analysis Data Saved Results Exist

Figure 2 DateLab analysis screen

DateLab Functionality

The basic operation of DateLab is via a simple graphical user interface (Figures 1 and 2 above) and is detailed in the user manual (http://www.datelab.org). DateLab is user-friendly and produces high-quality graphical output (for examples see Figures 3 and 4) and detailed reports. Examples of standard reports can be seen at http://www.datelab.org.

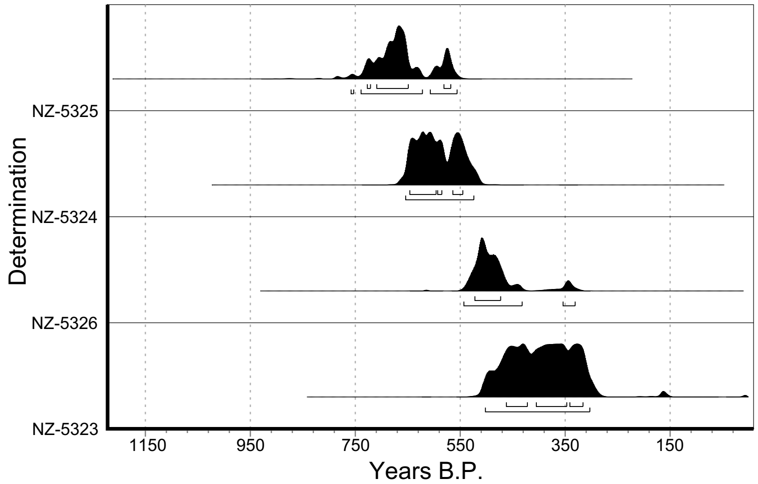


Figure 3 Combined histogram output from DateLab for the likelihoods (standard calibrated distributions) of the CRAs given in Table 2

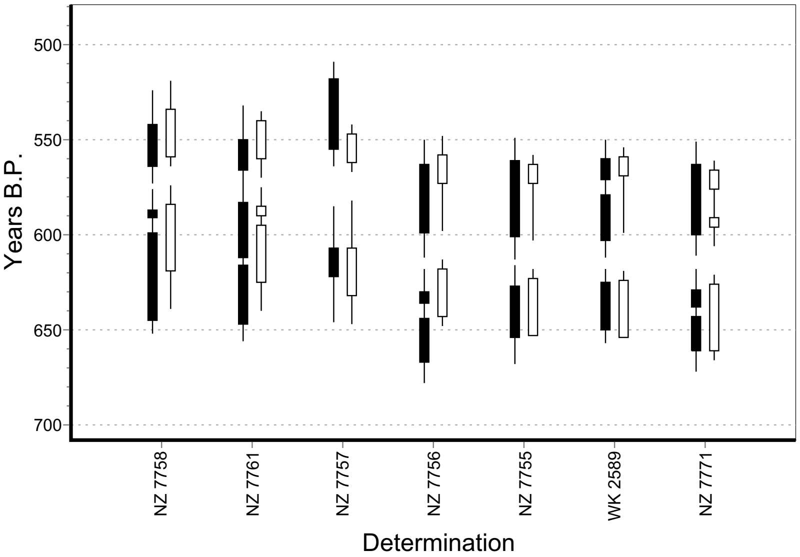


Figure 4 Posterior bar graph for the Shag Mouth data under $\aleph_1$: The solid bars represent the likelihoods (standard calibrated distributions) and the white bars represent the posterior distribution.

DateLab is currently only available for Win-32 systems. Future versions of the full DateLab system will be platform independent. The basic DateLab functionality is as follows:

1. Basic calibration of independent CRAs in the same fashion as found in packages such as Calib or Oxcal.
2. Calibration of dates using correlated reservoir offsets as discussed in Jones and Nicholls (2001) and Nicholls and Jones (1998, 2001).
3. Bayesian calibration of serial phases of dates following the methodology outlined by Nicholls and Jones (1998, 2001). Nicholls and Jones (1998, 2001) define a new class of prior models for

Bayesian calibration that are in general more appropriate than those used in existing software packages.

4. Sampling of the posterior via either rejection or MCMC analysis. As already discussed, rejection is an optimal sampling approach. However, rejection will be too slow for some problems and in this case a Metropolis-Hastings MCMC analysis following that given in Nicholls and Jones (1998, 2001) is implemented.
5. Model comparison via Bayes factors. In applying Bayesian analysis, it is often the case that a number of plausible models could be used. DateLab offers a sound method to objectively compare the relative likelihood of different models.
6. Results of analysis are summarized as intelligible documents in either HTML, RTF, or LaTeX formats with multiple graphics formats supported.

In the following, we detail the mathematical framework underlying the analyses described above.

Models

The Bayesian Calibration model used in DateLab is a direct representation of the relative chronometric information imposed by stratigraphic constraints in an archaeological excavation (see Nicholls and Jones 1998, 2001 for a detailed description and motivation for this model). All dates are treated as coming from one of a number of phases that occur as a single series. Within phases there are no prior constraints on the relative age of any of the dates. However, we know *a priori* the relative ordering of the phases and add a further constraint that there is no overlap of the phases. While this can be extended to analyze multi-phase models where there is complete independence between the phases, more general phase models, of the kind applied in Zeidler et al. (1998), cannot currently be analyzed using DateLab.

For further discussion of the DateLab model it is necessary to define the following notation. Dates are regarded as arising from a single series of M abutting phases. N_m ^{14}C age determinations are gathered from phase m, making $K \equiv \sum N_m$ dates in all. For $n \in \{1,2... N_m\}$ let $y_{m,n}$ denote the value of the n'th ^{14}C age measured in the m'th phase, reported with associated standard error $\sigma_{m,n}$. For all quantities $X_{m,n}$ let X denote the corresponding vector in the natural ordering, so that $y \equiv (y_{1,1},..., y_{M,NM})$, etc. Let $\theta_{m,n}$ be a calibrated date for specimen (m,n), with units calendar years AD, and assumed to equal the context date associated with the (m,n)'th specimen. For $m \in \{0,1... M\}$ let ψ_m denote the boundary date at the lower boundary of phase m. We have a total $K+M+1$ unknown parameters: the $M+1$ layer boundary dates $\psi_0... \psi_M$, and the K unknown object dates, $\theta_{1,1}... \theta_{M,NM}$. Let P and A, $P \leq A$ be given termini, setting lower and upper bounds on the dates. Possible parameter sets (ψ,θ) take some value in a parameter space Ω. This space is simply the set of all states (ψ,θ) satisfying the stratigraphic constraints:

$$\Omega \equiv \{(\psi,\theta);\ \mathrm{P} \leq \psi_{\mathrm{M}} \leq \theta_{\mathrm{M},.} \leq \psi_{\mathrm{M}-1} \leq ... \leq \psi_1 \leq \theta_{1,.} \leq \psi_0 \leq \mathrm{A}\}$$

Following the standard Bayesian inferential framework, the posterior distribution of Ψ and θ conditional upon the observed dates y (with density $h\ (\Psi, \theta \mid y)$) is defined in terms of an unnormalized prior density $f\,(\Psi, \theta)$, and likelihood L (y, θ), as in Equation 1, as

$$h\ (\theta, \Psi \mid y) = L\ (y \mid \theta) \times f\,(\Psi \mid \theta) \tag{2}$$

We now outline the likelihood employed in DateLab and the family of prior models that DateLab is presently able to analyze.

Likelihood

The likelihood $L\ (y \mid \theta)$ used here follows the standard definition of the ^{14}C likelihood (e.g. Buck et al. 1991). The observation model for CRA $y_{m,n}$ is

$$y_{m,n} \sim \text{Normal}\ (\mu(\theta_{m,n}),\ \sigma(\theta_{m,n})^2\ )$$

where

$$\sigma(\theta_{m,n})^2 \equiv \hat{\sigma}(\theta_{m,n})^2 + \tilde{\sigma}^2{}_{m,n}$$

and $\mu(\theta_{m,n})$ and $\tilde{\sigma}\ (\theta_{m,n})$ are standard, empirically determined ^{14}C calibration functions (e.g. Stuiver et al. 1998). For DateLab we use the INTCAL98 calibration data available from http://depts.washington.edu/qil/ in decadal tabulation. We spline these decadal values so that μ and σ^2 are functions piecewise linear by year. When a specimen (m,n) is of terrestrial origin, terrestrial calibration functions are used. Otherwise, the marine calibration functions are used. Material type dependence is implicit in our notation. Let $\ell(y_{m,n} \mid \theta_{m,n})$ denote the likelihood of parameter $\theta_{m,n}$,

$$\ell(y_{m,n} \mid \theta_{m,n}) = \frac{1}{\sigma(\theta_{m,n})\sqrt{2\pi}} \exp\left(-(y_{m,n} - \mu(\theta_{m,n}))^2 / 2\sigma(\theta_{m,n})^2\right)$$

Thus $\ell(y_{m,n} \mid \theta_{m,n})$ is a density distribution normalized over $y_{m,n}$ values, and unnormalized over $\theta_{m,n}$ values. Observations are assumed independent, so the joint likelihood, $L\ (y \mid \theta)$, is

$$L(y \mid \theta) = \prod_{m=1}^{M} \prod_{n=1}^{N_m} \ell(y_{m,n} \mid \theta_{m,n})$$

In the case that a correlated reservoir offset (Jones and Nicholls 2001) is used the likelihood needs to be modified. For details, see Nicholls and Jones (1998, 2001).

Prior

The prior density, $f(\psi,\theta)$, summarizes our state of knowledge before the ^{14}C determinations are available. It is natural to model the object dates θ conditional on the layer boundary dates ψ, so we break our prior up in two pieces,

$$f(\psi,\theta) = f_{\Theta|\Psi}(\theta \mid \psi) f_{\Psi}(\psi)$$

In the absence of ^{14}C age determinations, the age parameters $\theta_{m,n}$ might take any value between $\psi_{m-1,n}$ and $\psi_{m,n}$ with equal probability, a state of knowledge represented by the choice

$$f_{\Theta|\Psi}(\theta \mid \psi) = \prod_{m=1}^{M} \frac{1}{(\psi_{m-1} - \psi_m)^{N_m}}$$

for (ψ,θ) restricted to Ω. What prior density $f_\Psi(\psi)$ should we take for the set ψ of phase boundary event dates? A set of ψ values is "legal" if $P \le \psi_M \le \psi_{M-1} \ldots \le \psi_0 \le A$. It seems natural to say "any legal set of dates ψ is *a priori* equally likely", and we will call this choice, $f_\Psi(\psi) = 1$, the uniform prior density. Surprisingly enough, the uniform density for ψ weights the prior in favor of more widely spread sets of dates, and this can bias the whole analysis, though the effect is often slight. The date span, $\delta(\psi) \equiv \psi_0 - \psi_M$, which measures the number of years spanned by dated strata, exhibits the bias: it can be shown that, under the uniform prior density, a span of 2 δ is favored over a span of δ by a factor of approximately 2^{M-1} (the approximation is good when $\delta \ll R$, where $R \equiv A-P$). Nicholls and Jones (1998, 2001) give several reasons for favoring the choice

$$f(\psi,\theta) = \frac{1}{(R-\psi_0-\psi_M)} \frac{1}{(\psi_0-\psi_M)^{M-1}} \tag{3}$$

the most important being, that this is simply more representative of the typical state of knowledge prior to the arrival of the data. It is non-informative for $(\psi_0 - \psi_M)$, the span of events. The prior in Equation 3 is akin to the prior belief that "any span value is equally likely, and then any legal set of dates ψ is equally likely given the span". Equation 3 can be motivated in a number of ways. It may be derived by taking the simplest plausible physical model of the specimen deposition process, assuming constant deposition rates in layers, and allowing random thinning of deposited specimens. The suitability of the model choice expressed in Equation 3 over the constant prior may be tested, for any particular data set, using the model comparison tools described below.

Note that the prior density is not normalizable when no *terminus post quem* can be established, since $P = -\infty$ in that case. A prior with no finite normalization is said to be *improper*. Sample-based Bayesian inference with an improper prior is meaningful if the posterior density is normalizable, which is the case whenever $\sigma(\theta_{m,n}) < \infty$, under mild conditions on μ, the calibration function. Rejection sampling is infeasible when a very conservative P is asserted.

SAMPLING PROCEDURES

Sample-based inference is a numerical mode of analysis that allows us to form summarizing statements from the posterior density, i.e. integrate out marginal posterior distributions of interest. The inference is quite straightforward. The probability assertion X is the case, given the data and prior knowledge represented in the posterior, is estimated by sampling parameter sets (ψ,θ) from the posterior distribution, and then calculating the proportion of samples in which the event X occurs.

For example, a scientist may propose that the number of yr spanned by the modeled phases, $\delta(\psi) \equiv \psi_0 - \psi_M$, is less than 100 yr. In that case, S is the set $S = \{(\psi,\theta): \delta(\psi) \leq 100)\}$ and we are interested in $\Pr\{(\Psi,\Theta) \in S \mid y\}$, which stands for the probability the scientist is correct, given the data, and any other substantial knowledge quantified in the prior distribution. Let

$$Z^h = \int_\Omega h(\psi,\theta|y) d^{(M+1)}\psi d^{(k)}\theta$$

denote the normalizing constant for the posterior density $h(\psi,\theta|y)$. Then, in terms of $h(\psi,\theta|y)$, the probability we have to estimate is

$$\Pr\{(\Psi,\Theta) \in S|y\} = \frac{1}{Z^h} \int_{S\cap\Omega} h(\psi,\theta|y) d^{(M+1)}\psi d^{(k)}\theta \tag{4}$$

Instead of calculating the integral given in Equation 4 in closed form, we estimate its value by Monte Carlo integration. The posterior probability for "the span is less than 100 yr" is estimated by the proportion of samples from the posterior distribution in which $\psi_0 - \psi_M \leq 100$. Similarly, histograms of sampled parameter sets may be used to summarize marginal posterior probability distributions.

The problem then is to generate samples from the posterior. In general, this is done using Markov chain Monte Carlo (MCMC) algorithms, which employ Gibbs sampling or some more general Metropolis-Hastings algorithm. There is a problem here. The output of MCMC algorithms is not in general guaranteed to have the desired (posterior) distribution. A sufficient condition for MCMC convergence to equilibrium, which may readily be checked, does not exist. In many applications, it can be established with reasonable confidence that the output is correctly distributed. Unfortunately, in the case of ^{14}C calibration, with a posterior density of the kind we have defined, it is particularly difficult to get Markov Chain Monte Carlo algorithms to converge reliably.

Thus, close attention needs to be paid to the output from MCMC analyses to ensure that the results are correct. However, for reasonably small data sets, the rejection algorithm given by Nicholls and Jones (1998, 2001) is suitable. This sampling algorithm has the advantage that, when it returns a sample, that sample is returned with a probability density that coincides with $h(\psi,\theta \mid y)$ up to machine precision. Moreover, samples are independent, so estimation of standard errors is straightforward. The main disadvantage of rejection sampling, compared with MCMC in general, is that it is very slow for certain types of calibration problems: roughly speaking, those in which the number of dates is large. DateLab implements both the rejection and Metropolis-Hastings MCMC sampling algorithms of Nicholls and Jones (1998, 2001). While the Metropolis-Hastings MCMC sampling routine is much faster than rejection, rejection sampling is the preferred approach. Unfortunately, under some sampling problems the rejection routine is too slow to be practical, and we are obliged to fall back on less reliable MCMC methods.

Model Comparison

When we carry out Bayesian calibration, it is usual that there will be several prior models that we may realistically apply to the data. In this case it is useful to be able to make a comparison between the competing models. For example, this allows us to address questions such as do these dates actually come from sequentially ordered strata (i.e. are the dates from a primary context) or has the material been mixed. In DateLab, model comparison is based around Bayes factors.

Methodology

Bayesian model comparison is based on a quantity called the Bayes factor. This quantity plays a similar role, in Bayesian inference, to the p-value of frequentist inference. Suppose $\aleph_1$ and $\aleph_0$ are 2 models, and the Bayes factor for Model $\aleph_1$ over model $\aleph_0$ is some number B. The analysis is telling us that model $\aleph_1$ is B times more probable than model $\aleph_0$, in the light of the data. This statement may be interpreted quite literally. See the examples below. Table 1, taken from Raftery (1996), gives a standard interpretation of the Bayes factor (see Raftery 1996 for further discussion and references).

We now define the Bayes factor in more detail. For $i \in \{0,1\}$, let $f(\psi,\theta \mid \aleph_i)$ be the unnormalized prior density under model M_i, with normalizing constant Z_i^f. Let Ω_i denote the space of parameters in the ith model. The mean likelihood, $f_{Y|\aleph_i}(y \mid \aleph_i)$, for the data under model i is

$$f_{Y|\aleph_i}(y \mid \aleph_i) = \frac{1}{Z_i^f} \int_{\Omega} L(y \mid \theta) f(\psi,\theta \mid \aleph_i) d^{(M+1)}\psi d^{(k)}\theta$$

DateLab uses the method outlined by Meng and Wong (1996) to calculate the mean likelihood. Details of the algorithm employed are given in Nicholls and Jones (1998, 2001).

The Bayes factor for comparison of models is

$$B(1v.0) \equiv \frac{P(y|\aleph_1)}{P(y|\aleph_0)}$$

On the basis of this, we can say Model 1 is $B(1v.0)$ more times likely than Model 0. The level of support this indicates for Model 1 is shown in Table 1.

Table 1 Interpretation of Bayes factor $B(1v.0)$, from Rafterty (1996)

$B(1v.0)$	Support for $\aleph_1$
<1	Supports $\aleph_0$
1 to 3	Barely worth mentioning
3 to 12	Positive
12 to 150	Strong
>150	Very strong

Examples

To demonstrate the use of Bayes factors we consider 2 archaeological date sets in Tables 2 and 3.

Table 2 Charcoal dates from Dart River Mouth, New Zealand (Anderson and Ritchie 1986; Simmons 1973). Column $y_{m,n}$ lists standard ^{14}C determinations, with no corrections. Column (m,n) lists m, the layer index and n, the specimen index within a layer. See text.

Date nr	(m,n)	$y_{m,n}$	$\sigma_{m,n}$
NZ 5323	(1,1)	337	55
NZ 5326	(1,2)	442	41
NZ 5324	(3,1)	587	56
NZ 5325	(3,2)	723	57

Table 3 Charcoal dates from Shag River Mouth, New Zealand (Anderson et al. 1996). The column $y_{m,n}$ lists standard ^{14}C determinations, with no corrections. Column (m,n) lists m, the layer index and n, the specimen index within a layer. See text.

Date nr	(m,n)	$y_{m,n}$	$\sigma_{m,n}$
NZ 7758	(1,1)	580	47
NZ 7761	(2,1)	600	50
NZ 7757	(3,1)	537	44
NZ 7756	(4,1)	670	47
NZ 7755	(5,1)	646	47
WK 2589	(5,2)	630	35
NZ 7771	(6,1)	660	46

In Table 2, we list a set of K=4 charcoal dates from the Dart River Mouth site (Anderson and Ritchie 1986; Simmons 1973), southern New Zealand. Anderson and Ritchie (1986) suggest that 2 discrete occupation phases are represented in the Dart River archaeological record, with 2 of the dates presented in Table 2 deriving from each of these proposed phases. A real question is, do the dates support this model? A graph of the calibrated date distributions (Figure 3) does not make it obvious that 2 phases of occupation would necessarily be favored over a single longer-term phase of activity. We will compare the model where the phase structure suggested by Anderson and Ritchie (1986) is imposed ($\aleph_1$) with the model in which the dates are regarded as arising from a single phase of occupation ($\aleph_0$). The form of the prior density is the non-informative density $f(\psi,\theta)$ specified above. Under model $\aleph = \aleph_1$, M=3, and the parameter space is

$$\Omega_1 \equiv \{(\psi,\theta); P \le \psi_3 \le \theta_{3,.} \le \psi_2 \le \psi_1 \le \theta_{1,.} \le \psi_0 \le A\}$$

while under model $\aleph=\aleph_0$, we have $M = 1$, and

$$\Omega_0 \equiv \{(\psi,\theta); P \le \psi_1 \le \theta_{M,.}, \theta_{M-1}, \ldots\ \theta_{1,.} \le \psi_0 \le A\}$$

Computing the mean likelihood under each model with A = 100 BP and P =1000 BP we report

$$P(y|\aleph_0) = 1.1(1)\times 10^{-12} \qquad P(y|\aleph_1) = 5.2(2)\times 10^{-12}$$

Throughout this report, the quantity quoted in brackets is a standard error for the last reported digit. Here, the mean likelihoods have been estimated sufficiently accurately to determine their relative magnitude. $B(1v.0) = 4.7(2)$, which means that the occupation phase sequence described by Anderson and Ritchie is around 5 times more likely in light of the available dates than the suggestion that there is simply a single phase of occupation. From Table 1, we can state that the Bayes factor analysis provides positive support for the two occupation phase model described by Anderson and Ritchie (1986) in contrast to a single occupation phase model.

In the 2nd example, we consider a set of 7 dates from a single series of strata (Table 3, Figure 4). Do the dates support the assertion that the strata are primary context for the dated artifacts? Many stratigraphic models might be constructed. Each model constrains the θ in different ways. We will compare the model with the full set of stratigraphic constraints $\aleph_1$ with the model in which all stratigraphic constraints have been removed $\aleph_0$: a result significantly in favor of $\aleph_0$ would be sufficient grounds to reject $\aleph_1$, and thereby reject the strata as primary context. The form of the prior density is the non-informative density $f(\psi,\theta)$ specified above. Under model $\aleph = \aleph_1$, M=6, the N_m m=1,2... 6 are given by the data in Table 3, and the parameter space is

$$\Omega_1 \equiv \{(\psi,\theta); P \le \psi_M \le \theta_{M,.} \le \psi_{M-1} \le \theta_{M-1} \le \ldots\ \psi_1 \le \theta_{1,.} \le \psi_0 \le A\}$$

while under model $\aleph = \aleph_0$, we have M=1, N_1=K=7 and

$$\Omega_0 \equiv \{(\psi,\theta); P \le \psi_1 \le \theta_{M,.}, \theta_{M-1}, \ldots\ \theta_{1,.} \le \psi_0 \le A\}$$

Computing the mean likelihood under each model with A = 100 BP and P = 1000 BP we report

$$P(y|\aleph_0) = 1.3(1)\times 10^{-18} \qquad P(y|\aleph_1) = 1.7(2)\times 10^{-18}$$

Since $B(1v.0) = 1.3(2)$, there is evidence in favor of $\aleph_1$. Thus, the data provides some support for the proposed stratigraphic sequence, however it is inconclusive. Certainly, there is no reason to reject the notion that the observed stratigraphic sequence observes temporal superposition.

DATELAB OUTPUT

DateLab produces a standard output report of each analysis run. This is produced as an HTML document by default, however LaTeX and RTF versions can also be produced. The reports are selfexplanatory, and consist of a number of pages of summary analysis and output depending upon the type of analysis that has been performed. All reports contain:

- Details of the Analysis Models that have been applied.
- Summary reporting of analysis statistics (such as the mean Likelihood, HPDs etc.).
- Graphical output of analysis results in a range of formats.

Readers are encouraged to view the examples on the DateLab website (www.datelab.org).

Graphics

DateLab produces 2 basic forms of graphics, histograms (e.g. Figure 3) and bar graphs (e.g. Figure 4), which can be combined or presented individually. Histograms are a standard graphic for presentation of calibrated distributions and are well understood. However, histograms are not entirely suitable for presenting large numbers of calibrated distributions in a combined graph. While combined histograms such as those given in Figure 3 are useful, this type of graph rapidly becomes unmanageable for a large number of dates. Combined bar graphs, such as Figure 4, are more satisfactory for this type of data. In these plots the boxes represent the 68% highest posterior density (HPD) for the distribution (roughly analogous to the 1-sigma interval) and the lines represent the 95% HPD. Other types of graph are produced where a different format is more appropriate.

All images can be output to GIF, EMF, EPS, TIFF, or BMP versions. Full details are given in the manual.

CONCLUSION

DateLab is a simple software package that offers a limited range of Bayesian calibration models. However, DateLab can analyze many common CRA datesets and has been designed to produce robust, high-quality results and output. DateLab will be a useful tool for researchers wishing to perform simple calibration of CRA data or Bayesian calibration of serially ordered date sets, especially when the problem allows rejection sampling to be performed at a reasonable speed.

The DateLab software may be obtained via the internet from http://www.datelab.org.

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NATURAL ABUNDANCES OF CARBON ISOTOPES (^{14}C, ^{13}C) IN LICHENS AND CALCIUM OXALATE PRUINA: IMPLICATIONS FOR ARCHAEOLOGICAL AND PALEOENVIRONMENTAL STUDIES

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ABSTRACT. Radiocarbon ages of calcium oxalate that occurs naturally on rock surfaces have been used recently in archaeological and paleoenvironmental studies. Oxalate rock coatings are found globally, with most appearing to be residues from epilithic lichens. To explore the source(s) of carbon used by these organisms for the production of oxalate we measured the natural abundances of ^{14}C and ^{13}C in 5 oxalate-producing lichen species, 3 growing on limestone in southwestern Texas and 2 on sandstone in Arkansas. We also examined the distribution of the isotopes between the calcium oxalate and lichen tissues by separating these components and measuring the $^{13}C/C$ independently. The results demonstrate that the limestone species were slightly enriched in ^{14}C, by 1.7‰, relative to the sandstone species, which suggests that "dead" carbon from the limestone substrate does not constitute a significant source of carbon for the production of oxalate. The calcium oxalate produced by the lichens is also enriched in ^{13}C by 6.5‰ compared to the lichen tissues, demonstrating that there is a large carbon isotope discrimination during oxalate biosynthesis. These results support the reliability of ^{14}C ages of calcium oxalate rock coatings used for archaeological and paleoclimate studies.

INTRODUCTION

It is becoming increasingly evident that the calcium oxalate minerals whewellite ($CaC_2O_4 \cdot H_2O$) and weddellite ($CaC_2O_4 \cdot (2 + x)H_2O$) are common on rock surfaces worldwide (Table 1). The oxalate generally occurs as thin ($\lesssim$1 mm) rock patinas with most appearing to be deposits from epilithic lichens (Del Monte et al. 1987; Edwards et al. 1993; Russ et al. 1996; Hofmann and Bernasconi 1998), although unlichenized microbes (Bonaventura et al. 1999) and organic acid aerosols (Watchman 1991) have been proposed as sources of some oxalates on rock surfaces. Recently, radiocarbon dates of oxalates that cover, encapsulate, or are incorporated within prehistoric rock paintings (pictographs) have been used to constrain or estimate the ages of the artifacts (Watchman 1993; Hedges et al. 1998; Russ et al. 1999; Watchman et al. 2000; Steelman et al. 2001). Oxalate ^{14}C ages have also been used in paleoclimate reconstructions that are based on the assumption that temporal variations in oxalate production can be correlated to fluctuations in lichen productivity in response to climate change (Russ et al. 1996; 2000).

The source(s) of carbon that leads to the formation of oxalate coatings is unknown, although assumed to be ambient CO_2. Here we report a study of the relative abundances of ^{14}C and stable carbon isotopes in living, oxalate producing (pruinose) lichens that allowed us to address (1) whether lichens incorporate significant levels of limestone (carbonate) carbon for the production of oxalate and (2) the distribution of carbon isotopes between the lichen tissues and oxalate coating (pruina).

Lichens are symbiotic associations between fungi (mycobiont) and photosynthetic microbes (photobiont) integrated within the fungal matrix (thallus). Calcium oxalate is the most common lichen byproduct, which generally occurs as a coating on the upper/outermost surface of the organism. The production of oxalate might benefit lichens by removing excess calcium ions and/or providing an external source of water stored in the calcium oxalate crystal lattice (Wadsten and Moberg 1985). After the death of the organism, the oxalate pruina can remain stable on the rock surface for millennia (Watchman 1993; Russ et al. 1996).

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Table 1 Summary of occurrences of calcium oxalate rock coatings

Location	Association	Proposed origin	$\delta^{13}C$ (‰)	Reference
Italy	Coatings on ancient monuments, old buildings & rock surfaces	Lichens	NA	Del Monte et al. 1987
Australia	Natural coatings on pictographs & rock surfaces	Organic acids in rain	NA	Watchman 1991; Watchman et al. 2000.
Italy	Encrustations on Renaissance frescoes	Lichens	NA	Edwards et al. 1993, 1997
California (USA)	Natural coatings on pictographs & rock surfaces	None proposed	NA	Scott and Hyder 1993
Utah (USA)	Natural coating on a single pictograph	None proposed	NA	Chaffee et al. 1994
SW Texas (USA)	Natural coating on pictographs & rock surfaces	Lichens	−10.6 ± 1.9 (18)[a]	Russ et al. 1996; 2000
Northern Mediterranean	Coatings on ancient monuments, buildings & rock surfaces	Various mechanisms	NA	Various authors 1996
Argentina	Component in pictograph paint	Cacti used in paint recipe	−10.3 (1)[a]	Hedges et al. 1998
Argentina	Natural rock coating	None proposed	−26 (2?)[a]	Hedges et al. 1998
Arizona (USA)	Natural rock coating	Lichens	−11.8 (1)[a]	Hofmann and Bernasconi 1998
Switzerland	Natural rock coating	Lichens	−11.7 (1)[a]	Hofmann and Bernasconi 1998
Italy	Stone monuments	Microbes	NA	Bonaventura et al. 1999
Brazil	Natural coating on pictograph	None proposed	−11.67 (1)[a]	Steelman et al. 2002

[a] Number of individual analyses

The reliability of ^{14}C ages and stable carbon isotope ratios ($\delta^{13}C$) from oxalate deposits from lichens depends largely on whether or not inorganic carbon (carbonate or bicarbonate ions) from the basal rock is incorporated in the oxalate. Native carbon in ancient carbonate rocks such as limestones is completely depleted in ^{14}C, and enriched in ^{13}C by ~8‰ compared to atmospheric CO_2 (Craig 1953; Degens 1969). If such "dead" carbon from the substrate is included in the oxalate, either via biosynthesis, exchange reactions, or reactions of oxalic acid at the rock surface, then oxalate ^{14}C ages would be anomalously old and $\delta^{13}C$ values would represent a ^{13}C enrichment independent of metabolic processes.

Indirect evidence that lichen mycobionts can metabolize carbonate/bicarbonate ions was demonstrated by Lapeyrie et al. (1987; Lapeyrie 1988) by showing that oxalate ion production by the fungus *Paxillus involutus* was greater when grown on calcareous soil compared to acidic soil, and that this particular fungus incorporated bicarbonate ions from a growth medium for the biosynthesis of oxalic acid. Additional evidence of a possible bicarbonate effect is that calcium oxalate rock coatings are significantly enriched in ^{13}C compared to living lichens (Table 1). For example, Hofmann and Bernasconi (1998) reported $\delta^{13}C$ values of −11.8‰ and −11.7‰ from analyses of oxalate rock crusts in Arizona (USA) and Valais, Switzerland, respectively, while Steelman et al. (2001) reported an oxalate $\delta^{13}C$ value of −11.67‰ from a coating in Toca do Bastina, Brazil. Furthermore, 18 calcium oxalate crust samples from 12 sites in southwestern Texas yielded a mean $\delta^{13}C$ value of −10.6 ± 1.9‰ (Russ et al. 2000). Reported $\delta^{13}C$ values of living lichens, on the other hand, range from −35‰ to −14‰ (Lange et al. 1988), indicating a significantly lower ^{13}C content compared to the oxalate coatings. This isotope discrepancy might be due to utilization of carbonate in the rock substrate that would cause ^{14}C measurements to be unreliable, or other metabolic processes for which corrections can be made. The latter is the case for the intracellular calcium oxalate in cacti which is enriched in ^{13}C by ~5‰ compared to the cactus tissues (Rivera and Smith 1979).

To investigate whether lichens incorporate substrate carbonate and/or bicarbonate ions for the production of oxalate we measured the relative abundances of the carbon isotopes (Δ^{14}C and δ^{13}C values) in 5 living, pruinose lichen species collected from limestone and sandstone surfaces in Texas and Arkansas, respectively (Table 2). We sampled areas on the specimens that appeared to have recent growth—edges and areas with fruiting bodies—and so expected Δ^{14}C values that reflected contemporary atmospheric CO_2. Then, if substrate carbon was incorporated in the oxalate the ^{14}C/C of the limestone species would be lower (smaller Δ^{14}C values) and the ^{13}C/C higher (less negative δ^{13}C values) compared to the lichens growing on the sandstone. However, as Bench et al. (2001, 2002) recently demonstrated, there is considerable internal carbon recycling and/or carbon turnover in the 2 lichens they studied. Such processes would cause a discrepancy between the contemporary atmospheric ^{14}C record and the ^{14}C content of the organisms, and limit our ability to predict the relative amount of carbonate carbon, if any, included for the production of oxalate.

Table 2 Radiocarbon (Δ^{14}C) and stable carbon isotope (δ^{13}C) results from pruinose lichens in southwestern Texas and northeastern Arkansas

Species	Location	Substrate	AMS lab nr	Δ^{14}C	δ^{13}C (‰)
Flavoparmelia baltimorensis	NE Arkansas	Sandstone	50964	115.3 ± 5.0	–23.65
Dirinaria frostii	NE Arkansas	Sandstone	50970	142.7 ± 4.9	–22.13
Caloplaca saxicola	SW Texas	Limestone	50966	206.6 ± 5.6	–17.07
Caloplaca saxicola	SW Texas	Limestone	50967	219.1 ± 5.7	–17.76
Caloplaca saxicola	SW Texas	Limestone	50968	189.9 ± 5.5	–20.33
Lecania Sp.	SW Texas	Limestone	50969	162.3 ± 5.4	–17.92
Lecania Sp.	SW Texas	Limestone	AA42664	192.5 ± 7.1	–18.22
Lecidea Sp.	SW Texas	Limestone	AA42662	163.6 ± 6.3	–18.93

We also explored the distribution of the stable carbon isotopes between the lichen tissues and oxalate pruina by separating these components and measuring the δ^{13}C values of each. There is greater variability in δ^{13}C values reported for lichens than for higher plants—including both C_3 and C_4 plants—despite all lichen photobionts using the C_3 metabolic pathway. The δ^{13}C of lichens is governed primarily by moisture conditions, specifically the amount and phase of water required to activate and maintain photosynthesis. Three categories of lichens have been identified based on carbon isotope compositions, and which is related to the water requirements of the different photobionts whether cyanobacteria (cyanobionts), green algae (phycobionts) or a combination of both (photosymbiodemes). Phycobionts, for example, can initiate photosynthesis and reach maximum activity with lower water contents and when the water source is vapor alone (high humidity, dew or fog). Cyanobionts, on the other hand, require considerably more water and in the liquid phase (Lange et al. 1986; Lange et al. 1988). Fractionation of the carbon isotopes is induced by diffusion resistance of CO_2 through water-filled membranes (Lange et al. 1988) and/or structural changes in the photobiont and mycobiont cells caused by hydration and dehydration processes (Scheidegger et al. 1995). The presence of a carbon concentrating mechanism (CCM) employed by cyanobionts and some phycobionts has also been deduced, and that influences the overall isotopic composition of these particular organisms. Thus, lichens with cyanobionts (and phycobionts with a CCM) are "C_4-like" with δ^{13}C values $\gtrsim$–23‰; phycobionts without a CCM are more "C_3-like" with δ^{13}C values $\lesssim$ –24‰; and photosymbiodemes have the lightest isotopic composition, with δ^{13}C values ~ –33‰ (Máguas et al. 1993, 1995; Smith and Griffiths 1996).

MATERIALS AND METHODS

Lichen samples were collected from southwestern Texas (29°53′N, 100°54′W) and northeastern Arkansas (92°45′N, 36°12′W) by removing a portion of the basal rock with the lichens intact. Subsamples were sent for identification to B Ryan (Arizona State University). To prepare for the analyses the sample surfaces were rinsed using E-pure (18.2 Mega Ohm/cm) water to remove loose detritus then dried in a 90 °C oven. A small (~1 mg) aliquot of the specimen was removed and analyzed using Fourier transform infrared analysis (FTIR) to establish the presence of oxalate.

AMS $\Delta^{14}C$ and $\delta^{13}C$ Analyses

^{14}C and stable carbon isotope ratio analyses were performed on 5 lichen species, 3 collected from limestone surfaces in southwestern Texas and 2 growing on sandstone in northeast Arkansas (Table 2). Samples were prepared by removing ~2 cm^2 of the lichen from the substrate with a dental pick, followed by grinding using an agate mortar and pestle. Approximately 150 mg of the powdered sample was placed in a Teflon beaker and 40 mL of dilute phosphoric acid (pH ~2.4) added to remove carbonates. The solution was maintained at pH <3.0 by drop-wise addition of concentrated phosphoric acid, and allowed to stand for ~48 hr with intermittent stirring. The sample was filtered using a micropore glass filter (10–15 μm) and the filtrate consisting of the lichen tissue and calcium oxalate pruina dried at 90 °C. Each sample was split, with one aliquot used for the accelerator mass spectrometry (AMS) ^{14}C measurement and the other for the stable carbon isotope analysis.

The AMS measurements were performed at either the Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Laboratories or the University of Arizona NSF-Arizona AMS Facility. Samples were further processed for the AMS assay by combusting the powder to CO_2 at 950 °C in the presence of CuO, and graphite targets produced using standard protocol (Vogel et al. 1987). The $\delta^{13}C$ analyses were performed using a Finnigan Delta Plus isotope ratio mass spectrometer (IRMS) interfaced with a Carlo Erba EA-1108 elemental analyzer; samples were combusted to CO_2 in the elemental analyzer, and the isotopic composition of the CO_2 was determined by continuous flow IRMS. $\delta^{13}C$ values are expressed relative to the V-PDB standard, and precision was $\leq 0.1‰$.

$\delta^{13}C$ Analysis of Separated Oxalate Pruina and Lichen Tissues

As above, the lichens were removed from the substrate using a dental pick and ground in an agate mortar and pestle. Approximately 100 mg of the powdered lichen was placed in a Teflon beaker along with 30 mL of 1.5 N HCl to remove carbonates and dissolve the calcium oxalate. The solution was stirred at 80 °C for 2 hr, then allowed to stand overnight to completely dissolve the calcium oxalate. The acid insoluble tissue, mainly thallus, was isolated from the solution by filtering through a micropore (10–15 μm) glass filter, and the solid residue dried at 90 °C.

We precipitated the calcium oxalate from the solution under a stream of N_2 to prevent contamination from atmospheric CO_2 by first neutralizing the mother liquor with boiling 3 N NaOH then adding 3 mL of saturated $CaCl_2$ solution. The calcium oxalate precipitate was filtered using a micropore (4– 8 μm) glass filter and dried at 90 °C. The stable carbon isotope ratios of each component was measured as described above. This method was tested via 5 separate trials of a single homogenized lichen (*Lecania* sp.) sample, measurements of 3 different areas of a single *Lecania* specimen, and experiments using a calcium oxalate standard.

RESULTS AND DISCUSSION

All lichens used in this study had calcium oxalate pruina, as established using FTIR. Another common feature was that each species contained green algal photobionts.

$\Delta^{14}C$ Values of Lichens Growing on Limestone and Sandstone

The AMS ^{14}C results show that the mean $\Delta^{14}C$ value of the sandstone species ($\Delta^{14}C = 129.0 \pm 19.4‰$) is similar to that of contemporary atmospheric CO_2 (Nydal and Lövseth 1983; Levin and Kromer 1997), while the mean $\Delta^{14}C$ value of the lichens growing on limestone ($\Delta^{14}C = 189.0 \pm 22.8‰$) is enriched in ^{14}C by 60‰ compared to the sandstone species (Table 2). Moreover, the ^{14}C values from the limestone samples are consistently greater than values obtained by Bench et al. (2001; 2002) from analyses of *Caloplaca trachyphylla* (mean $^{14}C = 175.8 \pm 70.5‰$) growing on sandstone and *Rhizocarpon geographicum* (mean $\Delta^{14}C = 152.6 \pm 19.8‰$) growing on siliceous rocks (Table 3). Thus, the limestone specimens have more atmospheric carbon that must have been incorporated during an earlier period when the atmospheric $^{14}CO_2$ concentration was higher (due to bomb ^{14}C production). This could be due to inclusion of older portions of the lichens during the sampling/scraping process or that the recent growth includes considerably more recycled carbon, a phenomenon demonstrated by Bench et al. (2001; 2002) for the 2 lichen species they studied.

Table 3 Average radiocarbon ($\Delta^{14}C$) data from lichens analyzed in this study and reported by Bench et al. (2001, 2002).

Substrate	Nr of species	Nr of measurements	Mean $\Delta^{14}C$ (‰)	Standard deviation (‰)
Limestone (this study)	3	6	189.0	22.8
Sandstone (this study)	2	2	129.0	19.4
Siliceous rocks (Bench et al. 2001)	1	24	152.6	19.8
Sandstone (Bench et al. 2002)	1	44	175.8	70.5

The comparison of the stable carbon isotope ratios between limestone and sandstone species, however, gives evidence that the $\delta^{13}C$ values for lichens growing on limestone are greater than those for lichens growing on sandstone. One interpretation is that the evident enrichment in ^{13}C for the lichen growing on limestone could be due to incorporation of carbonate from the substrate, but the ^{14}C results indicate this interpretation is unlikely. Instead, the difference in the $\delta^{13}C$ values might be attributed to differences in the moisture conditions in which these lichens were collected. Lichens growing in more moist conditions are depleted in ^{13}C compared to lichens growing in relatively drier environments (Shomer-Ilan et al. 1979; Teeri 1981). Because the collection sites in Arkansas (mean annual rainfall ~1200 mm/yr) are considerably wetter than the sites in Texas (mean annual rainfall ~ 450 mm/yr), this difference in the $\delta^{13}C$ values could be expected.

Two-sample t-tests and Wilcoxon rank sum tests were performed to determine if a statistical difference is present in the $\Delta^{14}C$ and $\delta^{13}C$ values for lichen species growing on limestone and lichen species growing on sandstone. A summary of these results follows, with p-values and observed test statistics appearing in Table 4. It should be pointed out that the statistical power of these tests is hindered by the small sample sizes. When sample sizes are small, the size of the effect must be large for it to be evident.

First, the $\Delta^{14}C$ and $\delta^{13}C$ data from the *Caloplaca saxicola* and *Lecania* lichen species growing on limestone were compared. The results suggest there is not a statistically significant difference in the

$\Delta^{14}C$ and $\delta^{13}C$ values among different lichen species growing on the same substrate. This proposition is further supported by comparing the $\delta^{13}C$ values for the different limestone species contained in Table 5. The data provides no evidence of a significant difference between the $\delta^{13}C$ values for the *Caloplaca* and *Lecania* species when comparing their isolated calcium oxalate pruina separated from the lichen tissues, the lichen tissues themselves, nor the $\delta^{13}C_{tis} - \delta^{13}C_{ox}$ differences for the 2 species. This suggests we can pool the $\Delta^{14}C$ and $\delta^{13}C$ values from the different lichen species growing on the same substrate and focus our attention on comparing the limestone values to those from other substrates.

Table 4 Results of statistical analyses: Two-sample t-tests and Wilcoxon rank sum test

		Test statistic		p-value
Comparisons between different lichen species growing on limestone	Location of data	t-test Wilcoxon	Degrees of freedom	t-test Wilcoxon
$\Delta^{14}C$	Table 2	t = 1.6062	1.645	0.275
		W = 11	NA	0.4
$\delta^{13}C$	Table 2	t = –0.3157	2.09	0.781
		W = 10	NA	0.8
$\delta^{13}C$ for isolated calcium oxalate	Table 5	t = –0.2378	3.37	0.826
		W = 50	NA	0.7989
$\delta^{13}C$ for lichen tissues	Table 5	t = –0.3362	7.40	0.7460
		W = 50	NA	0.7986
$\delta^{13}C_{tis}$ - $\delta^{13}C_{ox}$	Table 5	t = 0.0551	9.57	0.957
		W = 55	NA	0.6711

		Test statistic		p-value
Comparisons after pooling values from different lichen species	Location of data	t-test Wilcoxon	Degrees of freedom	t-test Wilcoxon
$\Delta^{14}C$ limestone vs. $\Delta^{14}C$ sandstone	Table 2	t = –3.6253	2.044	0.9669
		W = 33	NA	1
$\Delta^{14}C$ limestone vs. $\Delta^{14}C$ silica	Table 2 and	t = 3.60	7	0.009
	Bench et al. (2001)	—	—	—
$\Delta^{14}C$ limestone vs. $\Delta^{14}C$ sandstone	Table 2 and	t = 0.94	22	0.359
	Bench et al. (2002)	—	—	—
$\delta^{13}C$ limestone vs. $\delta^{13}C$ sandstone	Table 2	t = 5.076	1.831	0.0219
		W = 33	NA	0.0357

The two-sample t-test and Wilcoxon rank sum test did not provide any statistical evidence that the $\Delta^{14}C$ values for lichens growing on limestone are less than the values for lichens growing on sandstone. In fact, the data contained in Table 2 suggest that the average $\Delta^{14}C$ values for lichen growing on limestone is at least 11.7‰ higher than the average $\Delta^{14}C$ for lichen growing on sandstone. A comparison of the $\Delta^{14}C$ values for the limestone species (Table 2) with those for *Rhizocarpon geographicum* growing on siliceous rocks (Bench et al. 2001) and *Caloplaca trachyphylla* growing on sandstone (Bench et al. 2002) was carried out utilizing two-sample t-tests with a Bonferroni correction (Bonferroni 1936; Miller 1981). The results suggest that the average $\Delta^{14}C$ values for lichen growing on limestone is at least 12.5‰ higher than the average $\Delta^{14}C$ for lichen growing on silica, and that there is no significant difference between the $\Delta^{14}C$ values for the limestone and sandstone substrates.

The mean $\Delta^{14}C$ value for the lichens growing on limestone is consistent with atmospheric CO_2 as the sole carbon reservoir if the lichen material was produced within the last 20–25 yr. However, we cannot rule out that even older material was present in the samples, produced when the atmospheric ^{14}C abundance was even greater, and that this was combined with "dead" carbon from carbonate substrate to yield the observed $\Delta^{14}C$ values.

$\delta^{14}C$ Values of Isolated Calcium Oxalate Pruina and Acid-Insoluble Lichen Tissues

One purpose for separating the pruina from living lichens and measuring the $\delta^{13}C$ of the calcium oxalate and lichen tissues independently was to explore why the $\delta^{13}C$ values of calcium oxalate rock coatings are enriched in ^{13}C compared to the values reported in the literature for living lichens. The separation procedure was tested using a calcium oxalate standard, with a mean $\delta^{13}C$ value = –17.42 ± 0.02‰. The mean $\delta^{13}C$ value of the processed standard (–17.72 ± 0.06‰) indicated a 0.30‰ shift in the isotope composition of the treated calcium oxalate.

The results of the analysis of 5 aliquots from a single homogenized *Lecania* sample (oxalate pruina mean $\delta^{13}C$ value = –15.62 ± 0.08‰; lichen tissue mean value = –22.50 ± 0.37‰) demonstrated the method was reproducible (Table 5). The oxalate $\delta^{13}C$ values of the second *Lecania* sample, in which 3 separate areas from the same lichen specimen were removed and analyzed, also confirms the reproducibility of the method (oxalate pruina mean $\delta^{13}C$ value –14.83 ± 0.03‰; lichen tissue mean value = –22.50 ± 0.14‰).

The calcium oxalate pruina proved to be consistently enriched in ^{13}C compared to the lichen tissues. Specifically, the oxalate pruina (mean $\delta^{13}C$ = –15.19 ± 0.92‰) was estimated (with 95% confidence) to be enriched in ^{13}C by between 5.7‰ and 7.3‰ relative to the lichen tissue (mean $\delta^{13}C$ = –21.66 ± 1.15‰). This is even greater than the 5‰ difference between the tissues and intercellular calcium oxalate in cacti reported by Rivera and Smith (1979). It also shows that the $\delta^{13}C$ values of calcium oxalate rock coatings ($\delta^{13}C$ ~ –11.5‰) are consistent with lichen sources with $\delta^{13}C$ values ~ –18‰, well within the $\delta^{13}C$ range reported for lichens (from –35‰ to –14‰; Lange et al. 1988).

Table 5 Stable carbon isotope ratios of lichen oxalate pruina and tissue separates

Species	Sample location	Substrate	Oxalate $\delta^{13}C$ (‰)	Tissue $\delta^{13}C$ (‰)	$\delta^{13}C_{ox} - \delta^{13}C_{tiss}$ (‰)
Lecania	SW Texas	Limestone	–15.62 ± 0.08[a]	–22.50 ± 0.37[a]	6.88
Lecania	SW Texas	Limestone	–14.83 ± 0.03[b]	–22.50 ± 0.14[b]	7.67
Lecania	SW Texas	Limestone	–15.05	–23.42	8.37
Lecania	SW Texas	Limestone	–15.45	–21.06	5.61
Lecania	SW Texas	Limestone	–14.96	–22.61	7.65
Lecania	SW Texas	Limestone	–16.04	–19.52	3.48
Lecania	SW Texas	Limestone	–16.37	–20.44	4.07
Lecania	SW Texas	Limestone	–15.00	–21.90	6.90
Caloplaca	SW Texas	Limestone	–15.22	–21.16	5.94
Caloplaca	SW Texas	Limestone	–14.98	–21.61	6.63
Caloplaca	SW Texas	Limestone	–13.37	–20.36	6.99
Caloplaca	SW Texas	Limestone	–17.30	–22.90	5.60
Verrucaria	SW Texas	Limestone	–13.99	–21.57	7.58
Verrucaria	SW Texas	Limestone	–14.30	–22.20	7.90
Unidentified	SW Texas	Limestone	–15.45	–20.00	4.55
Lecanora	Arizona	Quartzite breccia	–15.03	–22.87	7.84
			–15.19 ± 0.92	–21.66 ± 1.19	6.48 ± 1.47

[a] Average ±1 σ of 5 aliquots from 1 homogenized lichen sample

[b] Average ±1 σ of 3 different regions of a single lichen sample treated independently

CONCLUSIONS

We measured the ^{14}C content of what appeared to be recent growth of 5 oxalate-producing lichen species, 3 of which were growing on limestone in southwestern Texas and 2 on sandstone in northeastern Arkansas. The Δ^{13}C values of the sandstone species were consistent with contemporary atmospheric $^{14}CO_2$ levels, while the limestone species were enriched in ^{14}C by approximately 60‰ compared to those growing on the sandstone. This result is opposite of what was expected if substrate carbonate was a significant source of carbon.

While it might be reasonable to expect that little or no "dead" carbon from the limestone was incorporated by the lichens growing on this substrate, these results do not provide definitive evidence that this is the case. Without knowing the true age of the sampled areas, and thus the actual amount of atmospheric ^{14}C incorporated by organisms, the amount of carbon from the limestone, if any, remains ambiguous.

The stable carbon isotope composition of the calcium oxalate pruina produced by these lichens is 6.5‰ enriched in ^{13}C compared to lichen tissues. Calcium oxalate rock coatings thought to be byproducts of past lichen activity have δ^{13}C values that range from −6.8 to −13.7‰ (Table 1), and thus produced by lichens with δ^{13}C values that range from −13.3 to −20.2‰. These values are consistent with the stable carbon isotope composition of lichens that have cyanobacterial photobionts, or green algal photobionts that employ a CO_2 concentrating mechanism.

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^{14}C DATING OF THE 'TITULUS CRUCIS'

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ABSTRACT. The authors study the radiocarbon dating of a relic believed to be the tablet that was placed on the cross of Jesus Christ at the time of his crucifixion.

INTRODUCTION

The scope of the present work was the dating of a religious relic preserved in the Basilica of "Santa Croce in Gerusalemme" in Rome, Italy. This relic is believed to be the tablet that was placed on the cross of Jesus Christ. Dating was performed in a new radiocarbon (^{14}C) laboratory ("E. Amaldi" Physics Department of "Roma Tre" University) using 2 highly sensitive liquid scintillation spectrometers. Control measurements were performed on samples of known historical dates. A comparison between the ^{14}C dates and the historical dates of the control samples indicates the good operation of the analytical system and validates the calculated age of the "Titulus Crucis".

SAMPLE DESCRIPTION

The sample studied is a religious relic preserved in the Basilica of "Santa Croce in Gerusalemme" in Rome (Italy). This relic is believed to be the tablet that was placed on the cross of Jesus Christ, following the practice of the Roman penal code for every condemned prisoner. Inscribed on the tablet (Rigato 1999, 2001) is the phrase "Jesus of Nazareth, King of the Jews", written in Latin, Greek, and Hebrew.

The irregularly shaped tablet is approximately 26 × 14 × 4 cm in size, weighs about 687 g, and consists of a single piece of walnut wood (species *Juglans Regia* L) (E Corona, personal communication 2001). A sample for ^{14}C dating was collected from the lower central part of the back of the "Titulus Crucis", an area characterized by surface alteration of about 4 mm that covers wood that is completely intact, very compact, and perfectly preserved. Six samples were collected from different locations in and around a knot in the wood.

SHIELDING AND ELECTRONIC REGISTRATION

Dating was performed in the new ^{14}C laboratory located in the E Amaldi Physics Department of Roma Tre University. The laboratory has 2 independent, highly sensitive liquid scintillation spectrometers (A and B) as well as the capability to synthesize benzene. The characteristics of this laboratory are very similar to those of the ^{14}C laboratory at La Sapienza University of Rome, of which dates are prefixed by the letter *R* (*Radiocarbon* 1964 and successive years) and both laboratories were designed and constructed by the same researcher (Bella et al. 1960; Alessio et al. 1970; Alessio et al. 1973). Each spectrometer consists of the following:

1. A system for shielding the soft component of cosmic rays, consisting of iron, borate paraffin and lead;
2. Two photomultipliers which, in coincidence, monitor the benzene-filled sample vial;
3. A vial holder. The watch-shaped vial is constructed of low-sodium glass and has a volume of 2 cm^3, an external diameter of 3 cm and a depth of 0.6 cm. A small tube is fused onto the outer

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surface of the vial and closed with a flame using a technique adopted by the laboratory (Alessio et al. 1978);

4. A system for shielding the penetrating component of cosmic rays, consisting of a plastic scintillator with 8 photomultipliers that surround the coincident detector and which, through a Fan-in/Fan-out, create a classic anti-coincident system.

The new registration system is different from its predecessor at La Sapienza University because:

1. Highly stable electronic components, which allow for resolved real times within 6 ns, have been used;
2. For more control, it allows calculating ^{14}C dates directly from the coincidence between the two photomultipliers monitoring the sample, in addition to that from the anticoincidence.

CHEMICAL PREPARATION OF THE SAMPLES

Sample preparation was conducted very carefully, as the quality of the dating depends greatly on the mechanical and chemical pretreatment. The preparation of benzene was performed following the well-tested suggestions of Prof H A Polach (Polach et al. 1967). The small amounts of non-coeval contamination were eliminated first by unaided visual examination and then by using a stereoscopic microscope. The fragmented samples were then subjected to chemical treatment, including hot 5% HCl to eliminate carbonate contamination and 2% NaOH to remove secondary humic acids. A final 5% HCl treatment completes the cleaning procedure. After each acid treatment, the samples were neutralized with successive washing and boiling in distilled water and then desiccated in an oven at 70 °C.

The production of benzene is relatively complex, and is based on the 4 fundamental steps that are summarized below.

1. Combustion of the sample in a flux of O_2, followed by purification and storage of the produced CO_2;
2. Fluxing of the CO_2 in a titanium catalytic reactor containing metallic Li at 700 °C to obtain Li_2C_2. The exothermic reaction is completed at 900 °C in 1 hr, after which the sample is cooled to room temperature and distilled H_2O is injected in the vessel to convert the Li_2C_2 into C_2H_2;
3. The acetylene is transformed into benzene in the presence of an aluminum silicate catalyst activated with potassium-bichromate. Benzene is produced when the acetylene adsorbed on this material is heated to 150 °C and then collected in a small cryogenic oxygen trap.
4. Finally, a stoichiometric amount of PBD scintillator (Fluka) is added to the obtained benzene.

There was little problem in preparing the benzene for this study because the 4 samples were well-preserved. Notably, the weight of the wood treated from the "Titulus Crucis", sample 43, was 5.8 g before chemical and mechanical treatment and 5.1 g before combustion.

DATING

The activity of our "modern standard", consisting of wood that grew near Rome between 1949 and 1953, was checked repeatedly with 95% of the counting rate of NBS oxalic acid and was found to be within 1 σ. All dates are reported in conventional ^{14}C yr, using the Libby half-life of 5568 ± 30 yr, with AD 1950 as the standard yr of reference. The dating of the studied samples was performed under different background conditions by varying the discriminator thresholds and the voltage of the photomultipliers. Besides the accurate count checks, each series included ^{14}C age measurements of the following samples:

Sample 43. **"**Titulus Crucis" (see Tables 1–3)

Sample 29. A piece of a wooden lintel from a small storeroom in the attic of the Basilica of "S. Croce in Gerusalemme" where the "Titulus Crucis" may have been kept (Rigato 1999)

Radiocarbon age[2] **t = 960 ± 260 BP**

Sample 31. Part of the wooden plank, on which the body of Giovanni de Alessandro de Medici was laid when he was buried around AD 1360, taken from the center of the nave in the Florence Duomo (Azzi et al. 1973, 1974).

Radiocarbon age **t = 650 ± 73 BP**

Sample 40. Wood from Roman ships housed in the "Museo delle Navi Romane" in Nemi (Italy). This sample was 1 of 3 distributed among many ^{14}C researchers in order to perform the first interlaboratory cross-check. The average value calculated using all of the values was 1990 ± 85 BP (Alessio et al. 1964). Historically, construction of the ships occurred during the reign of Caligula, from AD 37 to 41.

Radiocarbon age **t = 1943 ± 40 BP**

The following 3 tables give the results of the datings performed.

Table 1 Dating of wood samples from the Nemi ships and the "Titulus Crucis" using spectrometers A and B under different conditions by varying the discriminator thresholds and the voltage of the photomultipliers. Measurements were collected directly by coincidence of the 2 photomultipliers that monitor the vial. The errors of the single age are calculated as experimental errors.

	Age measurements using only coincidence		
Counter	Background C/min	Nemi wood (BP) sample 40	"Titulus Crucis" wood (BP) sample 43
A	21.6 ± 0.1	2063 ± 163	973 ± 115
	22.5 ± 0.2	1676 ± 199	1084 ± 157
	21.1 ± 0.2	1907 ± 90	1051 ± 123
B	3.77 ± 0.07	2340 ± 240	986 ± 135
	4.7 ± 0.1	1919 ± 152	1073 ± 210
	4.37 ± 0.03	1985 ± 307	999 ± 208
	4.80 ± 0.03	1937 ± 133	970 ± 96
	Weighted mean:	1941 ± 56	1006 ± 50

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[2] This sample is subject to a large error due to the uncertainty of the volume of benzene used for the radioactivity counting and by the imperfect closure of the vial.

Table 2 Dating of wood samples from the Nemi ships and the "Titulus Crucis" using spectrometers A and B under different conditions. The measurements were acquired by the anticoincidence between the coincidence of the 2 photomultipliers monitoring the benzene-filled vial and the fan-in/fan-out system. The errors of the single age are calculated as experimental errors.

	Age measurements using anticoincidence		
Counter	Background C/min	Nemi wood (BP) sample 40	"Titulus Crucis" wood (BP) sample 43
A	2.47 ± 0.03	2030 ± 155	951 ± 90
	2.20 ± 0.03	1898 ± 232	1052 ± 214
	2.43 ± 0.03	2012 ± 152	1162 ± 110
	2.43 ± 0.03	1959 ± 87	1026 ± 51
B	0.97 ± 0.03	1893 ± 173	991 ± 103
	1.13 ± 0.03	1611 ± 269	1063 ± 219
	1.10 ± 0.03	1859 ± 163	1025 ± 154
	1.30 ± 0.03	1948 ± 122	915 ± 193
	Weighted mean:	1945 ± 51	1023 ± 36

Table 3 Comparison between the historical and ^{14}C dates of the reference samples, with the latter being calculated with a half-life of $\tau = 5568$ yr, the value agreed upon at the 5th Conference on Radiocarbon Dating (Cambridge 1962)

Sample nr	Sample description	Historical dates (BP)	Radiocarbon dates (BP)
29	Attic lintel	~1000	960 ± 260
31	Burial plank	~600	650 ± 73
40	Roman ship wood	~1990	1943 ± 40
43	Wood from the "Titulus Crucis"	–	1020 ± 30

CONCLUSION

The experimental confirmation of the known historical dates of the control samples indicates a good operation of the analytical system and validates the calculated age of the "Titulus Crucis":

Radiocarbon age of the "Titulus Crucis" = 1020 ± 30 BP

Calendar age of the "Titulus Crucis" = AD 996–1023 (1 σ)
AD 980–1146 (2 σ)

The calendar age has been calculated using the INTCAL98 program (Stuiver et al. 1998). This dating permits one to consider alternative hypotheses regarding the origin of the tablet, such as the possibility that it is a copy of the original, as discussed by Rigato (2002).

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THE ANTIQUITY OF PEARL SHELL (*PINCTADA* sp.) BURIAL ARTIFACTS IN PALAU, WESTERN MICRONESIA

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ABSTRACT. Pearl shell was an important and highly valued resource for producing tools and ornaments in Oceania. One pearl shell artifact type that is quite rare in Micronesia, however, is the crescent-shaped scraper/grater. These artifacts have recently been found in 2 burial caves in Palau, Western Caroline Islands, suggesting they may have played important social and symbolic roles in society. The first direct accelerator mass spectrometry (AMS) dating of this tool type, found in association with an in-situ female burial at the Chelechol ra Orrak site, provides a date of AD 150–270, while associated dates range from 770 BC–AD 180. These dates help contextualize human burials and associated artifacts from one of the earliest and most diverse burial sites in Austronesia.

INTRODUCTION

Prehistoric peoples in the Pacific used pearl shell (*Pinctada* sp.) for producing a variety of tools and ornaments over a period of 3000 yr or more. In Melanesia, early Lapita settlers in the Mussau Islands (Kirch 1997:213–4) and late preceramic inhabitants in the northern Solomons (Wickler 2001:200) used whole valves of pearl shell as vegetable or coconut peelers and produced trolling hook shanks (Wickler 2001:199). Pearl shell "crescents", often considered wealth or status items, are also worn today as neck ornaments in parts of New Guinea (Sillitoe 1988:396) and Vanuatu (Speiser 1990:165). Pearl shell is known archaeologically and ethnographically in Polynesia for manufacturing fishhooks (Davidson 1968; Sinoto and McCoy 1974; Emory 1975:199–205; Bellwood 1978; Kirch 1979:176; Walter 1989, 1990; Kirch et al. 1992; Allen 1994), and sometimes harpoon points (Sinoto 1968). In Micronesia, pearl shell was used for making fishhooks (Intoh and Leach 1985:100–4), trolling lures (e.g. Pohnpei, Kosrae, and the Marshall Islands; Ayres 1990:191–7; Intoh 1998), and various other objects (Sinoto 1984:36; Intoh and Leach 1985:101; Shun and Athens 1990:236-37; Carucci 1992:94). Pearl shell scraper/graters, similar to ones found outside Micronesia, were also recently recovered in archaeological investigations. All are from Palauan burial caves (Rieth and Liston 2001; Fitzpatrick 2003), but their chronology and function are poorly documented.

With the advent of accelerator mass spectrometry (AMS) radiocarbon dating, archaeologists can now date smaller samples of artifacts with a higher degree of accuracy than using conventional techniques. This is especially crucial in contexts that may be disturbed, contaminated, or have only associated dates (e.g. Rick 2001). In addition, direct dating of artifacts using AMS can refine artifact chronologies with relatively high precision (Vellanoweth 2001).

In this paper, we describe the first direct dating of this tool type at the Chelechol ra Orrak site in Palau using AMS. Our goals include: 1) determining the antiquity of burials and grave goods at the site, and 2) developing a chronology for these important artifacts throughout the Pacific. We first provide a brief background on archaeological research at Chelechol ra Orrak and discuss the provenience and significance of the pearl shell scraper/grater tools found in association with early human burials. Four additional ^{14}C dates from strata that contained the artifacts and human skeletal remains are used to assess the importance of directly dating artifacts in complex cultural deposits.

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BACKGROUND

Palau is located in the Western Caroline Islands of Micronesia approximately 600 km equidistant from the Philippines to the west and New Guinea to the south. Recent archaeological investigations indicate that the archipelago was probably settled by at least 1000–1400 BC (Liston et al. 1998; Wickler 2001; Fitzpatrick 2002a), although paleoenvironmental data suggest that colonization could have occurred even earlier (Athens and Ward 2001). Despite the increase in archaeological research in Palau, and especially on the large volcanic island of Babeldaob, dated contexts that extend past 500 BC are rare (Wickler 2001; Fitzpatrick 2003).

The Chelechol ra Orrak ("beach of Orrak") site is located along the western fringe of Orrak, a small "Rock Island" approximately 1 km east of Babeldaob's southeastern tip (Figure 1). This raised limestone island has rough karst topography and numerous caves and rockshelters. Blaiyok (1993) originally identified the site as a stone money quarry used within the last few hundred yr by Yapese Islanders (Fitzpatrick 2001, 2002b). Earlier cultural deposits (~1000 yr BC) were discovered at the site during recent excavations and produced the pearl shell artifacts discussed here (Fitzpatrick 2002a, 2003).

Human remains were found in all 4 test units in stratified deposits usually deeper than 60 cm. Preliminary osteological analysis by Nelson et al. (2002) indicates that at least 25 individuals were buried in the cave, 14 of which were recovered in Test Unit 1. Human remains include neonates, prenates, adolescents, and adults of both sexes, comprising one of the largest, earliest, and most diverse burial assemblages in the Pacific Islands.

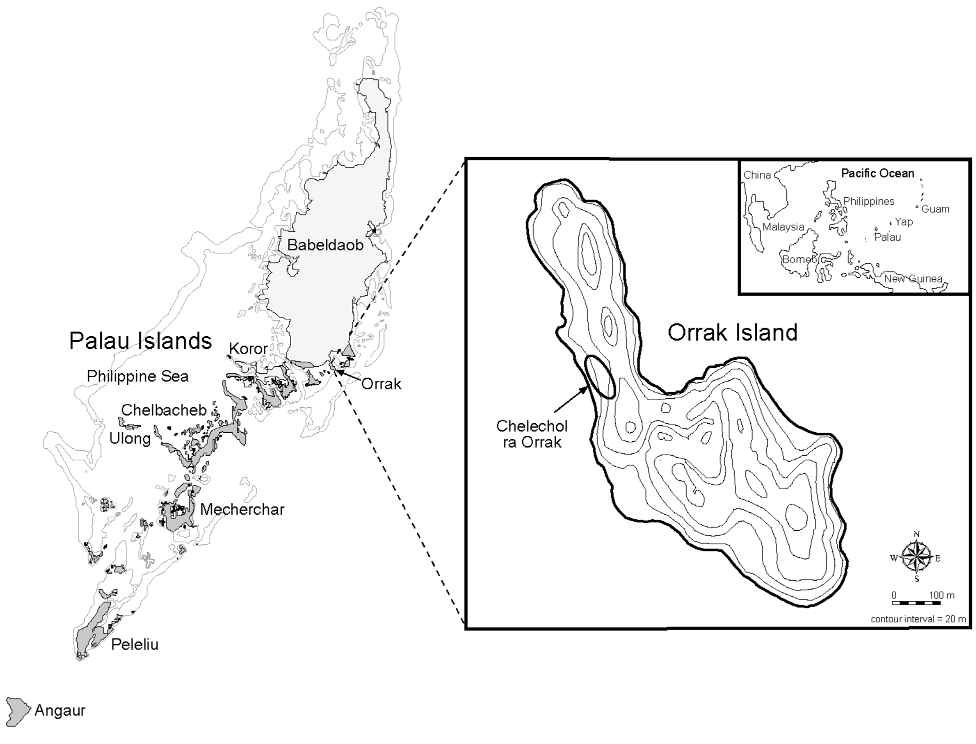

Figure 1 Map of main Palauan archipelago with Orrak Island inset

Due to the sandy nature of the deeper deposits and the likelihood that burial activity took place over at least a 1000-yr period (with later burials disturbing earlier ones; Fitzpatrick 2003), the skeletal assemblage was highly fragmented and rather poorly preserved. However, 2 undisturbed in-situ burials were discovered a meter deep in Test Unit 1. Three pearl shell scraper/graters were located directly above the left femur of a supine female burial in Layer 9 and are the only definitive burial goods discovered at the site. Two other fragments of similar tools were also found in Test Unit 1 (Layer 7) and one in Test Unit 2 (Layer 5; Table 1).

A variety of artifacts such as unworked marine shell, shell ornaments, shark teeth, bone tools, and ceramic vessels have been reported from burial sites in Palau (Beardsley 1998; Liston et al. 1998; Rieth and Liston 2001), but only Ngermereues Ridge and Chelechol ra Orrak have produced pearl shell artifacts. To determine the antiquity of these artifacts and their associated burial contexts in Test Unit 1, 5 samples, including a small piece from a pearl shell scraper/grater tool were submitted for AMS ^{14}C dating.[2]

Table 1 Pearl shell scraper/grater artifacts from Chelechol ra Orrak

TU	Layer	cmbs	Length (mm)	Width (mm)	Thickness (mm)	Weight (g)
1	7	50–60	47.5	36.1	2.5	5.6
1	7	50–60	19.8	10.1	2.0	0.5
1	9	~110	126.2	75.4	4.8	50.7
1	9	~110	71.6	68.5	2.8	17.5
1	9	~110	94.6	69.1	3.5	23.0
2	5	40–50	62.9	49.4	2.6	11.7

METHODS

All specimens submitted for ^{14}C dating were recovered from the site in situ, cleaned of extraneous soil using distilled water, air dried, and individually bagged in airtight containers for transport. The pearl shell tool (OS-33447) and charcoal (OS-33568) sample were submitted to the National Oceanic Sciences Accelerator Mass Spectrometry (NOSAMS) facility at the Woods Hole Oceanographic Institution for AMS ^{14}C dating. Laboratory preparations were conducted using standard techniques, details of which can be found at the NOSAMS website <www.nosams.whoi.edu>. Three additional samples (two human bone [AA-40957, AA-43054] and one burned fishbone [AA-43050]) were submitted to the NSF–University of Arizona AMS Facility. Pretreatment and analysis procedures for the bone are outlined in Fitzpatrick (2002a). All samples were calibrated at 1 σ using Calib 4.3 (Stuiver and Reimer 1993)[3]. A local ΔR for shell in Palau has not yet been determined, so the mean global reservoir correction (~ 400 ^{14}C yr) was used (Stuiver and Reimer 1993)[4].

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[2] See Fitzpatrick (2003) for a complete list of AMS dates from Chelechol ra Orrak to date, including those discussed here.

[3] Because the prehistoric diet is unknown but is presumed to contain both marine and terrestrial flora and fauna, the human bone samples were calibrated as 50% marine and 50% terrestrial to better reflect a mixed diet of shellfish, fish, and aroids (Hunter-Anderson 1991; Weisler 1999, 2000) common to the region. It should be noted that Ambrose et al. (1997) suggest marine protein consumption in the Mariana Islands (western Pacific) was around 20–50% and would thus make human bone dates in Palau slightly older if recalibrated.

[4] See Kennett et al. (1997), Phelan (1999), Guilderson et al. (2000), Kuzmin et al. (2001), Yoneda et al. (2001), and Hideshima et al. (2001) for recent attempts to determine ΔR correction values in other parts of the Pacific.

RADIOCARBON DATES

^{14}C dates from the basal layers of Test Unit 1 revealed a range of human activity spanning over a 1000-yr period from cal 970 BC to AD 270 (Figure 2). The earliest date was cal 970–840 BC from charcoal in Layer 8 (OS-33568). That this date was earlier than the others was not surprising considering the deposit was truncated by later burial episodes (Layer 9). The 4 bone and shell samples, all from Layer 9, dated from cal 770 BC to AD 270. The broad age range in this deposit can be attributed to one of the bone samples (AA-40957) dating to cal 770–550 BC; the other 3 dates fall within the range of cal AD 80–270. The earlier date in Layer 9 can probably best be explained as resulting from soil disturbance due to subsequent periods of burial activity (Fitzpatrick 2003). The later date of cal AD 150–270 obtained from the pearl shell scraper/grater lends support to this conclusion due to its direct association with the burial in Layer 9, as does the fragmentary nature of other skeletal remains recovered in Layers 7–9.

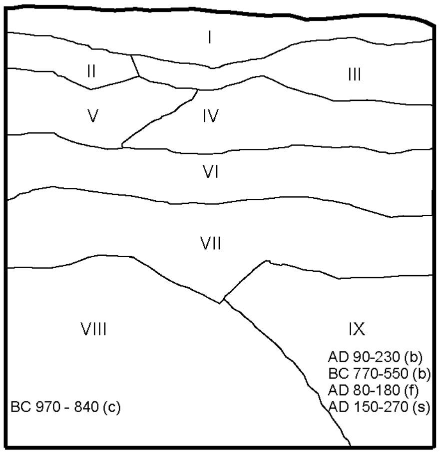

Figure 2 Stratigraphic profile from the east wall in Test Unit 1 (b = human bone; c = charcoal; f = fishbone; s = shell scraper grater)

PEARL SHELL ARTIFACTS

Pearl shell is found in tropical or subtropical waters throughout the Indo- and western Pacific. *Pinctada maxima* (silver or golden-lipped pearl oyster), *Pinctada margaritifera* (black-lipped pearl oyster), and *Pinctada radiata* (Ceylon pearl oyster, also known as *P. imbricata*) are the most common species found in the western Pacific. *P. maxima* has a restricted range, commonly found around Australia's north coast, the Arafura Sea, and the Aru Islands in eastern Indonesia. It has a silver-yellow nacre with greenish tint and is the largest species, with lengths of up to 30 cm. *P. margaritifera's* nacre is a rich silver gray with blue, green, or rose overtones edged with grayish-black and is slightly smaller with maximum lengths around 20 cm. *P. radiata* are smaller, generally brownish with shades of red, and reach lengths ranging from 5 to 10.5 cm (sometimes misidentified as *P. margaritifera*; Dance 1974; Wye 1991). *Pinctada* sp. typically attach themselves to hard substrata (e.g. under stones, in crevices of rocks) in intertidal and subtidal environments at depths ranging from very shallow to 190+ m in depth. Today, all 3 species are prized for their pearls and commercially cultivated.

Judging from the size and coloration of the pearl shell scrapter/grater tools from Chelechol ra Orrak and comparative specimens from Ngermereues Ridge (Rieth and Liston 2001), they appear to be made from *P. margaritifera* (Linnaeus 1758). The artifacts exhibit no external lamellae that would prove more useful in identifying the particular species. However, nearly all other pearl shell refuse found at the site have a grayish-green exterior, typical of *P. margaretifera*.

This tool type appears to be rare in Micronesia. Somewhat surprisingly, a review of the literature reveals only 1 other example of these tools found in archaeological sites in the region, also from Palau (Rieth and Liston 2001:44–8), despite pearl shell having been used throughout Micronesia for producing other objects. The artifacts found associated with the burial in Test Unit 1 at Chelechol ra Orrak range from 6.9 cm to 7.5 cm in width and 7.2 to 12.6 cm in length. All 3 are roughly half-moon in shape and retain the natural curvature of the shell (Figure 3). Serrations are present on one side of each tool to create a grating or scraping edge useful for peeling vegetables, grating coconut, or some other cutting function (Figure 4).

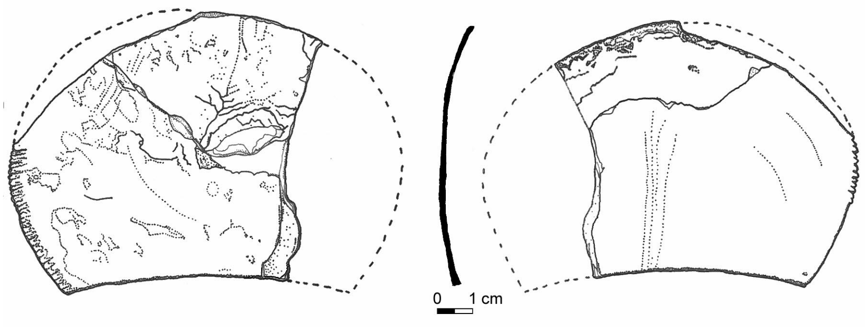

Figure 3 Pearl shell scraper/grater (drafted by Jenna Boyle)

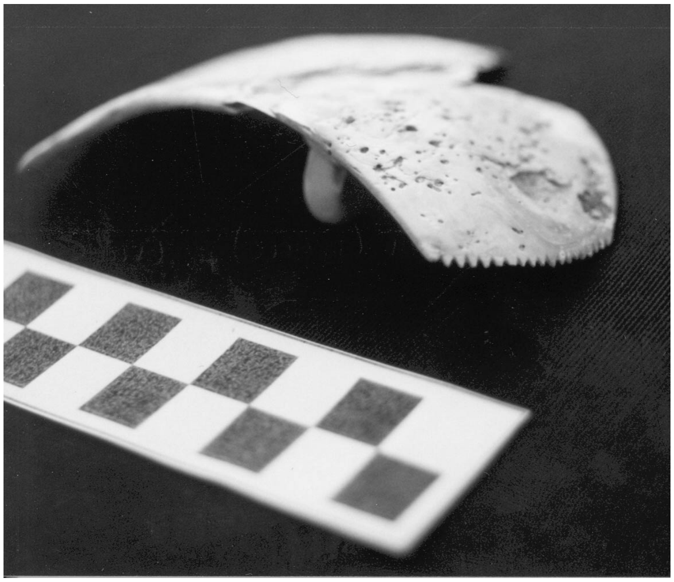

Figure 4 Photo of the serrated edge from a pearl shell scraper/grater (photo courtesy of Brian D Diveley)

DISCUSSION

^{14}C dates indicate that pearl shell grater/scraper tools were used at least 2000 yr ago as burial goods at Ngermereues Ridge (Rieth and Liston 2001) and Chelechol ra Orrak. They are similar to ones described as coconut graters in early ethnohistoric accounts (Adams et al. 1997), suggesting these artifacts have a long antiquity in Palau. Rieth and Liston (2001) did not directly date any of these tools, but ^{14}C dates of human bone from burial chambers dated from cal 470 BC to AD 780 (2 σ) and are within the range of the pearl shell artifacts from Chelechol ra Orrak.

Rieth and Liston (2001:49) note that "[t]he association of marine shell with the burials most likely represents the need to supply the soul with subsistence and tools to carry with them on their journey to the afterlife." Although this is speculative, the question remains as to what role these tools played in Palauan social systems, since grave goods are often indicators of an individual's gender, status, or occupation (Pearson 2000).

In ethnographic references (Adams et al. 1997:49), pearl shell is listed as a form of women's money (*chesiuch*) and often associated with female tasks such as grating taro or coconut. These tools were also apparently added along with food to a basket prepared for pregnant women by their parents as part of the birth ritual (Adams et al. 1997:31). Of the 6 pearl shell scraper/graters found by Rieth and Liston (2001), 3 were associated with a single individual from Chamber 7A, 2 with individuals in Chamber 4 (MNI=4), and 1 with an individual from Chamber 9 (Rieth and Liston 2001:71). Although sexing of the skeletons was difficult and could not be determined for most of the assemblage, Rieth and Liston (2001:31) report that at least 1 of the burials from Chamber 4 may be female. The Layer 9 burial at Chelechol ra Orrak is also female, suggesting the distribution of these particular pearl shell tools may be indicative of gender and status.

This research is the first direct dating of a pearl shell scraper/grater tool in Palau, a rare documented use of *Pinctada* sp. shell for this artifact type in Micronesia. AMS ^{14}C dating of shell artifact, bone, and charcoal samples from burial deposits at Chelechol ra Orrak indicates that burial activity at the site began around 3000 yr ago, with pearl shell scraper/graters used as grave goods roughly 1000 yr later. The interment of these tools with female burials is indicative of the high status some women attained in early Palauan society. This also supports ethnohistoric and ethnographic accounts stating that pearl shell was a gender and probably a status marker used almost exclusively by women for processing coconuts and starchy root crops like taro (*Colocasia* sp.) and giant swamp taro (*Cyrtosperma* sp.). Additional fieldwork in western Micronesia is expected to supply a greater variety of pearl shell artifacts in which better comparisons can be made with those found in other parts of Oceania. Our data support the findings by other researchers suggesting that directly dating artifacts using AMS is a critical step for developing and refining artifact chronologies, especially in sites with complex stratigraphic relationships.

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ARCHAEOLOGICAL SAMPLES

OS-33447. Chelechol ra Orrak **2140 ± 50**

$\delta^{13}C = 0.36\%$

This determination was obtained from a fragment of a pearl shell (*Pinctada* sp.) scraper/grater (1.6 g) tool recovered from Test Unit 1, Layer 9, at a depth of 100–110 cmbs. The artifact was one of three found directly above the left femur of burial 1. Calibrated date range at 1 σ: AD 150–270.

OS-33568. Chelechol ra Orrak **2770 ± 30**

$\delta^{13}C = -25.9\%$

This determination was obtained from wood charcoal (0.1 g) recovered from Test Unit 1, Layer 8, at a depth of 100–110 cmbs. Calibrated date range at 1 σ: 970–840 BC.

AA-40957. Chelechol ra Orrak **2680 ± 40**

$\delta^{13}C = -15.7\%$

This determination was obtained from a human cranial bone fragment (3.0 g) recovered from Test Unit 1, Layer 9, at a depth of 90–100 cmbs. Calibrated date ranges at 1 σ: 890–800 BC (100% terrestrial) and 770–550 BC (50% marine and 50% terrestrial).

AA-43050. Chelechol ra Orrak **2220 ± 40**

$\delta^{13}C = -12.6\%$

This determination was obtained from an unidentified burned fish bone (1.1 g; probably pelagic) recovered in Test Unit 1, Layer 9, 100-110 cmbs. Calibrated date range at 1 σ: AD 80 (130) 180. Petchey and Higham (2000) suggest that reliable ^{14}C dating of fish bone (barracouta – *Thyrsites atun*) may be accomplished if the reservoir conditions of fish are similar to those of locally collected shellfish. This, however, has not been tested in Palau.

AA-43054. Chelechol ra Orrak **2030 ± 40**

$\delta^{13}C = -15.4\%$

This determination was obtained from a human left navicular bone fragment (0.8 g) from Test Unit 1, Layer 9, 80–90 cmbs. Calibrated date range at 1 σ: 90 BC–AD 50 (100% terrestrial) and AD 90 (140) 230 (50% marine and 50% terrestrial).

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MARINE CARBON RESERVOIR AGE ESTIMATES FOR THE FAR SOUTH COAST OF PERU

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ABSTRACT. In order to estimate the apparent age of seawater (R) and the corresponding local offset from the global marine radiocarbon calibration curve (ΔR) on the far south coast of Peru for 2 periods in the past, 6 pairs of associated marine shell and unburned wood samples from archaeological excavations at Loreto Viejo were ^{14}C dated. Three pairs from about cal AD 1280–1380 indicated larger and more variable ΔR estimates than have been obtained for other periods in nearby regions, suggesting that ΔR may vary considerably over space and/or time. Three pairs from about 1870–1680 cal BC yielded consistent shell dates, but only one reasonable terrestrial date and ΔR estimate, probably due to stratigraphic mixing in antiquity. The one early ΔR estimate falls slightly outside the range of the later ones, suggesting either still greater spatial variability in ΔR, or some temporal variability.

INTRODUCTION

In order to calibrate radiocarbon ages from archaeological marine shell excavated near Ilo, Peru (17°37′S, 71°20′W) (Figure 1), and to estimate how much a partially marine diet could have perturbed the apparent ^{14}C ages of human bones and tissue from this area, it is necessary to know the apparent age of local sea water (R) and the corresponding local offset from the global marine carbon calibration curve (ΔR) at the time of interest (Beavan and Sparks 1998; Beavan-Athfield 2001; Ingram 1998; Kennett et al. 1997, 2002; Molto et al. 1997; Stuiver et al. 1986; Stuiver and Braziunas 1993; Tauber 1983). Taylor and Berger (1967) assayed 4 shells collected at known times in the early 20th century on the coast of Peru and northern Chile, from about 10°S to about 33°S. The local reservoir ages indicated by 3 of these shells were in rough agreement, yielding ΔR estimates from 171 ± 34 to 307 ± 77. The shell from closest to Ilo (a gastropod collected around 15°S) indicated a much greater reservoir age (ΔR = 664 ± 45). This value, while not unprecedented, is very high and was excluded by Stuiver et al. (1986: Table 1) from their survey of regional ΔR estimates. The shell may have provided a false estimate because it was old when collected or was affected by geological carbonate (Dye 1994), or it may reflect a correct localized extreme value due to fluvial water depletion (Little 1993) or other factors.

Southon et al. (1995) estimated ΔR for a region south of Ilo (around 20°S) during roughly cal AD 200–900, reporting 4 tightly clustered values that were close to Stuiver et al.'s (1986) regional mean. This finding suggested that the regional value was correct, and that it was relatively constant during the last two millennia. Estimates by Southon et al. (1995) were based on composite archaeological artifacts made from marine bird tissues and terrestrial plant fibers, ensuring excellent contemporaneity of the samples. Because marine birds are mobile, their tissues may average out small-scale spatial variations in the marine carbon reservoir. This could contribute to the good agreement among the estimates, but might mask localized extremes.

Kennett et al. (2002) tried to estimate ΔR for the Ilo region during the Late Archaic Period (about 6000–4000 cal BC) using associated shell and charcoal samples from the site of Kilometer 4, but were thwarted because the ancient inhabitants apparently collected very old wood for fuel.

MATERIALS AND METHODS

Six pairs of marine shell and terrestrial plant samples were ^{14}C dated. Each pair was from a single archaeological stratum at the site of Loreto Viejo (17°36′8″S, 71°13′45″W) in the coastal Osmore valley, about 13 km inland from Ilo (Owen 1993). The samples were collected from 1/4 inch screens in the course of stratigraphic excavations. Extreme aridity and salty soil contribute to extraordinary preservation of plentiful desiccated plant material. Three sample pairs were from domestic middens that contained Chiribaya style ceramics and dated to cal AD 1280–1380 (1 σ; Table 1). The other 3 pairs were from a cooking area in a preceramic and early ceramic sector of the site, dating to 1870–1680 cal BC (1 σ; Table 1). Strata selected for sampling were well-defined by changes in soil texture and color, in order to reduce the chance of conflating samples from different depositional events. The selected strata were of low volume (Table 1), in order to ensure that each pair of samples was probably deposited within a relatively short period.

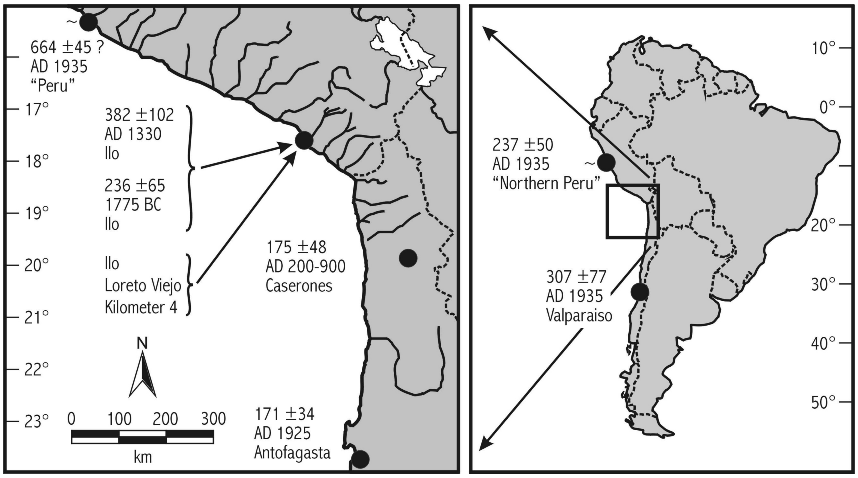

Figure 1 ΔR estimates for the Andean coast at various periods (with 40-yr Southern Hemisphere correction)

SAMPLES

Five of the 6 terrestrial samples were small unburned twigs of *Schinus molle* (California pepper tree, or molle) with bark. The remaining sample included twigs of both molle and unidentified species. The twigs ranged from 1 to 3 mm in diameter, which should ensure that they contain atmospheric carbon fixed during a period of no more than 2 or 3 yr. Such small twigs are unlikely to be “old wood” that was curated or collected from long-dead sources.

The marine samples were all fragments of *Choromytilus chorus* (choro mussel, or purple mussel) shell. Subtidal shellfish are preferable for marine carbon reservoir studies, because they are not exposed to atmospheric carbon during life (Goodfriend and Rollins 1998). Sandweiss et al. (1989) classify archaeological *C. chorus* from the nearby Ring Site as subtidal, and other sources treat *C. chorus* as primarily or entirely subtidal (Hancock 1969; Jaramillo et al. 1992; Soot-Ryen 1955). Intertidal *Choromytilus* are reported, but they are scarce, small, and of poor quality (Hancock 1969; Lasiak 1991). The archaeological specimens tend to be gigantic by modern standards, and they are ubiquitous in archaeological deposits, suggesting subtidal sources. If any of the samples were intertidal, ΔR estimates from them would be incorrectly low. Some grazing gastropods metabolize old carbon from seafloor minerals, exaggerating ΔR estimates based on their shells (Dye 1994; Phelan 1999). As a sessile bivalve, *C. chorus* should be less subject to this source of error. Since *C. chorus* shells in this region were rarely, if ever, worked, the mussels were probably collected live for food, making them contemporary with young twigs in the same contexts.

Table 1 Marine-terrestrial date pairs from Loreto Viejo (see notes on page 705)

Sample pair	Sample ID	Material and context	δ^{13}C (‰)	^{14}C age[a]	Apparent age of seawater[b]	Cal AD ±1σ[c] No SHC[e]	Model marine age[d] No SHC[e]	ΔR[f] No SHC[e]	Cal AD ±1σ[c] With SHC[g]	Model marine age[d] With SHC[g]	ΔR[f] With SHC[g]
Chiribaya midden, cal AD 1280–1380 (1σ)											
Unit 2505-5-6 midden, near top (279 liters)	Beta 51073	Small twigs of *Schinus molle* and other species	-26.3	730 ±60		1220–1390			1270–1390		
	AA37160	*Choromytilus chorus* shell	0.3	1358 ±46	628 ±76	640–770	1100 ±85	258 ±97	660–770	1070 ±70	288 ±84
Unit 2505-11-17 midden, near bottom (48 liters)	AA37161	Small twigs of *Schinus molle*	-27.7	701 ±38		1270–1390			1285–1390		
	AA37162	*Choromytilus chorus* shell	0.4	1428 ±56	727 ±68	560–665	1070 ±70	358 ±90	600–690	1060±60	368 ±82
Unit 2503-6-6 midden (44 liters)	AA40291	Small twigs of *Schinus molle*	-26.3	662 ±34		1285–1390			1300–1395		
	AA40292	*Choromytilus chorus* shell	0.0	1530 ±40	868 ±52	430–600	1060 ±60	470 ±72	535–620	1040 ±60	490 ±72
Weighted means[h]					772 ±36			384 ±49			393 ±45
Means w/ std dev[i]					**741 ±121**			**362 ±106**			**382 ±102**
Early cooking area, 1870–1680 cal BC (1σ)											
Unit 2513-5-4 around hearth, near top (36 liters)	AA37163[j]	Small twigs of *Schinus molle*	-27.3	1895 ±28[j]		65–135			125–220		
	AA37164	*Choromytilus chorus* shell	-0.8	3936 ±55	2041 ±62[k]	-2560–-2310	2250 ±30	1686 ±63[k]	-2470–-2300	2190 ±35	1746 ±65[k]
Unit 2513-14-33 pit fill, middle (9 liters)	AA40293[l]	Small twigs of *Schinus molle*	-25.7	1519 ±31[l]		440–610			540–620		
	AA40294	*Choromytilus chorus* shell	-0.1	3951 ±46	2432 ±55[k]	-2560–-2340	1885 ±75	2066 ±88[k]	-2470–-2310	1840 ±33	2111 ±57[k]
Unit 2513-11-18 ashy surface, near bottom (75 liters)	AA37165	Small twigs of *Schinus molle*	-26.7	3439 ±43		-1870–-1680			-1750–-1620		
	AA37166	*Choromytilus chorus* shell	-0.1	3961 ±47	**522 ±64**	-2570–-2350	3790 ±70	**171 ±84**	-2470–-2310	3725 ±45	**236 ±65**

Table 2 Marine-terrestrial date pairs from neighboring regions (see notes on page 705)

Sample pair	Sample ID	Material and context	$\delta^{13}C$ (‰)	^{14}C age[a]	Apparent age of seawater[b]	Cal AD $\pm 1\sigma$[c] No SHC[e]	Model marine age[d] No SHC[e]	ΔR[f] No SHC[e]	Cal AD $\pm 1\sigma$[c] With SHC[g]	Model marine age[d] With SHC[g]	ΔR[f] With SHC[g]
Composite artifacts from Caserones, northern Chile, cal AD 200–900 (Southon et al.											
CAS572	CAMS 10320	Plant fiber	-23.5	1270 ±60		660–860			690–890		
	CAMS 10321 and 10322	Fish vertebra and skin or stomach (mean)	-12.8 -14.5	1770 ±35	500 ±69	220–340	1640 ±95	130 ±101	250–390	1610 ±95	160 ±95
CAS512	CAMS 9372 and 9373	Wool yarn (mean of two samples)	-20 (est.)	1580 ±40		430–540			430–600		
	CAMS 7610 and 9374	Bird-skin cape (mean of two samples)	-13.4	2060 ±40	480 ±57	-160–1	1925 ±45	135 ±60	-90–60	1895±75	160 ±80
CASTr6/7	CAMS 10314	Wool yarn	-20.2	1690 ±60		250–430			260–530		
	CAMS 10315 and 10316	Feather and bird skin (mean of two samples)	-11.5 -12.2	2225 ±50	535 ±78	-380—200	2045 ±75	180 ±90	-360—170	2000 ±110	245 ±85
CAS93.031	CAMS 10317	Wool yarn	-20.4	1850 ±70		80–250			120–330		
	CAMS 10318 and 10319	Bird-skin cape (mean of two samples)	-13.1 -13.6	2295 ±50	445 ±86	-410—210	2195 ±70	100 ±86	-400—200	2145 ±85	135 ±110
Weighted means[h]					490 ±35			**136 ±40**			180 ±45
Means w/ std dev[i]					**490 ±38**			136 ±33			**175 ±48**
Historical shells, early 20th century (Taylor and Berger 1967; Stuiver et al. 1986, table 1)											
"Northern Peru"	UCLA 1282	*Strombus peruvianus* shell collected 1930–1940	-0.2	700 ±49	685 ±49		463 ±10	237 ±50			
"Peru"	UCLA 1279	*Oliva peruviana* shell collected 1930–1940	1.2	1127 ±44	1112 ±44[k]		463 ±10	664 ±45[k]			
"Antofagasta, Chile"	UCLA 1277	*Concholepas concholepas* shell collected 1925	0.1	626 ±34	601 ±34		455 ±4	171 ±34			
"Valparaiso, Chile"	UCLA 1278	*Tegula aler* shell collected 1930–1940	1.3	770 ±76	755 ±76		463 ±10	307 ±77			
Weighted means[h] excluding 1279					**643 ±26**			**206 ±27**			
Means w/ std dev[i] excluding 1279					680 ±21			238 ±21			

Table notes:

[a]Conventional radiocarbon age BP, including $\delta^{13}C$ adjustment, no Southern Hemisphere correction.

[b]Apparent age of seawater is the difference between the conventional ^{14}C ages of the marine and terrestrial samples (Stuiver and Braziunas 1993:137). The error estimate is $(\sigma_{marine}^2 + \sigma_{terrestrial}^2)^{1/2}$. This value is in ^{14}C yr and is independent of the Southern Hemisphere correction.

[c]Calibrated ±1 σ date range calculated by OxCal v3.5 (Ramsey 2000), atmospheric calibration data from Stuiver et al. (1998).

[d]The model marine age is the hypothetical age and error estimate that, when calibrated using OxCal v.3.5, the marine calibration curve (Stuiver et al 1998), and $\Delta R=0$, produces the associated terrestrial calibrated 1 σ date range.

[e]"No SHC" indicates that no Southern Hemisphere correction has been included in the calculation.

[f]ΔR is the difference between the measured marine ^{14}C age and the model marine age. The ±1σ error estimate is $(\sigma_{measured}^2+\sigma_{model}^2)^{1/2}$.

[g]"With SHC" indicates that the Southern Hemisphere correction suggested by Stuiver and Braziunas (1993) has been included in the calculation by subtracting 40 yr from the conventional ^{14}C age of the terrestrial sample prior to calibration and determination of the model marine age.

[h]Weighted means and error estimates after Bowman (1990:59).

[i]Arithmetic mean and standard deviation of values.

[j]Weighted mean of one measurement on each of 2 graphite targets prepared from the same sample (Bowman 1990:59).

[k]Unreasonably large, presumably erroneous value.

[l]Weighted mean of 2 measurements on 1 graphite target and 1 measurement on a second target prepared from the same sample (Bowman 1990:59).

Eleven of the 12 samples were pretreated and assayed at the NSF–University of Arizona AMS Facility under the direction of George Burr. The remaining sample was measured conventionally by Beta Analytic. The wood samples were given AAA pretreatment. The shell samples were pretreated with an HCl bath to remove surface layers that could contain recrystalized calcium carbonate.

ANALYTICAL METHODS

ΔR is calculated here as outlined by Stuiver et al. (1986) and Stuiver and Braziunas (1993), except that instead of a graphical intercept method (Southon et al. 1995; Facorellis and Maniatis 1998), a probability distribution method is adopted, using OxCal v3.5 (Ramsey 2000) with atmospheric and marine calibration data from Stuiver et al. (1998). The model marine age and error estimate are determined by successive approximations as the values that, when calibrated with the marine calibration curve, produce the same 1-σ date range as the terrestrial sample produces when calibrated using the atmospheric calibration curve. The smooth shape of the marine calibration curve ensures that there is almost always a unique solution.

Stuiver and Braziunas (1993) recommend subtracting a 40-yr Southern Hemisphere correction (SHC) from the terrestrial date before calibration. Marine dates calibrated with the resulting ΔR are comparable to terrestrial dates that incorporate the 40-yr SHC. However, few Andean archaeologists apply the SHC. Using a ΔR that includes the SHC would produce calibrated marine dates that are biased 40 ^{14}C yr younger than terrestrial dates calibrated without the SHC. On the other hand, most ΔR estimates are based on historically dated marine shells, the age of which is independent of any SHC. Marine dates calibrated with ΔR estimates from historical shells are comparable to calibrated terrestrial dates only if the terrestrial dates are correctly adjusted for hemispheric and other reservoir offsets—whatever those actually are. Moreover, the appropriate value for the SHC is still under discussion (Stuiver and Braziunas 1998; McCormac et al 1998). Given these uncertainties, ΔR is calculated here both with and without a 40-yr SHC.

Multiple values are summarized as weighted means of the values and error estimates (Bowman 1990:59) and as simple means and standard deviations of the values. Following Stuiver et al. (1986: 982), the method that results in the larger error estimate in any given case is adopted.

RESULTS AND DISCUSSION

The results of the 6 date pairs are given in Table 1. Data from Southon et al. (1995), Taylor and Berger (1967), and Stuiver et al. (1986) are recalculated in the same manner in Table 2. The recalculated values differ only slightly from the originally published results.

The weighted mean terrestrial conventional age for the 3 Chiribaya contexts is 687 ± 23 BP, or cal AD 1280–1380 (1 σ) without SHC. This range is as expected for the Chiribaya ceramic style. The 3 ΔR estimates are of credible magnitudes and are roughly consistent, averaging 363 ± 106 without SHC or 382 ± 102 with SHC. These estimates are considerably higher and more variable than those from previous studies (Table 2).

The tight clustering of the terrestrial dates suggests that the archaeological deposits accumulated over a brief period, so the variability is probably not due to poor contemporaneity of the paired samples, nor to temporal variations in ΔR. If the variation reflects spatial clines in ΔR, these must occur over very small distances, since all the shell probably came from within walking distance of Loreto Viejo. Such small-scale variation might be caused by the contribution of fluvial water at the river's mouth (Little 1993).

Estimated ΔR near Ilo about cal AD 1330 is high compared to the general region in the early 20th century (Taylor and Berger 1967; Stuiver et al. 1986) and to northern Chile around cal AD 200–900 (Southon et al. 1995). This could indicate temporal or spatial variability in ΔR. Alternatively, some of the new or previous ΔR estimates could be inaccurate.

The results from the 3 earlier date pairs were erratic. The 3 shell dates were very close, suggesting that they correctly date the deposits. The weighted mean of the conventional ^{14}C ages of the marine shell is 3951 ± 28 BP, or 1860–1680 cal BC (1 σ) without SHC using the single ΔR estimate discussed below. The 3 terrestrial dates were wildly variable. Two were over 2000 yr younger than the shells, indicating such extreme reservoir ages that they must be in error. The third indicates a believable ΔR estimate of 171 ± 84 without SHC or 236 ± 65 with SHC.

Both anomalous terrestrial dates were re-measured using new graphite targets prepared from remaining portions of the samples. One dated within 1 σ of the first target, and the other agreed within 2 σ, but both still suggested unreasonable reservoir ages. The twigs had been inspected under 10× magnification, and no decay or mold were noted. The discrepancy is probably not due to diagenetic changes or contamination of twigs that were contemporary with the shells, because an unreasonably high percentage of the carbon in the samples would have to be recent or modern to shift the apparent ages this much. The samples are unlikely to include post-occupation twigs, because the excavated area is on a barren desert hillside, over 200 m from the nearest irrigable land capable of sustaining a tree and 40 m above it. This is not an "old wood" problem, because the wood appears too *young*. It is unlikely to be an "old shell" problem, not only because the shell would have been collected fresh for food, but also because old shells collected on three different occasions would be unlikely to have such close ^{14}C ages.

The tightly clustered shell dates and the shallow cultural layers suggest that the entire sequence of deposits accumulated in a brief period, so there is no obvious source for drastically younger archaeological material. However, the stratum (2513-11-18) with a twig that gave a reasonable date is stratigraphically the earliest and best isolated from later deposits (Figure 2). The other 2 strata could contain twigs from a much more recent occupation, mixed in antiquity with shells from the earlier deposits that were brought up by digging the pit of 2513-14-33. Unfortunately, the material culture of this period is too simple and conservative to indicate on stylistic grounds whether the strata differ so significantly in time. This explanation implies exceedingly bad luck in sample selection, but it is possible. If it is correct, the sample pairs from 2513-5-4 and 2513-14-33 are not contemporary and cannot be used to estimate ΔR. On the other hand, this explanation gives no reason to reject the pair from 2513-11-18, providing one usable estimate of ΔR for the early period.

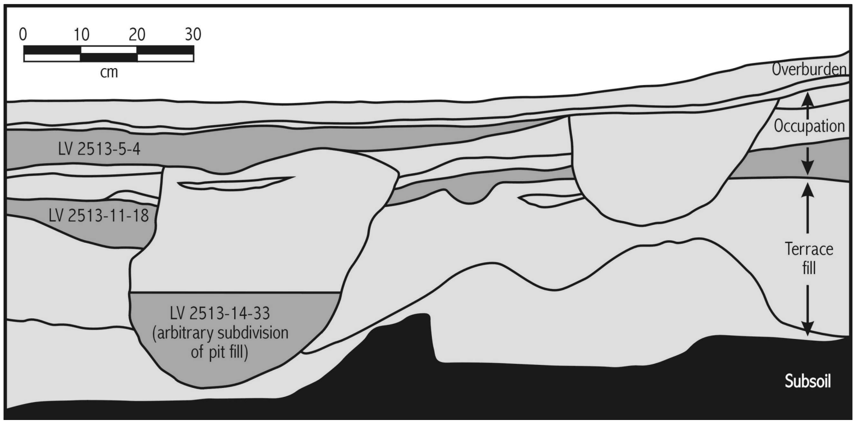

Figure 2 Strata sampled in the early cooking area at Loreto Viejo; north profile of unit LV 2513.

CONCLUSIONS

ΔR near Ilo, Peru, around cal AD 1280–1380 (1 σ) was about 362 ± 106 with no Southern Hemisphere correction, or 382 ± 102 with a 40-yr Southern Hemisphere correction. These values are higher than previous estimates for nearby regions at different times (Taylor and Berger 1967; Stuiver et al. 1986; Southon et al. 1995). This disagreement suggests that estimates of ΔR for this region should be used with caution, since there may be considerable spatial or temporal variation in marine ^{14}C reservoir depletion, or errors in some of the estimates.

One sample pair suggested a ΔR estimate for around 1870–1680 cal BC (1 σ) of about 171 ± 84 with no Southern Hemisphere correction, or 236 ± 65 with the 40-yr Southern Hemisphere correction. This estimate should be regarded as tentative, because 2 similar date pairs from nearby contexts gave erroneous results. While its lower magnitude is more in line with previously published estimates, it falls slightly outside the range of the other estimates reported here, accentuating the apparent variability of ΔR over short distances and/or over time.

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STABLE CARBON ISOTOPE MEASUREMENTS ON HAIR FROM WILD ANIMALS FROM ALTIPLANIC ENVIRONMENTS OF JUJUY, ARGENTINA

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ABSTRACT. The use of stable carbon isotopes as dietary tracers is an application that is widening its scope within the fields of ecology and paleoecology. Although hair is potentially one of the most favorable animal tissues for isotopic measurement for dietary studies, this tissue is rarely included in research works. This may be due to the fact that many aspects related to hair tissue are not fully understood, especially in the case of wild animals whose diets consist of plants with contrasting $^{13}C/^{12}C$ ratios, their abundance depending on seasonality. The present isotopic study of hair from animals inhabiting the Andes in northwestern Argentina, at heights ranging from 3500 to about 5000 m above sea level (asl) shows that 1) $\delta^{13}C$ values measured on hair from herbivores with a mixed and isotopically contrasting diet, and from their carnivorous predators, differ in their respective trophic levels, 2) in primary consumers, different types of hair from the same individual have different $\delta^{13}C$ values, whereas hair values are homogeneous in carnivores, and 3) some types of hair from rodents, such as whiskers, show $\delta^{13}C$ values similar to those of less metabolically active tissues such as bone collagen.

INTRODUCTION

Although the isotopic signatures that can be correlated with mammals' diets can be measured on any type of material from the organism, several circumstances have contributed to make bone the material preferred by researchers. The physical and chemical pretreatment for obtaining bone collagen or bone gelatin, on which measurements are generally made, involves strong acid and alkaline reagents, and temperatures ranging from −70° to 90° C. The features involved in this process may create doubts about the validity of some of the results obtained. In modern environments, and also under exceptional conditions in prehistoric environments, in order to carry out isotopic measurements, any other animal material is preferable to bone. Moreover, $\delta^{13}C$ values measured on bone collagen average the isotopic values of the food eaten and, thus, mask the dietary variations that could have taken place during the life of human or animal individuals. Therefore, seasonal variations in the carbon isotopic composition of the diet cannot be established using collagen due to its slow turnover rate. Information about variations in diet can be very important when studying food chains that involve isotopically contrasting plants, such as C_3 and C_4. Other tissues with faster turnover rates can provide information on the food eaten during short periods of time prior to death (Schwarcz and Schoeninger 1991).

The use of hair for isotopic measurements has several advantages. Individuals under study do not have to be sacrificed and, thus, can be re-examined over long periods of time under either constant or varying environmental and dietary conditions. Sampling is quick and, after surveying the samples under a microscope to verify their state of conservation and the absence of parasites, eggs, or dejections, pretreatment consists only of washing them with extremely volatile solvents. Also, the turnover rate of all the dietary isotopes is fast. The isotopic enrichment factor relative to diet, which is slightly over 5‰ in collagen, decreases to 1‰ in hair (Minson et al. 1975; DeNiro and Epstein 1978; Jones et al. 1981; Tieszen et al. 1983). In the case of natural feeding, fractionation has been greater, around 1.3 and 3.4 (Nakamura et al. 1982; Schoeller et al. 1986; Katzenberg and Krouse 1989; White 1993; Fernández C et al. 1999).

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[1] Deceased 24 December 2001.

Despite the important information provided by hair analysis, the use of this tissue is limited. It has been used mainly for comparison almost exclusively during the initial stage of dietary research using isotopic tracers. This research was carried out with laboratory animals and under controlled and isotopically constant diets (Minson et al. 1975; DeNiro and Epstein 1978; Jones et al. 1981; Tieszen et al 1983). Therefore, little is known about what happens in natural settings where animals fill their energy needs freely and completely. We aim here to contribute to the knowledge of these wild environments and to provide a better understanding of the way in which stable carbon isotopes are transferred following the energy flux from plants to animals, both herbivores and carnivores, belonging to different trophic levels, living in the remote and unpolluted environment of the Andes in northwestern Argentina.

NATURE AND PROVENANCE OF SAMPLES

The Puna of Jujuy is a high, arid plain located between the western and eastern Andes (Figure 1). It shows remarkable ecologic variability, possibly due to its relief, which includes heights ranging from 3500 to over 6000 m asl. The increases in height occur within short horizontal distances, influencing plant and animal distribution. The distribution resembles a complicated biological chessboard, changing within short distances due to factors determined by altitude.

A remarkable aspect of the animal community of this region is the strong predominance of herbivores in spite of the scarcity and discontinuity of vegetal cover. Out of this variety of plants and animals, the small sample presented in this work corresponds to 3 well-defined ecosystems, for both its altitude and its biological content. These ecosystems constitute strips of land with an E–W alignment and with a latitude almost coinciding with the Tropic of Capricorn.

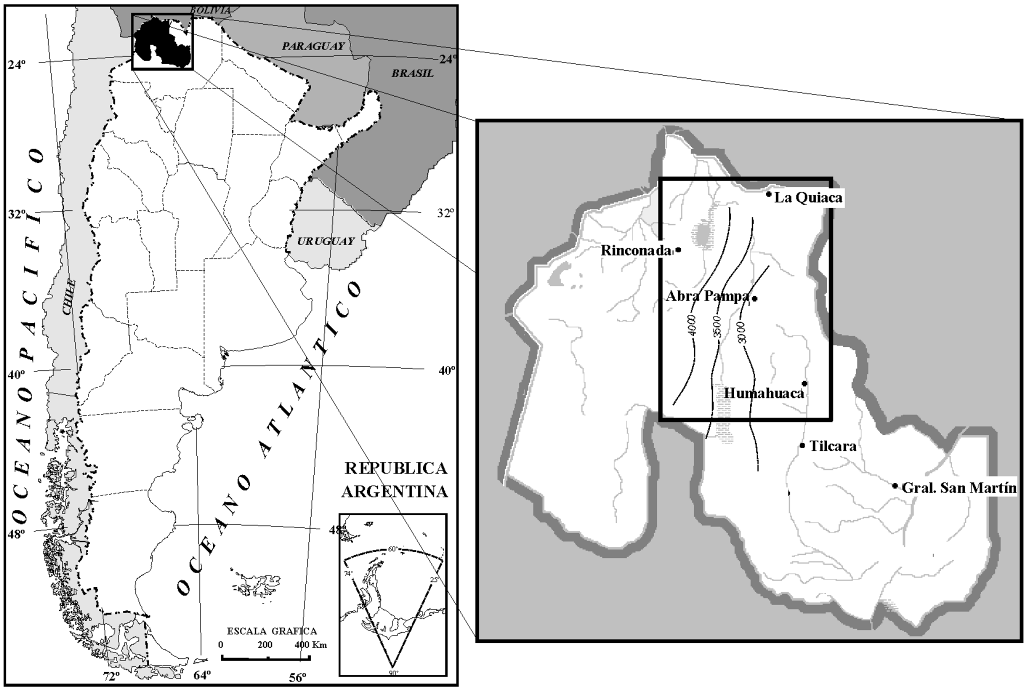

Figure 1 Geographic location of the study area

The first ecosystem occurs on the sides of isolated mountains, at heights varying from 4000 to about 5000 m asl, where annual rain, concentrated in the summer months, is 300 mm. In spite of the latitude, height makes night temperatures reach glacial values. The vegetation of this semi-arid region consists of a succession of scrub and grass steppes which follow the C_3 photosynthetic pathway, with the exception of *Muhlenbergia*. The large herbivores that inhabit this ecosystem are 2 wild camelids, the guanaco (*Lama guanicoe*), and the vicuña (*Vicugna vicugna*). Regarding their feeding habits, both camelids are indistinctly grazers and browsers. The puma (*Puma concolor*) is their main predator.

The second environment is characterized by the intense diaphaneity and great luminosity of its sky and corresponds to the mountainsides with heights ranging from 3600 to 4000 m. There is a marked difference between day and night temperatures. The land is covered by rock detritus and dunes, on which shrubs and mats grow forming discontinuous groups. Most of the monocotyledon plants follow the C_3 photosynthetic pathway, but there are also localized grasslands of *Pennisetum chilensis* and *Sporobolus rigens*, both C_4 species. This is the habitat of a digging rodent, *Ctenomys sp.*, a type of mole that lives in underground burrows and feeds on roots.

The third ecosystem is found at lower heights (3500 m), in the Altiplano itself. It is hyper-arid, the sky is clear and luminous, and annual rain values barely reach 100 mm. Nights are extremely cold. The relief consists of rocky outcrops where some shrubs and cacti grow in cushion-like shapes. Colonies of medium-size herbivorous rats, the chozchori (*Octodontomys gliroides*), live in this rocky habitat. Because there is little information about these rats in specialized literature, we will discuss some of their biological characteristics in more detail below.

Apparently due to the dangers posed by its feathered (owl, eagle, falcon) and terrestrial predators (fox, *Dusicyon sp.*; wildcat or oscollo, *Felis sp.*), the octodontic rat chozchori cannot have direct access to water. Its habitat being limited to cracks and holes in the rocks, this species seems to have developed strategies to cope with this lack of water availability by 1) developing the ability to regulate water consumption by means of urine concentration, 2) building shelters with available materials consolidated with indurated urine, and 3) feeding on vegetal species that accumulate water excess, such as cacti. In some aspects, the behavior of the chozchori rat is similar to that of the North American packrat (*Neotoma*), whose middens occasionally reach ^{14}C ages over 20,000 BP. They represent a primary source for the reconstruction of the climatic and biological past in arid regions. Taking into account the paleoenvironmental and paleoclimatic information that could be potentially preserved in the indurated middens of the *Octodontomys*, this species has been chosen for the present study.

Each of the animal species inhabiting the 3 environments considered can, to a certain extent, modify the composition of its diet mainly due to seasonal availability of food rather than by their own free will. Some of the vegetal species disappear completely during the toughest seasons and herbivores are forced to replace them by other species. This change in diet does not modify the isotopic values of the tissue of the herbivore, as long as it does not imply the consumption of plants that follow different photosynthetic pathways.

Direct field observation shows that the guanaco and the vicuña seasonally modify the vegetal composition of their diet, browsing more during winter (Table 1A–B). But this change does not imply an intake of plants with a different photosynthetic pathway. The same applies to the rodent (*Ctenomys*) living in the environment at intermediate heights.

On the other hand, field observations concerning the dietary habits of the chozchori (*Octodontomys*) show important seasonal changes. During winter, there is a marked dependence on cacti of the genus *Maihueniopsis* that is a CAM-type plant (*crassulacean acid metabolism*) photosynthesizing as C_4 and during summer, there is a supplementary intake of C_3 fresh grasses (Table 1C).

Table 1 $\delta^{13}C$ values of plants from environments at different altitudes in the Argentine Altiplano

Species	Common name	Altitude (m)	$\delta^{13}C$ ‰ (PDB)
A) Plants eaten by camelids (guanaco and vicuña)			
Ephedra breana	Paraguay	4000–5000	–21.6
Trifolium amabile	Paja blanda	4000–5000	–25.5
Poa juyensis	Suico	4000–5000	–26.4
Tagetes multiflora	Cebadilla	4000–5000	–27.3
Poa anua	Pasto blando	4000–5000	–25.9
Poa lilloi	Cienego	4000–5000	–250
Cotula mexicana	Pasto cebadilla	4000–5000	–27.8
Bromus catharticus	—	4000–5000	–285
Poa jujuyensis	—	4000–5000	–26.7
Festuca orthophylla	—	4000–5000	–27.5
B) Plants eaten by tojos (*Ctenomys sp.*)			
Nassauvia axillaris	—	3600–4000	–26.8
Ephedra breana	Pingo-pingo	3600–4000	–21.6
Nassauvia axillaris	—	3600–4000	–26.8
C) Plants eaten by chozchoris (*Octodontomys gliroides*)			
Maihueniopsis glomerata	Espina binacilla	3500	–15.3
Maihueniopsis boliviana	Quepo	3500	–15.5
Maihueniopsis sp.	Pushcaia	3500	–13.1
Tarassa sp.	Malva	3500	–23.7
—	Oreja-oreja	3500	–26.1
Gnaphalium lacteum	Vira-vira	3500	–31.6
Senecio graveolens	Canchalagua	3500	–22.4

METHODS AND MATERIALS

With the exception of chozchori (*Octodontomys*) specimens 3 and 4, none of the animals considered in this study has been intentionally sacrificed for the experiments. Therefore, in some cases we have not been able to get and analyze perishable parts of the animals.

The bodies of the large Andean herbivores (guanaco, *Lama guanicoe*; vicuña, *Vicugna vicugna*) inhabiting the highest ecosystem (3500–5000 m) are covered by an animal "fiber" similar to wool and possibly sharing the same physical and chemical properties. Therefore, sampling was limited to the short hair present in the inner side of the thigh.

Since the hair of the rodent from the second ecosystem (tojo, *Ctenomys* sp.) is very short in the ventral part and on the tail, samples were taken only from the dorsal part.

In the case of the chozchori (*Octodontomys sp.*), we found 3 types of hair that can be differentiated both for its location as for its functionality and possibly also for its physical and chemical characteristics: 1) whiskers, extremely long, as in all rodents, 2) hair on the tip of the tail, which forms a kind of tuft or brush, and 3) snowy-white hair that covers the animal's belly. To establish the grade of homogeneity of the carbon isotopic distribution in the chozchori (*Octodontomys gliroides*), we collected samples of these 3 types of hair from each individual analyzed. The same procedure was followed in the case of the identified chozchori carnivorous predators from the same locality and ecosystem: the wildcat or oscollo (*Felis sp.*) and the fox (*Dusicyon sp.*).

Hair samples were examined under a microscope and magnifying glass to remove any possible contaminants (dirt, parasites, or eggs) and later washed with sulfuric ether and dried in a stove.

Parts of diaphysary bones were obtained from the animals' skeletons. The remaining tissues were removed with a scalpel and the bone fraction was defatted in a Soxhlet with sulfuric ether. The decalcification organic residuum was extracted following the method of Sealy (Sealy and van der Merwe 1986), with modifications. The product resulting from the bone decalcification treatment described is informally referred to as collagen in this paper.

In order to calibrate the $^{13}C/^{12}C$ ratios both in the bone decalcification organic residuum and in mammals' hair, samples of plants conforming their diet were taken along a height and humidity gradient from the low lands at 3500 m inhabited by the chozchori (*Octodontomys*), up to the snow line, at almost 5000 m, the height reached by the camelids (guanaco and vicuña) in the region. We were not able to compare the feeding habits observed with what was actually eaten by the animals. Materials such as stomach contents and dejections, which would make this control possible (Jones et al. 1979), cannot always be easily identified and collected in the case of small animals living in burrows in rough areas. Plants were identified by species and, on the basis of leaf anatomy and $\delta^{13}C$ values, were grouped according to their photosynthetic pathway in C_3, C_4, and CAM (Table 1, A–C).

$\delta^{13}C$ analyses were done at the Instituto de Geocronología y Geología Isotópica (INGEIS) in Buenos Aires, Argentina following the technique described by Panarello (1987). About 8 mg of each sample of hair, bone decalcification residuum, and vegetal matter was mixed with 100 mg V_2O_5 in a Pyrex vial and flame sealed under a vacuum of about 10^{-2} Pascal. The sample was combusted for 8 hr at 550 °C in an electric furnace. CO_2 gas was then purified by cryogenically removing water and non-condensable gases, trapped with liquid nitrogen, and transferred to a Micromass 602-D McKinney type mass spectrometer. Results are expressed as δ, defined as follows:

$$\delta^{13}C=1000\frac{\left[{}^{13}C/{}^{12}C\right]_S-\left[{}^{13}C/{}^{12}C\right]_R}{\left[{}^{13}C/{}^{12}C\right]_R}\ ‰ \tag{1}$$

where:
$\delta = \delta^{13}C$ isotopic deviation in per mil, ‰
R = $^{13}C/^{12}C$ isotopic ratios
S = sample
R = reference standard

The standard is V-PDB as defined by Gonfiantini (1984) on the basis of PDB (Craig 1957). The standard measurement error is ±0.1‰. Table 1 shows the stable carbon isotopic values from the plants that compose the diet of the chozchori (*Octodontomys gliroides*) and of the other groups of animals we have selected for comparison. Table 2 shows the $\delta^{13}C$ values measured on hair and collagen of camelids and of *Ctenomys sp.*, and on the different types of hair and on the bone decalcification residuum of the chozchori (*Octodontomys gliroides*) and of its predators: 2 carnivores inhabiting the same locality, the wildcat or oscollo (*Felis sp.*) and the fox (*Dusicyon sp.*).

Table 2 $\delta^{13}C$ values of different types of hair and of bone collagen from *Octodontomys gliroides,* from its predators (*Felis sp.* and *Dusicyon sp.*), and from other animals from the Andean Altiplano

Species	Common name	Altitude (m)	Hair ($\delta^{13}C$ ‰ PDB) Whiskers	Tail	Belly	Collagen ($\delta^{13}C$ ‰)
Octodontomys gliroides	Chozchori	3500	−12.5	−15.8	−16.2	−12.3
Octodontomys gliroides	Chozchori	3500	−13.0	−15.2	−17.7	−12.0
Octodontomys gliroides	Chozchori	3500	−12.4	−16.7	−14.2	−12.2
Octodontomys gliroides	Chozchori	3500	−13.2	−12.4	−13.8	−10.9
Felis sp.	Wild cat	3500	−15.5	−15.2	−15.9	−13.6
Dusicyon sp	Fox	3500	—	—	—	−13.8
Ctenomys sp.	Tunduco	3700	—	—	−19.8	−13.8
Lama guanicoe	Guanaco	4500	—	—	−21.7	−20.1
Vicugna vicugna	Vicuña	4500	—	—	−22.1	−20.0
Puma concolor	Puma	4000	—	—	−17.0	−16.1
Puma concolor	Puma	4300			−19.1	−18.0

RESULTS

$\delta^{13}C$ values of plants from the mountain slopes between 4000 and 5000 m (Table 1A), grazed or browsed by the camelids (guanaco and vicuña), average −26.2‰. All these plants correspond to C_3 species. Assuming that $\delta^{13}C$ values of collagen from animals of the size of those considered in this study show an enrichment of 5.1–5.3‰ relative to diet (van der Merwe 1982, 1986), we find that $\delta^{13}C$ values of collagen from camelids (guanaco = −20.1‰ and vicuña = −20.0‰) agree with the expected values. $\delta^{13}C$ values of hair are more positive than those of diet by up to 1‰ (DeNiro and Epstein 1978), but this is not seen in the case of camelids. On the contrary, $\delta^{13}C$ values of hair from camelids are similar to those measured on collagen, possibly because the "hair" that covers these animals (pelage) has characteristics that are more similar to wool than hair, with its resulting chemical differentiation.

The plants eaten by the tojo (*Ctenomys sp.*) in the intermediate environment of the Altiplano, at heights of 3700 to 4000 m (Table 1B), include a C_4 gramineae *(Sporobolus rigens)*; therefore, the average $\delta^{13}C$ value of the diet is estimated at −20.1‰. This value is reasonably reflected in the $\delta^{13}C$ values of the tojo collagen (−13.8‰) and hair (−19.8‰).

The cacti eaten by the chozchori (*Octodontomys*), in the Altiplano itself, are CAM species that photosynthesize following the C_4 pathway. They show a mean $\delta^{13}C$ value equal to −14.6‰. The $\delta^{13}C$ value of the C_3 species on which the chozchori complementary feeds averages −26‰. The difference between both ends of the combined diet is −11.5‰. Taking an enrichment of 1‰, these dietary values will be fixed in the hair of animals feeding on either 100% C_3 or 100% CAM plants with values equal to −25.0 and −13.5‰, respectively. If we compare this last value with the isotopic values measured on ventral hair of chozchori specimens 3 and 4, captured at the beginning of winter, we can see that their diet was exclusively formed by CAM plants. On the other hand, specimens 1 and 2 reflect the mixed summer diet, with up to 20–35% of C_3 plants (Table 2). This seasonality in diet cannot be detected by the isotopic composition of collagen (−12.2 and −10.9‰).

If we compare $\delta^{13}C$ values of white hair from the belly with that of the whiskers and tail, we can see that $\delta^{13}C$ values of whiskers are generally the most positive, reaching values similar to those of collagen, whereas hair from the tail shows intermediate values between those of the belly and whiskers.

The differences mentioned above are not observed in any of the types of hair (whiskers, tail, or belly) of the oscollo (*Felis sp.*), the carnivorous predator of the chozchori rodent, which allows us to conclude that the distribution of $\delta^{13}C$ values of the animal pelage is homogeneous.

CONCLUSIONS

Hair is generally considered a short-term animal product, that is, a product of a short-term absorption of nutrients. Therefore, it should be expected that knowing the$^{13}C/^{12}C$ ratio of hair tissue, it would be enough to subtract 1‰ from this value to estimate the approximate percentile composition of the original plant diet.

Nevertheless, we find that this statement is not true in all the cases analyzed. For instance, the woolly hair of the guanaco and the vicuña, 2 mammals inhabiting the same environment at the upper heights of the Andes, shows $\delta^{13}C$ values that are about 5‰ more positive than those of diet, which means that $\delta^{13}C$ values of hair are more similar to those of bone collagen. This difference is worth studying in a larger number of cases due to its potential use in archaeology. Archaeological cloth made with this same type of animal fiber is often found in prehistoric burial sites, and is sometimes used for ^{14}C dating. The measurement of $\delta^{13}C$ values in archaeological cloth with known ^{14}C ages would allow one to obtain information about the type of pasture (C_3 or C_4) grazed upon by the prehistoric animals, and would thus be valuable as a paleoenvironmental indicator.

The rodents studied (*Octodontomys*) represent the first active link of the local food chain. Rough seasonal conditions affect the composition of their predominant diet, which changes from cacti in winter to an apparently graded mixture of CAM species with a proportion of up to 30% of C_3 grasses in summer.

The analytical results of the present study show that the passage or fixation in hair tissue of the isotopic values of a typically contrasting diet (C_3/CAM photosynthesizing as C_4) is not verified with equal speed and homogeneity in primary consumers. Some types of hair seem to be more metabolically active than others or, at least, hair from different parts of the body of the same individual can have significantly different isotopic values.

Apparently, the factor that determines the distribution of carbon isotopes in hair is the residence time of hair in the living individual. Hair on the rodent's belly, which is directly related to reproductive functions, appears to have the shortest life-span, since it shows faster turnover rates. Nevertheless, sampling of free animals necessarily involves the inclusion of hair from different growth stages, ages or seasons, which results in a mixture of isotopic values. Therefore, it is only relatively true that $\delta^{13}C$ values of hair can give information about the food eaten during a short period prior to death.

Other types of hair, such as whiskers, whose function is important to rodents throughout their lives, remain on the individual for a longer period of time and, thus, tend to accumulate isotopic values corresponding to older components of the diet. Therefore, whiskers tend to have a slower turnover rate than hair and their $\delta^{13}C$ values become similar to those measured on bone collagen of the same individual.

The remarkable shift in isotopic composition between diet and hair is only verified in the first link of the food chain, in our case, the large (camelids) and small herbivores (rodents).

In those animals that do not significantly modify their diets throughout the year, such as the puma, who eats camelids, and the wildcat or oscollo and the fox, who eat chozchoris *(Octodontomys)*, $\delta^{13}C$

values are relatively constant no matter the type of hair (with long or short residence time) on which the measurement is done, and these values coincide with those of collagen.

Taking into account the relevance of the facts observed and the dietary and paleodietary interest of the results obtained on a small number of cases, together with the possible paleoenvironmental implications and their potential use in archaeology, it would be interesting to extend this research to a larger number of individuals and their respective microenvironments.

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THE RADIOCARBON DATING AND AUTHENTICATION OF IRON ARTIFACTS

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ABSTRACT. The continuing improvements in accelerator mass spectrometry (AMS) dating technology mean that it is possible to work on ever smaller samples, which in turn, make an ever wider range of sample potentially available for dating. This paper discusses some of the difficulties arising with the interpretation of AMS dates obtained from carbon in iron. The overriding problem is that the carbon, now in chemical combination with the iron, could have come from a variety of sources with very different origins. These are now potentially an iressolvable mixture in the iron. For iron made over the last millennium, there are the additional problems associated with the use of both fossil fuel and biomass fuel in different stages of the iron making, leading to great confusion, especially with authenticity studies.

INTRODUCTION

The radiocarbon dating of the carbon contained within smelted iron was first proposed and accomplished by Nikolaas van der Merwe (1969). But the radioactive decay was perforce measured by the beta-counting methodology of the day that required approximately 1 kg of wrought iron to provide the necessary carbon and this precluded its serious practical application. Thus, the ^{14}C dating of iron seemed destined to linger in the limbo of good ideas that are fatally brought down by practical difficulties. The introduction of accelerator mass spectrometry (AMS) ^{14}C dating, capable now of operating on mg-sized carbon samples have transformed the possibilities of obtaining dates from smelted iron (Cresswell 1991, 1992; Harbottle et al. 1993; Kusimba et al. 1994; Cook et al. 2001, 2003). This is an exciting and important extension of ^{14}C dating generally and the recent review by Cook et al. (2003) shows that the dates obtained are very often commensurate with the age estimates made by other methods. However, the very fact that tiny sample weights are now dateable has raised dangers of serious misinterpretation. It is always potentially hazardous to work on samples when one can no longer identify the original form of the material that was the source of the carbon. Sometimes, even when it is apparently possible to identify the material, there are problems because it has been chemically modified, as exemplified below.

The rationale behind the feasibility of the ^{14}C dating of iron is that the carbon in the iron originates from the fuel of the smelting process, and for most ancient iron this fuel will have been charcoal. There seems to be an implicit assumption that the use of fossil fuel can be unambiguously recognized because of the geological age of the carbon (although the possibilities of Neanderthal iron smelting have been raised, apparently seriously, by Sherby and Wadsworth 2001). The potential dangers of serious misdating arising because of the intermixing of carbon from charcoal- and fossil fuel-smelted iron seem not to be appreciated (see Cook et al. 2003, discussed below).

It has been recognized that there is a possibility of contamination where limestone or other carbonate rocks have been used as a flux in the smelting process (Cresswell 1992), or where the iron is extensively corroded (Cook et al. 2003), and Eylon (2002) has documented the reuse of solid wrought iron, that was sometimes centuries old, welded to pieces of much more recent iron. However, it still seems that there has been insufficient attention paid to the potential problems arising from the carbon used in the actual smelting and other processes. This is probably due to a failure to fully realize the complexities of iron-smelting and how much the processes have changed over time. It also seems to be assumed that the histories of the various iron-making processes in use around the world are all known, and that evidence of the production stages to which any iron artifact will have

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been subjected will be preserved in its structure. In reality, the processes of iron production are both complex and very far from being understood, and very often the later stages of processing the iron will have completely removed the evidence of the earlier stages from the iron of an artifact, while the carbon they introduced remains unaffected but anonymous (Tylecote 1976; Rostoker and Bronson 1990; Craddock 1995:234–83, 1998, 2003).

The problems of the ^{14}C dating of iron were brought home to the authors when trying to get meaningful dates from a lump of iron, found at the village of Churchdown in Gloucestershire, England (Figures 1 and 2). The piece, apparently still containing lumps of its smelting fuel, is described in detail below. To anticipate the discussion of the piece, dates of both 35,000+ BP, and, more specifically, of 2800 BP, have led us to finally conclude that the piece is almost certainly of the late 18th to early 19th centuries AD. Clearly the interpretation of the AMS ^{14}C dates obtained on the carbon in iron is not a straightforward exercise!

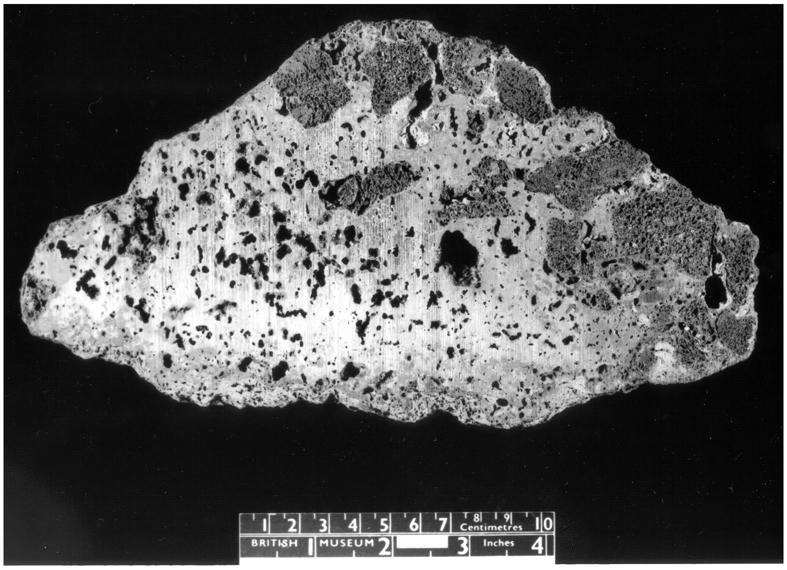

Figure 1 Half-sectioned lump of iron from Churchdown, Gloucestershire. It is a product of the fining of cast iron to make wrought iron. Note the rectilinear black areas exposed in the section. These turned out to be highly pyrolized hard wood charcoal upon which carbon from the coke-smelted cast iron had deposited. (Photo. British Museum / A. Milton)

This paper will explore some of the problems of interpreting ^{14}C dates obtained from iron, and the confidence with which they can be used. There are 2 rather different scenarios to consider:

1. When the age of the iron artifact is approximately known, stylistically or because the piece is from excavation, and in this case something should be known about the contemporary technology by which the iron is likely to have been produced.
2. When the age of the iron is completely unknown, or where there are 2 or more very different possibilities, such as with an authenticity determination.

In the first case, it is at least possible to predict likely problems arising from the technology, but in the second there will usually exist a bewildering range of possibilities, such that without additional

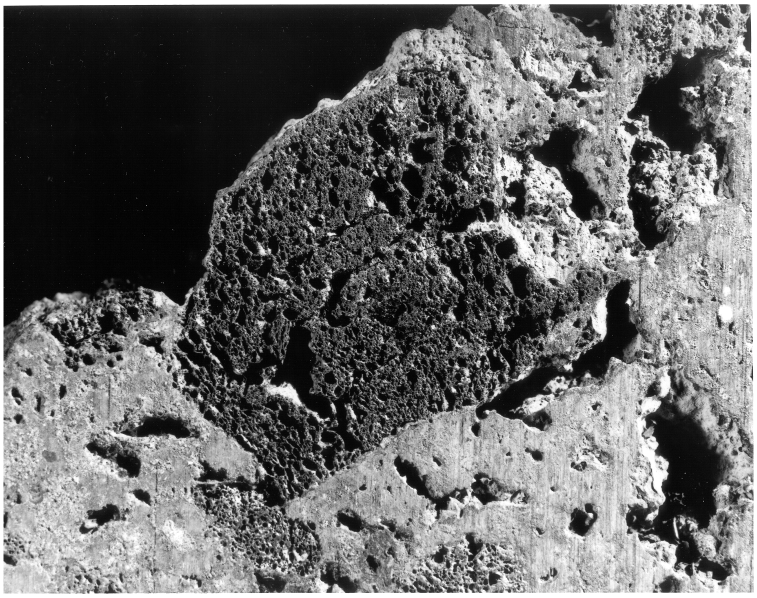

Figure 2 Detail of Figure 1 showing one of the highly pyrolized lumps of charcoal. (Photo. British Museum / A. Milton)

metallurgical or compositional information, any date is possible or explicable no matter what the real age of the object.

First, it is necessary to outline the smelting processes used through the last 3000 yr, as far as they are known, concentrating on the potential sources and ultimate disposition of the carbon present in these processes.

IRON AND STEEL PRODUCTION: CARBON IN—CARBON OUT

The iron ores themselves could well contain significant quantities of carbon in the form of carbonates; siderite, $FeCO_3$, was one of the more common ores used in the past (Pounds 1971), and other ores are often intermixed with limestone. For example, siderite was the main ore used by the Wealden iron industry in southern Britain from the Iron Age to the Industrial Revolution (Cleere and Crossley 1985:11–5). Carbon present in carbonates is expected to be of geological age.

Iron Production by the Direct Process

Because of its high melting point (about 1550 °C), iron was traditionally made by solid state processes often referred to as direct processes. In these, the fuel and reductant was invariably charcoal. In antiquity it is likely that wood of any age would often have been used as a source of the charcoal, and this state of affairs continued until at least the Renaissance in Europe, as illustrated in Biringuccio's *Pirotechnia* (Smith and Gnudi 1942:177–9), published in 1540, on all aspects of metallurgy and which contains a woodcut illustration showing a woodman about to place a venerable tree bole into the charcoal-making pit (Figure 3). Eylon (2002) has drawn attention to the traditional preference for old oak trees to make charcoal for metallurgical purposes in Lebanon. If these sources of charcoal were used to smelt or carburize iron, then the carbon date could predate the iron by centuries.

Figure 3 Illustration from Biringuccio's *Pirotechnia* of 1540, showing an old tree bole being converted into charcoal for use as a metallurgical fuel, the carbon from which could have been laid down centuries earlier.

However, in the post-Medieval period in Europe, and probably much earlier in the Roman period (Fiqueiral in Fulford and Allen 1992) and also in China, production of iron on an industrial scale required a regular supply of charcoal on a large scale. This could only realistically have been met from properly managed woodland producing timber from pollarded trees that were unlikely to have been growing for more than a very few decades when converted to charcoal. In these instances, the carbon date would be close to the date of the metallurgical operations.

It was common practice to roast the ores before smelting, which should have converted most of the carbonates present to oxides (Maréchal 1985; Rostoker and Bronson 1990:52–3). Once the ore was charged into the furnace, further roasting could be expected in the upper or outer reaches of the reaction zone (depending on the furnace type), which were more oxidizing before the ore entered the lower or central region of the furnace where the reduction took place. It seems, however, very likely that some unroasted ore would have dropped straight into the reaction zone where the CO produced by its decomposition could enter the forming iron in the same manner as the CO from the burning fuel.

In the reaction zone of the furnace, the iron minerals were reduced to tiny platelets of metallic iron protected from reoxidation by the slag. This is where the nascent iron began to coalesce into a solid mass which inevitably contained quantities of the slag in which it had formed. The resulting solid lump of iron, known as the *bloom* in English, had to be hammered to weld the iron into a more coherent mass and to expel as much of the slag as possible. In order to keep the slag molten, the bloom had to be kept at red heat which required frequent reheating on a hearth. Once an acceptable iron billet had been produced, it could then be forged, that is hammered to shape, an operation once again necessitating that it be frequently returned to the hearth. The hearths were normally fueled with charcoal, but mineral coal, often in conjunction with charcoal, has been found on metalworking hearths, dating from the last 2000 yr at least, all over the Old World in areas where coal abounds from Roman Britain (Webster 1955) to Han China (Wagner 1993).

These processes produced wrought iron with highly variable carbon contents, normally ranging between about 0.01% and 0.1% carbon. However, sometimes the carbon content was appreciably higher and there were a very few direct processes in various parts of the world that could regularly produce bloomery iron with a carbon content in region of 0.1 to 1.0%; that is, the iron was produced as a steel.

Otherwise, steel was produced by carburization of the smelted iron, that is, by prolonged heating of the solid iron billet or artifact in a charcoal hearth, where under intensely reducing conditions some of the CO gas from the burning charcoal would have been decomposed to deposit elemental carbon on the surface of the iron into which it could dissolve and diffuse. In addition to the charcoal, a variety of other carbonaceous materials, scraps of leather, horn, rags, etc., were recommended by various authorities on iron-making to encase the iron, and all of them were materials that are likely to have been growing very shortly before their use as carburizing agents. The process, sometimes referred to as *case hardening* if performed on the forged artifact, was slow, laborious, and the results uncertain and heterogeneous.

The obvious solution was to produce iron and steel in liquid forms, and this was achieved at earlier dates and was far more widespread than is presently credited (Craddock 1998, 2003). The implications of this for the viability of ^{14}C dating iron are disturbing.

Iron Production by the Indirect Process

In the so-called *blast furnace*, or indirect process, the iron was produced in liquid form known as *pig iron* or *cast iron* (irrespective of whether or not it was destined to be a casting). The blast furnace process operated at a higher temperature under markedly more reducing conditions than the bloomery process, such that the forming iron could dissolve the elemental carbon that was deposited on it. Even small quantities of carbon dissolved in iron reduce the melting point by several hundreds of degrees centigrade such that the product was molten iron that could be run from the furnace as pig iron, typically containing between about 3% and 5% of carbon. The very reducing conditions meant that separate fluxes had to be added to form the slag and limestone has always been a popular flux mineral. The limestone was not calcined before being charged to the furnace (Tylecote 1976:119).

For the blast furnace process, charcoal was the usual fuel initially, and has remained in use on a limited scale to the present day in countries such as Brazil where timber resources are perceived to be inexhaustible. Fossil fuel in the form of coke or anthracite, seems to have been used in China from the mid-1st millennium AD, developing into a major industry by the beginning of the 2nd millennium AD (Hartwell 1966, 1967). In the Far East, charcoal-smelting of iron continued on an industrial scale alongside fossil fuel-smelting and is still practiced today to a limited extent, with the obvious potential for confusion when scrap iron made by the 2 processes is mixed. Examination of a selection of Chinese iron castings showed that both charcoal- and coke-smelted iron was being used through the last thousand yr (Craddock et al., forthcoming). In the West, smelting with fossil fuel began in the 18th century and coke was the pre-eminent fuel by the 19th century (Tylecote 1976: 105–20), although, even in Europe and North America (Gordon 1996:90–124) charcoal-smelting continued on a limited scale well into the 20th century.

Cast iron is a rather intractable material such that the majority of it, in Medieval Europe for example, was converted into wrought iron by burning out the carbon in a process known in English as *fining*. Usually this was achieved by remelting the iron on a charcoal-fired hearth exposed to the air, and stirring. The iron became progressively more pasty as the carbon content fell and the melting point rose. Some of the iron oxidized and this was removed as a slag by adding fluxes that could themselves contain more limestone, as exemplified by the calcium-rich inclusions in the iron plate found on the Gizeh Pyramid (Craddock and Lang 1993). In the post-Medieval period in Europe, coal was sometimes used on a separate hearth to keep the metal at red heat, but conditions in this hearth were unlikely to have permitted much, if any carbon (or sulphur), to enter the metal. The wrought iron produced by fining is presently indistinguishable from bloomery wrought iron (Rostoker and Dvorak 1990; Starley 1999), and to no small degree this is the reason why the use of the blast furnace process over much of Eurasia in the distant past is so little appreciated. This exemplifies the problem prevalent with so much iron production, namely, that the final processes will have removed evidence of the putative earlier stages that potentially could have introduced carbon of geological age.

A variant process used in 18th and 19th century Europe, and possibly also in China at much earlier dates, is known as *puddling*. Here, the cast iron was melted by heat supplied indirectly by coal and the decarburization was achieved in part by the addition of iron oxide. The puddling was carried out in a reverbatory furnace where there was no direct contact between the coal and the metal. The gases burning over the metal would have been carbon monoxide from the coal, but this should not have entered the iron in any quantity as the whole point of the operation was to burn out the carbon the iron already contained, rather than introduce more.

The production of cast iron began in China early in the 1st millennium BC, and was probably used initially just for castings, but soon became the usual method of producing iron whether for castings, for fining to wrought iron, or for partial decarburization to steel (Wagner 1993).

In Europe, this sequence of developments seems to have been reversed, with all the early cast iron being turned into wrought iron. There is now archaeological evidence of iron-smelting sites where the blast furnace process was developing from the beginning of the 2nd millennium AD. These smelting sites have calcium-rich slags, and are always associated with fining hearths, and predate the first iron castings in Europe by several centuries (Magnusson 1985; Gassman et al. 1995; Knau and Sonnecken 1994). Note, the evidence is all from the smelting sites, there is presently no way of recognizing the fined iron produced by these furnaces in the surviving iron artifacts.

The situation in central and southern Asia and in the Middle East is not clear. Contemporary documentary evidence would seem to suggest that the blast furnace process may have been widely used throughout Islamic lands (Allan 1979:72-5) and India (Craddock 1998), but as yet there is no artifactual or archaeological evidence.

Steel

Steel could be produced from cast iron by controlled de-carburization, stopping just short of the total removal of the carbon, although this was only practiced to a minor extent. The usual practice was to start with wrought iron and subject it to a carburization process. In addition to the case hardening processes mentioned above, a number of other processes developed.

One method of exposing the wrought iron to carbon that it could easily absorb was to soak solid wrought iron in liquid cast iron, so that some of the carbon transferred to the wrought iron converting it to steel. The best description of this process is given by Biringuccio (Smith and Gnudi 1942: 67–70). It was practiced in Renaissance Italy, where it was known as the *Brescian process*, but it seems likely that the process was fairly widely used across Eurasia. In Biringuccio's description, the bath of molten cast iron, which would have been smelted with charcoal, was protected from oxidation by a slag formed from marble chips. This would have decomposed releasing the carbon from the geological-age carbonates, some of which could have dissolved into the cast iron. As with many other processes, the Brescian process is known mainly from a very few contemporary descriptions, the history and extent of the technology is completely unknown.

In post-Medieval Europe the *cementation process* was developed for the production of steel. Here, bars of wrought iron were packed into large stone chests with charcoal and heated for periods of up to several weeks before being removed and forged to bars, often referred to as *shear* and *double shear* steel (Barraclough 1984a). By the 19th century, there is the possibility that some of the wrought iron bars could have been smelted with coke, and after the cementation process with charcoal, the residual coke-derived carbon from the original iron-smelting process together with the charcoal-derived carbon could combine to make the iron appear some centuries older than it actually was.

Bulk steel production began with the Bessemer process in the 1850s, followed by the open hearth process in the 1880s (Barraclough 1990:39–56, 137–63). Both of these made steel by burning the carbon out of pig iron at very high temperatures. Usually the pig iron was freshly made metal, smelted with coke. Such fuel as was used in the open hearth process was coke and, thus, all the carbon was of geological age, and this could not be confused with charcoal-smelted iron. However, this was bulk steel, and a fair amount of the charge was often made up of scrap, which could of course include charcoal-smelted iron, Sweden, for example, was still a major producer of charcoal-smelted iron well into the 20th century. The remnant carbon from such a mixture could give a wide range of carbon dates.

Crucible Steel

Steel could also be produced as a liquid, and this technology has a long history, the extent of which is only now being fully realized (Bronson 1986; Craddock 1998, 2003; Allan and Gilmour 2000).

In Asia, 2 very different crucible processes for the production of liquid steel evolved from the latter part of the 1st millennium BC (Craddock 1998, 2003; Lang et al. 1998). In 1 process, wrought iron and carbonaceous materials were strongly heated in small crucibles, causing some of the carbon to enter the iron, forming a steel typically with about 1 or 2 percent of carbon. The carbonaceous material came from a variety of wood and other plant sources that are unlikely to have been of any great age at the time of their use in the crucible steel process. Alternatively, a mixture of wrought iron and cast iron could be melted together in the crucibles to give a steel once again typically containing between 1 and 2 percent of carbon. Little is known of the production of cast iron for the crucible steel process; if Chinese cast iron were used, then clearly it could contain carbon from fossil fuel. The Indian crucibles were regularly heavily tempered with straw, probably coming chiefly from the use of dung as a temper, and it is suggested that some of the carbon from these tempers could have entered the iron.

By the early Medieval period, crucible steel was in common use throughout the Middle East and South Asia as the usual quality steel for a wide range of tasks, well beyond the more familiar Damascus-patterned blades, as the recent study of Allan and Gilmour (2000) has demonstrated.

In Europe, a very different approach to the crucible steel process was developed utilizing cementation steel. This was melted to homogenize it and to remove the slag. The process was introduced by Benjamin Huntsman in Sheffield in the 1740s and by the early 19th century it was the usual method of producing crucible steel in Europe and North America. A recent report suggests that liquid steel was being produced in Saxon England almost a thousand yr earlier, although nothing is presently known about the process that could have produced it (Mack et al. 2000). This does, however, serve to illustrate how incomplete our knowledge is of the various ironmaking processes.

Latterly, the Huntsman process was largely supplanted in Europe and in North America by other crucible processes. The co-fusion process was quite widely used in the 2nd half of the 19th century, and in-situ carburization of the iron in the crucible with charcoal became general by the end of the 19th century (Carnegie 1913:51; Craddock and Wayman 2000).

Although the carbon in the cementation iron was derived from charcoal, and the reductants, where used, were also of charcoal, it was common practice to add either graphite, or latterly coke dust to the clay from which the crucibles were made, and this increased the carbon content of the steel (Barraclough 1984b: Table 1, p 54; Freestone and Tite 1986). Also, from the end of the 19th century at the latest, the wrought iron feedstock in all of these crucible processes would normally have been coke smelted.

Thus, overall, there are strong possibilities of intermixing carbon from recent and ancient sources, especially from charcoal and fossil fuel, and where charcoal- and fossil fuel-smelted cast iron exist together to be mixed as scrap. The problems created by multi-stage processes using carbon from different sources are well-exemplified by the following case history.

THE CHURCHDOWN LUMP

The lump of iron shown in Figure 1 was found buried in the surface of pasture land in the village of Churchdown in Gloucestershire, where it had clearly lain for some time. There were no other indications of iron production in the immediate vicinity, although Churchdown lies only a few km from the traditional iron-smelting area of the Forest of Dean, from where the lump very possibly came. From its shape and the vitreous material on the surface, the iron had clearly been very hot, and the finder, Mr Michael Hynd, initially wondered if it could be a sideritic meteorite, and cut a large slice from the center of the lump. This revealed the presence of numerous subrectangular areas, which although very ferruginous, were also very dark and had an open, somewhat pyrolized structure. Mr Hynd took his find to the Natural History Museum in London, where they were able inform him straight away that it was not a meteorite but could be an iron bloom, and suggested that he take it to the British Museum.

On first seeing the sectioned lump, the rather pyrolized structure of the black areas suggested that they were coke. If this was the case, then the iron could either have been a lump of cast iron, which had been coke-melted in a cupola furnace at some foundry and would have been of little interes,t or it could be a piece of cast iron in the process of being fined with coke, an unusual but not impossible scenario, and a rare if not unique survival of an intermediate stage of wrought iron production from the time of the Industrial Revolution. The other method of converting cast iron to wrought iron, puddling, was unlikely as that process used coal out of contact with the iron and the forming wrought iron has a distinctive slag content that was entirely absent here. However, expert examination by Caroline Cartwright, of the Department of Scientific Research, British Museum, showed that the black pieces had in fact once been charcoal, now heavily pyrolized and infiltrated with iron, such that it was impossible to determine the species of wood, beyond that it was likely to have been a hard wood (Figure 2). Also, at this stage, preliminary metallographic examination showed that the iron was predominantly of ferrite with a little heterogeneously distributed pearlite, that is a wrought iron. Given the size, shape, unconsolidated appearance, and the inclusions of charcoal, it now seemed very likely that this was a bloom of iron straight from the direct process furnace. This would also be a very rare survival indeed, and one that the Museum would wish to acquire and study (It is hoped that the metallographic report on this piece be published in the *Journal of the Historical Metallurgy Society*). This immediately raised another more practical problem, the lump was without any immediate context but found near to the Forest of Dean where iron had been smelted from the Iron Age to the 20th century (Hart 1971:1–209), and, thus, we had no idea at all of the date. We did not even know which was the appropriate Department in the British Museum to access it!

Thus, a selection of the iron-rich black pieces was sent to Arizona for AMS ^{14}C dating. The sample was pretreated in 1N HCl for several days and a considerable amount of Fe was dissolved. The remaining material was combusted at 900 ^{o}C with CuO. Two separate combustions were made. The sample CO_2 was then reduced to graphite over iron powder and the graphite was pressed into an AMS target. The sample target was mounted in a sample wheel with other samples and 8 standards (oxalic acid I and II, NIST SRM-4990B and 4990C). The measurements were made as described by Donahue et al. (1990a, 1990b).

The results were:

AA - 42636A16.0% C 35,940 ∀ 760

AA - 42636B9.4% C 36,230 ∀ 800

This showed that fossil fuel had been used, seemingly suggesting that the black pieces were coke as originally thought. Caroline Cartwright once again examined the pieces and was adamant that, although severely modified, the pieces were, or had been, charcoal. This time she actually took a small sample of the material that she believed was charcoal, and that was not in contact with the iron (this was not as easy as it might seem, as the iron and carbonaceous material are in very intimate contact). This sample (4.5 mg) was sent to Arizona and pretreated with the standard acid-alkali acid pretreatment. The result was as follows:

AA - 44357 66.5% C2,798 ∀ 43

The high C content of this sample clearly indicates that it is charcoal. Since this represents the age of the wood, it is clearly a better estimate of the time of firing of the iron than the earlier date.

In the meantime, further metallography done on the lump itself showed that the iron had significant quantities of iron sulphide inclusions, which together with the geological age of the first sample, strongly suggested that the iron had indeed been smelted with fossil fuel.

The possibilities were either to take the dates at face value and argue that this was coke-smelted iron which was being fined with charcoal made from ancient bog timber, or to seek other explanations. The first option is extremely unlikely, there is no record of the use of bog timber for making charcoal or indeed for its use in industrial processes generally, and there is no known source of such material within the Forest of Dean area.

It is more likely that the intense reducing conditions within the burning charcoal can provide the explanation. The overall purpose of the fining was to burn the carbon out of the putative coke-smelted cast iron as gaseous CO and CO_2. The function of the charcoal was to provide the heat to melt the iron and energy necessary. Thus, the original AMS ^{14}C date showed that the small amount of carbon left within the iron was the original carbon from the smelting process. This was to be expected as the whole purpose of the fining process was to create conditions where carbon left the iron, not entered it. However, where the gases had permeated into the intensely reducing conditions inside the burning charcoal, some of the CO_2 and CO had been reduced back to elemental carbon and deposited on the charcoal. Thus, the charcoal, produced from timber that had very likely been growing recently, was coated with carbon of geological age, coming ultimately from the coke used in smelting. This process could also explain the anomalous early date of the charcoal lump in one of the Frobisher blooms (see below).

If in the first instance, the carefully selected 2nd sample of charcoal had been sent to Arizona, there is a possibility that the Churchdown ingot could now be hailed as an important survival of early Iron Age iron production technology in Britain. As it is the combination of the AMS ^{14}C date of geological age from the iron itself, the much younger date from the charcoal and the numerous sulphide inclusions allows us to identify the lump as belonging to the charcoal-fining of coke-smelted iron (NB as described below the distinctive sulphur contents are only found in early coke-smelted irons).

As explained above, the use of coke to smelt iron only became general in Britain after the mid-18th century and the charcoal-fining process was finally superseded by the puddling process by the early 19th century. Thus, the AMS ^{14}C dates of geological age for the carbon in the iron and 2800 BP for the charcoal really do allow us to confidently assign this piece to between the late 18th and early 19th century.

DISCUSSION

There are clearly a number of potential contributions to the smelted iron from carbon other than that contained in the charcoal of the smelting process, which may itself have been made from wood of some age. It is proposed to review these in technological sequence, discussing some of the inherent problems in recognizing the presence of old carbon, from whatever source.

1. At every stage there is the possibility of carbonates associated with the ore surviving a putative roasting process.
2. The blast furnace process regularly used limestone as a flux. Cresswell (1992) has already noted that the presence of calcium in the slag inclusions in the iron is a danger signal that the carbon may be contaminated with carbon from the flux. However, blast furnace iron should retain few, if any, slag inclusions from the primary smelting, and if the iron has been fined to wrought iron then it will be impossible to establish even that it had been blast furnace iron, let alone whether or not it was fluxed with limestone. The inclusions that are present will be associated with the secondary fining process, but the majority of the remaining carbon will be from the original smelting, by whatever process. Given that the history of the blast furnace process commences very early in China, and that it is of very uncertain, but potentially early use in the rest of Eurasia, this puts the ^{14}C dating of wrought and cast iron on a very uncertain basis for most places at most times through the last 1000 yr or so. Any piece of wrought iron from a large part of the Old World over the last 1000 yr or so potentially may have been made by fining cast iron smelted with a limestone flux, or it might not—we presently have no way of telling from the iron itself.
3. In the fining of the iron, limestone was sometimes added as a flux, and coal was used on a minor scale in the hearths.
4. Old wrought iron was sometimes re-used by welding it together with other pieces of recycled or newly smelted iron.
5. Once iron was being melted, then part of the feedstock could be scrap iron. The danger here is two-fold. There is a danger of old charcoal-smelted iron forming part of the feedstock along with more recent charcoal-smelted iron. However, the problem becomes much worse if charcoal- and coke-smelted scrap is mixed. Where such mixed scrap is used, then the carbon from the resulting wrought or cast iron could give any ^{14}C date across the entire range covered by ^{14}C dating, and there is no warning or way of ascertaining that such admixtures are present in the iron. Qui Shihua and Cai Lianzhen (1986, quoted in Wagner 2001:52–3), turned the question round and used AMS ^{14}C dates of between 11500 BP and 13800 BP obtained from 3 artifacts of cast iron belonging stylistically to the Song and Yuan periods in China, and thus in reality rather under 1000 yr old, to calculate the relative proportions of charcoal- and fossil fuel-derived carbon that was likely to be present in these castings. They assumed that the carbon all derived from a single operation and from the figures that they had obtained, calculated the proportions of charcoal and coke that were likely to have been present in the original smelting

charge. Given the other imponderables such as the possible of use of limestone fluxes etc., the precise figures quoted of a furnace charge of 30% charcoal and 70% coal seem a little optimistic. In fact, the use of a mixed charcoal-fossil fuel mixture is otherwise unknown. The more likely scenario is that the mixture arose from melting together charcoal- and coke-smelted scrap iron. It must be of concern that there is no way of determining the real explanation.

6. Recognition of the presence of fossil fuel in the iron. It might seem that it would be relatively easy to spot iron that has been smelted with fossil fuel, even if the carbon date was equivocal, because of the high sulphur content, as was the case with the Churchdown lump, described above, for example. The first Chinese irons smelted with fossil fuel do indeed have recognizably higher sulphur contents than those smelted with charcoal (Han Rubin 1996; Craddock et al. in press), as does the first coke-smelted iron from England, which has given the impression that all iron smelted with fossil fuel has elevated sulphur contents. This is simply not so, as even trace levels of sulphur result in serious embrittlement during hot working of the metal, every effort was and is made to keep the sulphur content in iron to a minimum. The much higher temperatures achieved in the iron-making processes from the mid-19th century effectively reduced the sulphur content in the pig iron down to levels similar to those found in earlier charcoal-smelted iron.
7. The problems of establishing the origin of the carbon in steels are basically similar to those for iron generally. With crucible steels there is the special problem of the widespread use of graphite or coke as a constituent of the crucibles that held the molten charcoal-smelted cementation steel.

As noted in the “Introduction”, the material for dating is likely to fall into 1 of 2 categories, those pieces where there is some chronological or typological indication of the likely age of the piece, and those where there is none.

DATING MATERIAL FROM EXCAVATED, OR AT LEAST KNOWN, CONTEXTS

Even where there is some indication of period, and so that the processes behind the iron should be understood, there can still be problems of interpretation. The Frobisher blooms provide an interesting example.

The story behind the voyages of Martin Frobisher, who in the 1570s initially went in search of the North West passage around North America, before becoming side-tracked into an equally fruitless search for gold, is long and complex and has been told many times (McFee 1928; Kenyon 1975; Fitzhugh and Olin, editors 1993). Not least of the conundrums left by the expedition are the famous Frobisher blooms. A 19th century expedition to Frobisher’s base on Kodlunarn Island in Frobisher Bay off Baffin Island came back with several iron blooms, one of which was subsequently ^{14}C dated by Sayre et al. (1982) by gas counting using miniature counters. This gave a date of 679 ∀ 133 BP, calibrated to AD 1240–1400 at 1σ, using the calibration curve of Stuiver and Pearson 1986 (this and the following dates are taken from Harbottle et al. 1993:Table 10.1). The date is apparently much too old for the iron to be associated with the Frobisher expedition. The 1981 Smithsonian Expedition to Kodlunarn Island found 3 more blooms in association with a smithy. Two of the recently discovered blooms have been AMS ^{14}C dated (Harbottle et al. 1993). Once again, they gave dates that were apparently too old to have been associated with the Frobisher expedition. Suggestions had been put forward many years ago that the blooms discovered in the 19th century could have been made by the Vikings who were in the area centuries before Frobisher. However, the discovery of mineral coal at the smithy in association with the blooms raised other possibilities, and further complicated the matter. The main dating interest centers on Bloom 2, which was sectioned and samples taken from several locations. In 1991, Cresswell reported a date of 1340 ∀ 70 BP, (calibrated to 640–760 AD at 1σ) for a sample taken from the outside of Bloom 2, and a date of 920 ∀ 60 (calibrated to 1006–1150 AD at 1σ) for a small inclusion of charcoal found beneath when the section was cut. At this stage, although admitting the possibility of an earlier Viking origin, Cresswell seemed to have supported the explanation of Elizabethan smelting using a mixture of charcoal and mineral coal, but stated that additional dates from deeper within the bloom were necessary to resolve the problem. In 1992, Cresswell duly reported on 2 more dates on Bloom 2, 1 taken about 2 cm into the metal, which gave a date of 550 ∀ 60 BP (calibrated to AD 1307–1355 at 1σ), and one of 500 ∀ 60 BP (calibrated to AD 1400–1442 at 1σ), from about 5 cm depth (which was about the center of the sectioned bloom at that point). Cresswell noted the obvious, the further in the younger the age, but added, "These dates are closer to Elizabethan in origin, and suggest multiple sources of carbon used for the smelting." Thus, he still seemed to have believed the blooms to be Elizabethan. Yet, 1 yr on Harbottle et al. (1993) (with Cresswell as one of the authors) concluded on the basis of the same carbon dates that the blooms were after all likely to be Viking. A separate metallographic examination of all the metallurgical debris on the site including the blooms had concluded that although there was possible evidence of attempted smelting, the blooms were likely to have been earlier (Unglik 1993). Citing supporting documentary evidence (see below), both Harbottle et al. (1993) and Wayman and Ehrenreich (1993:213, fn) suggested that the Frobisher expedition had picked up the blooms locally, and had heated Bloom 2 strongly with coal, which had carburized the surface with geological age carbon.

The evidence supporting this interpretation is quite strong. It would seem to make no sense at all for the expedition to bring out half-formed iron as blooms from England when space on board the ships must have been at a premium (although there is an inventory of the materials carried by the expedition which lists "osmondes" of iron, and the word *osmonde* was sometimes used in Tudor England to denote ingots of iron) (Fitzhugh and Olin 1993:24). There is, however, another significant reference in the journal of Edward Fenton, captain of one of the expedition's ships on the third Frobisher expedition, that they had found "divers osmondes of iron", in the vicinity of Kodlunarn Island (Harbottle et al. 1993:174). These could be osmondes brought out on the previous expeditions, or the Frobisher expedition could have stumbled across an old Viking iron-making site, where unsuccessful blooms had been abandoned, which was common practice. So it could be that the expedition collected and tried to work them using some of the considerable quantity of mineral coal and charcoal brought out for the projected gold mining and working operations. Laeyendecker (1993) raised yet another possibility, namely that the expedition had smelted the iron locally, using old driftwood as a source of the charcoal.

However, our concern here is not primarily with the real origin of the Frobisher blooms, but merely to comment on the disparity of the dates. Bloom 2 has 4 dates spanning almost 1000 yr in the space of about 10 cm. If one takes the dates of the 2 inner samples as representing the age of the charcoal that was used for the near contemporary smelting of the iron, then there are still problems. The dates of the 14th and 15th centuries are rather too young for the Viking settlements that had shrunk away to almost nothing by this time, but conversely are still too old for the Elizabethans, unless some allowance is made for the possibility of some old carbon in the ore/flux, and for old wood being used for the charcoal or for coal being used on the hearth. Another problem is the discrepancy in the date of the charcoal inclusion in Bloom 2 and the dates of the 2 interior samples. The charcoal inclusion is apparently several centuries older than the carbon of the iron which it smelted. A possible explanation is that the later putative heat treatment with coal as well as carburizing the surface of the bloom also deposited some geological-age carbon on the charcoal, with a mechanism somewhat similar to that postulated for the charcoal in the Churchdown lump, described above, only with the carbon coming from the fuel rather than the iron.

Overall, the small counter and AMS ^{14}C dating of the Frobisher blooms has produced a lot of seemingly contradictory data, sometimes on the same bloom, without producing a firm "right" date or even consensus on the origin of the iron. It does exemplify again the problems of AMS ^{14}C dating of iron, especially where the possibility exists of both fossil fuel and charcoal being involved, together or at different stages in the operation.

DATING OF IRON WHICH HAS NO CONTEXT

In the 2003 paper by Cook et al. on the AMS ^{14}C dating of rusty iron, the opportunity was taken to publish the AMS ^{14}C dates obtained on some (unrusty) samples taken from 6 pieces of body armor that purport to be German or Italian of the 15th to 17th centuries AD, and are now in the collections of the Metropolitan Museum of Art, New York. Only 1 of the 6 dates was in broad agreement with the art historical dates, the other 5 had AMS ^{14}C dates varying from 570 BP back to 4250 BP. The authors suggest that some carbon could have come from the ore/ flux, the use of old charcoal (Figure 2), and the possible use of coal in the smithing hearths, which would all have tended to increase the apparent age and thereby explain some of the discrepancies**.** However, most of the discrepancies are very large, indicating to Cook et al. that a considerable percentage of the carbon in the iron must be of geological age. The explanation offered was that the iron was smelted with a mixture of charcoal and coal, and that the previously held view, that smelting iron with fossil fuel began later in Europe, would have to be revised.

There is, of course, a much more plausible explanation to accommodate the use of fossil fuel in the production of the iron that does not necessitate rewriting the history of metallurgy, namely that the armors are not of the age suggested by their typology, but are, in fact, 19th century copies. The carbon content of the iron could then be explained as either coming from the mixing of charcoal- and coke- smelted scrap iron, or by the carburizing of coke-smelted iron with charcoal.

There are a number of disturbing factors here. It is especially worrying that, no matter what the real date of the iron, the AMS ^{14}C dates show the general frequency of the admixture of carbon derived from both fossil fuel and charcoal in the iron. It is also disturbing that though the range of proffered AMS ^{14}C dates is very wide (from 230 to 4850 BP), with one exception they still manage to lie outside the almost equally wide range of realistic dates, which lie from 50 to 500 BP.

CONCLUSIONS

Overall, there is a strong possibility that iron submitted for ^{14}C dating will have been subjected to several processes involving heat, reduction, oxidation and carburization with a variety of fuels and fluxes containing carbon in a variety of forms, and this is likely to cause problems in the interpretation of the ^{14}C date.

In the examples quoted above of some Viking(?) blooms, "Medieval" armor, and the Industrial Revolution lump from Churchdown, one could have predicted that the carbon would have been from charcoal in the first 2 cases and coke in the 3rd. In reality, all turned out to be mixtures of unknown and unknowable proportions, fatally compromising the possibility of obtaining real dates by ^{14}C dating alone. That having been said, it must be stated that in each case the radiocarbon determinations did provide valuable information which, used in conjunction with other information, helped to elucidate the possible histories of the irons concerned, even if it could not provide a direct date as such.

However, the detailed and quite destructive sampling of the Frobisher bloom and the Churchdown lump, which was necessary, is clearly not possible on the majority of artifacts.

For authenticity investigations, an AMS ^{14}C date of many hundreds or thousands of yr for an iron artifact could be the result of charcoal-smelting at that time, or the fortuitous (or even possibly deliberate) mix of charcoal- and coke-smelted irons.

Where there is no independent indication of age, there are major and probably insurmountable difficulties in producing not just a reliable date, but even convincing evidence of age. This is due in the main to the possibility of unquantifiable intermixes of carbon of geological and recent age in the iron smelted or cast in the last couple of centuries being mistaken for charcoal-smelted iron of much greater age. For many classes of artifact, knowing that this possibility existed, it would be possible to offer a series of equally plausible explanations for almost any date from the recent past to the Upper Paleolithic.

For many periods and places, we simply do not know the basic technology by which the iron was produced. Even more worrying, in most instances the last process such as fining or crucible melting will have removed all tangible evidence of the earlier processes having taken place. Without the evidence of the original form of the carbon the various possible interpretations of the AMS ^{14}C dates obtained would be impossible either to refute or confirm.

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RADIOCARBON, Vol 44, Nr 3, 2002, p 733–738

THE ANTIQUITY OF THE PREHISTORIC SETTLEMENT OF THE CENTRAL-SOUTH BRAZILIAN COAST

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ABSTRACT. We discus here the prehistoric settlement of the central-south Brazilian coast, and, more specifically, 1 old radiocarbon date obtained for a costal shellmound, as well as its implications concerning the chronology attributed to the settlement process. The accelerator mass spectrometry (AMS) technique was used to determine the ^{14}C age of charcoal from a shellmound on the southern coast of Rio de Janeiro. The resulting age was 7860 ± 80 BP, an unexpected result that reinforces 2 similar previously obtained dates for the same region. Brazilian archaeologists, however, have questioned those 2 dates, because they would predate by some 2000 yr the antiquity consensually accepted for the settlement of the central-south Brazilian littoral.

INTRODUCTION

Prehistoric Settlement of the Central-South Brazilian Coast and Shellmound Chronology

This paper focuses on a phenomenon that happened in Brazilian prehistory: the settlement, in the central-south coast, of populations specialized in the exploitation of shellfish that reached a degree of social complexity that was unusual among hunter-gatherers (Lima 1997, 2000). The environmental characteristics of the Brazilian central-south coast, in association with socio-cultural factors, seem to have favored the emergence of the shellmound builders.

The dominant physiographic features of Brazil's south-southeast coast landscape are an abrupt, steep, forest-covered escarpment running parallel to the coast, reaching heights greater than 2000 m within a few km of the shoreline. There is also a narrow, quite irregular strip of coast, displaying a nearly continuous sequence of bays and lagoons edged with vast mangroves, as well as countless islands that are extensions of the continental relief.

Typical of estuarine systems where 2 neighboring ecosystems meet—land and sea—these ecotones comprise subsystems linked by the ebb and flow of tide and river waters, providing high nutrient influx, and displaying one of the highest productivity rates among marine ecosystems. These waters are veritable vivaria and one of the most fertile natural environments in the world.

Especially rich in mollusks, crustaceans, and fish, these ecosystems favored the settlement of prehistoric hunter-gatherers proceeding from inland highlands who arrived at the coast and became fishers and shellfish gatherers, shellfish being one of the most abundant and easily caught marine resources. These resources came to play a role of prime importance within their social system, as it was the search for greater proximity to mollusk beds that determined their settlement choices.

In these circumscribed landscapes, there is usually a high density of shellmounds, built by these fisher-gatherers, that stand out against the landscape. In some places, these mounds are as tall as 30 m. The most conspicuous and dense concentrations of shellmounds are found in the state of Santa

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Catarina. As one moves north through the states of Paraná, São Paulo, Rio de Janeiro, and Espírito Santo, they thin out progressively and disappear, as Brazil's straight northeastern coast does not offer the lagoonal areas with environmental conditions that favor this way of life. These shellmounds, dated in general between 6000 and 2000 BP, are the result of the intentional accumulation of food remains—notably shells and bones—and sediments. The material culture recovered in these shellmounds consists essentially of artifacts made from shells and bird bones, fish, sea and land mammals, including projectile points, ornaments, and other tools and weapons. Axes, milling stones, anvils, and hammer-stones made with basic rocks, together with abundant vein quartz flakes, make up the lithic industry, common to practically all shellmounds. These industries were simple in general, commonly crude, reflecting no great technical skill. Hearths, as well as human burials, are also found in the midst of this chaotic midden of shells, sediments, and bones.

Around 4000 BP, Santa Catarina was apparently the heartland of the phenomenon of progressive complexity between fishers-gatherers. As we move away from this core, there is a progressive diminishment in the evidence of emergent complexity among the shellmounds. In São Paulo and Rio de Janeiro, the concentrations of shellmounds are less dense and the mounds considerably smaller, having an average height of only 2 or 3 m.

The ^{14}C dating of shellmounds is crucial for the understanding of the rise, maintenance, and collapse of these socio-cultural systems that flourished in the course of 4000 yr, until the beginning of the Christian era, when their vestiges disappear. By this time, the bold and well-succeeded inland horticulturalists arrived at the littoral. Economically more powerful—since they were able to produce their own food—socially organized in more solid and concrete structures, technologically more advanced, and more expressive numerically, they ended up determining the absorption or extinction of the fisher-gatherers, in such a way that by the threshold of the 16th century, with the arrival of the Europeans to the Brazilian territory, those populations had long disappeared from the central-south coast.

There are currently some 290 ^{14}C dates available for Brazilian shellmounds, which attest to the initial occupation of the coast at around 6500 BP (Lima 2000). The frequency distribution of such dates shows that these cultures seem to have reached their peak between 5000 and 3000 BP (see Figure 1).

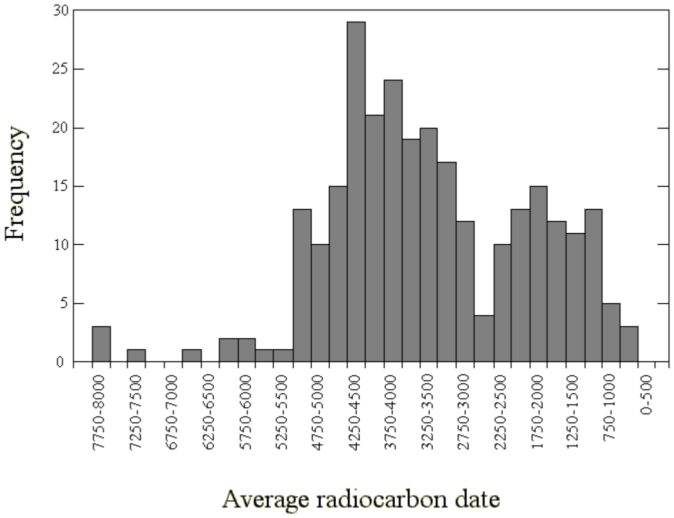

Figure 1 Histogram of average ^{14}C dates for the coastal shellmounds

Three very old ^{14}C dates were obtained decades ago for 2 different shellmounds; nevertheless, since they were considerably out of the average age range for these sites, they have been harshly contested by the professional community. In 1956, French archaeologists Joseph e Anette Laming Emperaire obtained 2 dates of 7803 ± 1300 BP and 7327 ± 1300 BP in Gif-sur-Yvette, France, for the Sambaqui de Maratuá, located in the region of Santos, São Paulo (Emperaire and Laming 1956). The dates were refuted due to the reason given above (Garcia 1979).

Twenty-four yr later, in 1981, while researching the Sambaqui de Camboinhas, in Rio de Janeiro (22°58′S 43°3′W), Lina Maria Kneip obtained a 2nd and slightly older date: 7958 ± 224 BP (SPC 207) (Kneip et al. 1981). Such a dating was also contested by the same reasons and by the supposition—which turned out to be mistaken (Muehe and Kneip 1995)—that the sand straps over which the shellmounds stood were more recent and would have been formed after the presumed date of the site. Until the present, both dates were viewed with strong distrust.

The Dating of the Sambaqui do Algodão

This paper is concerned with the dating of charcoal samples from the Sambaqui do Algodão. Located in a small island in the Ribeira Bay, in Angra dos Reis, Rio de Janeiro (22°55′48″S, 44°20′48″W), this site integrates a group of 7 shellmounds built very close together in different islets in the small Ariró Cove.

This site presents 2 distinct stratigraphic levels: the inferior, where an abundant capture of mollusks took place, and the superior, where there was a clear increase in fishing as a means of compensating the smaller availability of mollusks (see Figure 2). The superior level of the site has been dated previously as 3350 ± 80 (WSU 3359), in a study by Andrade Lima in the mid-1980s (Lima 1991). This result is compatible with the time range of occurrences for the shellmounds. The inferior level dating is reported in the present paper.

The accelerator mass spectrometry (AMS) technique was used for the ^{14}C dating of the charcoal sample, at the PRIME Lab of the Purdue University.

The charcoal sample was removed from the inferior level of the Sambaqui. The cleaning process began with the sample examination under microscope, where the gross impurities were removed. The usual chemical pretreatment for organic samples was performed. The sample was taken to PRIME Lab, where it was reduced to 1 to 2 mm thickness through the use of a razor blade. After being rinsed with ultra-pure water, the sample was treated with hydrochloric acid to remove the inorganic fraction. A base treatment, with sodium hydroxide, was done in order to remove the fulvic and humic fractions. Finally, another acid treatment removed the inorganic carbon that could have been incorporated to the sample during the base treatment. The entire chemical processing was performed at 95 °C. The sample was then dried and combusted at 900 °C in an evacuated quartz tube. Copper oxide was used as an oxidizing agent and a silver foil to remove sulphur compounds released during the oxidation process.

As carbon dioxide was obtained by combustion of the organic sample, the gas was injected into a vacuum line in order to be purified and transferred to the graphitization tube. A mixture of dry ice and ethanol was used to trap water from the gas and a liquid nitrogen trap froze the carbon dioxide while other gases were discarded. The graphitization tube consisted of a long quartz tube with a small tube, with iron, inside. A dimple in the bigger tube prevented the small one to touch its bottom, where the zinc was placed. In a vacuum, only the small tube was heated to 725 °C to remove any contaminants in the iron. The zinc was then heated to 550 °C to be distilled from the bottom to form a surface area layer in the upper part of the big tube. The bottom was sealed and removed. The CO_2 was transferred to the graphitization tube with a liquid nitrogen trap. Finally, the tube was sealed and heated in the oven for 700 °C for 10 hr. It was left to cool and heated again to 700 °C for 10 more hr. At this temperature, the CO_2 was reduced to CO by a reaction with Zn. Then, the CO decomposed, forming graphite by the iron-catalyzed reaction. After the process, the graphite was deposited into the inner tube with the iron and they were both pressed into the sample holder to be measured in the accelerator.

The 7.5 MV FN Tandem accelerator of the PRIME Lab was used for the measurement. The terminal voltage used was 4MV, and ^{13,14}C beams, with charge state of 3+, were selected by the high-energy magnetic analyzer and detected. The age was calculated as in Donahue et al. (1990) and the oxalic acid standard used was the NBS SRM 4990 C.

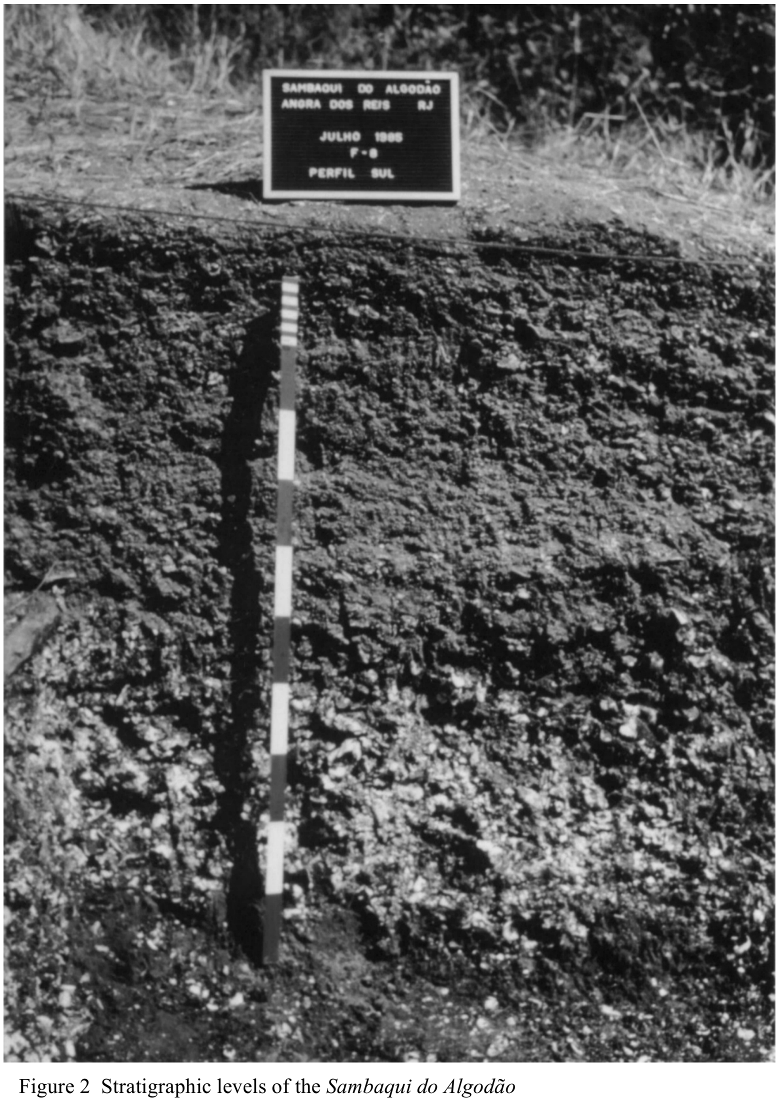

Figure 2 Stratigraphic levels of the *Sambaqui do Algodão*

RESULTS AND DISCUSSION

The results of the charcoal AMS measurements lead to 7860 ± 80 BP (PLID T00-0677) or cal AD 7050–6500 (2 σ) (OxCal Version 3.5; Oxford University). This result confirms and reinforces the 2 other dates previously considered dubious. This fact reopens the discussions about the antiquity of the settlement of the Brazilian coast.

What is interesting when we analyze the 3 occurrences together is the remarkable temporal proximity of the dates, practically contemporaneous—7958 ± 224 BP (Camboinhas), 7860 ± 80 BP (Algodão), 7803 ± 1300 BP (Maratuá)—as well as the geographical proximity between the sites, in the axis Rio de Janeiro/São Paulo. We are not dealing with dates obtained in distant points of the coast, but among very close sites, concentrated in the same area of the southeast region, and very far from the area of Santa Catarina.

CONCLUSIONS

In the context of the prehistory of the Brazilian central-south coast previously described, it would be admissible to assume that the oldest dates would be found among the shellmounds of Santa Catarina. Nevertheless, it is surprising that the oldest dates appear in Rio de Janeiro and São Paulo. This demonstrates that the initial settlements occurred in an area that does not coincide with the one in which those cultures reached their highest degree of expression.

The origin and routes through which the hunter-gatherers reached the coast and became the littoral's oldest fisher-gatherers are still questions to be answered, as there is no evidence of hunter-gatherers in the highland region of Rio de Janeiro. If we were to consider the Ribeira Valley, in São Paulo, as 1 of the few possible routes of communication between the coast and the countryside in the extensive barrier constituted by the Serra do Mar mountainous range, and still, the existence of sites with fluvial mollusks along the Ribeira River course, 1 of which dated 9000 BP (Blasis 2001), we could say that those fisher-gatherers could have originated in the São Paulo plateau, above all because the Sambaqui do Algodão, in the southern coast of Rio de Janeiro, is very close to the coast of São Paulo.

This 3rd date compels us to consider the 2 previously questioned dates, predating the traditionally accepted chronology for the settling of the coast by at least 2000 yr.

ACKNOWLEDGMENTS

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NO SYSTEMATIC EARLY BIAS TO MEDITERRANEAN ^{14}C AGES: RADIOCARBON MEASUREMENTS FROM TREE-RING AND AIR SAMPLES PROVIDE TIGHT LIMITS TO AGE OFFSETS

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ABSTRACT. Existing data and theory do not support a recent assertion that upwelling of old carbon has led to systematically 100–300 yr too old radiocarbon ages for the Mediterranean region. Similarly, the prehistoric tree-ring record produced over 3 decades by the Aegean Dendrochronology Project is shown to provide robust, well-replicated data, contrary to a recent unfounded assertion. ^{14}C and dendrochronology provide an accurate and precise chronometric framework for the Mediterranean region.

INTRODUCTION

In a recent paper, Keenan (2002) asserted that radiocarbon ages from the Mediterranean region from "earliest historical times (*sic*) until the mid-second millennium BC" are too old. He then put forward a hypothesis (upwelling of old carbon from the stagnant Mediterranean) to explain his initial assertion. Finally, he claimed that Anatolian dendrochronological evidence did not disprove his assertion or hypothesis. Further, he stated that the "Anatolian dendrochronology should be regarded as suspect and in need of independent scrutiny."

We respond as this paper is seriously flawed. We briefly review the evidence to show that:

1. There is no basis to his initial claim or starting point of systematically too old ^{14}C ages of "between one and three centuries", and instead good evidence to the contrary.
2. Keenan's review of literature in support for his theory is highly selective; there is, in fact, no sound database to support his claims.
3. The Anatolian dendrochronology, and in particular the key Bronze-Iron Age master sequence, is built on robust and well-replicated data using standard dendrochronological techniques.
4. Significantly, and inexplicably ignored by Keenan, ^{14}C research reported in 2001 using the Anatolian dendrochronology, in fact, demonstrates over long time intervals that there is no systematic distortion of Mediterranean ^{14}C ages versus those from the rest of the mid-latitude Northern Hemisphere. And, even at times of dramatic and rapid change in solar activity when a small short-lived offset has been detected between ^{14}C data on contemporary wood from the Mediterranean and Germany (and in turn Ireland), this is an order of magnitude less than Keenan's claim of disparities of between "one and three centuries."

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1. SYSTEMATICALLY TOO EARLY ^{14}C DATES IN THE EAST MEDITERRANEAN? NO

Keenan (2002:225) claims that ^{14}C ages are too old for the Mediterranean region from the "earliest historical times until the mid-second millennium BC" (*sic*–the earlier Holocene is meant). He makes this assertion not on the basis of unambiguous evidence, but instead, by the rather selective citation of some assorted publications. A few of these studies do report ^{14}C ages for some contexts older than the dates previously best estimated by archaeologists and ancient historians from little hard evidence through interpretation of various partial (2nd millennium BC) to largely non-existent (3rd millennium BC and earlier) proto-historical records and cultural associations, or speculative astronomical conjecture (e.g. Spence 2000; Rawlins et al. 2001), but none actually demonstrate ^{14}C ages systematically 100–300 yr older than any historically fixed date. The other literature cited consists of statements by archaeologists expressing concern that scientific dating techniques (most often ^{14}C) are sometimes yielding ages earlier than those conventionally assumed or best estimated but not known. Again, in no case, do any of these studies demonstrate ^{14}C ages significantly earlier than any actually known date. Moreover, in all cases careful and rigorous analysis of materials dated, and their association with the contexts for which dates are sought, would be necessary to support Keenan's assertion (cf. Bruins et al. 2003 and literature cited).

The major data resource is the study of Bonani et al. (2001), which reports ^{14}C ages for fragmentary organic samples obtained (with difficulty, in many cases) from a number of major Egyptian monuments. There are wide spreads of ages in several of the sets, which the team involved suggests to be partly accounted for by an "old wood" issue. All available trees in the region, of widely varying ages, were consumed by the pyramid builders and as older settlement debris was recycled in fires (Lehner et al. 1999); and the association of measured age for the sample (biological age unless other contaminating processes were involved) versus the date for monument construction is not demonstrated or clear in a number of instances (e.g. "charcoal" from mudbricks or from mortar [see Bonani et al. 2001:1297–98]—may easily represent "old" tree rings). Interestingly, the 2 secure datasets from early 2nd millennium BC Middle Kingdom monuments (Pyramid of Senusret II at Illahun and Pyramid of Amenemhet III at Dashur) yielded calibrated ages compatible with historical estimates (Bonani et al. 2001:1320 and Figure 1). This indicates no *a priori* problem with the 2nd millennium BC ^{14}C dates in the Mediterranean region, and, thus, negates Keenan's suggestion that other 2nd millennium BC ^{14}C series from the region may be too old. For the 3rd millennium BC Old Kingdom, Bonani et al. do report 17 date sets as older than the historical estimate, 6 as compatible, and 4 as more recent than the historical estimate. But, apart from noting that the historical age estimate is commonly regarded as ±100 yr for this period. The interpretation of Bonani et al. is based on the inappropriate use of average values for the ^{14}C age of sample sets, which contain significant internal variation, and is thus misleading. For example, an examination of Bonani et al. (2001:Figure 1) shows the Khafre Pyramid (object number 16) to yield one of the apparently tighter calibrated age ranges and to be some 2 centuries older than the estimated historical age. But examination of the 25 ^{14}C data from charcoal samples from the monument (Bonani et al. 2001:1306) reveals ages varying by 536 ^{14}C yr! As we show in Figure 1, a number of the individual samples do, in fact, offer calibrated ages compatible with the estimated historical age of 2558–2532 BC (Bonani et al. 2001:1316), and only some are older—"old" wood would appear the obvious 1st hypothesis (see Lehner et al. 1999). Such a pattern—younger ages corresponding to, or close to, context date and older ones reflecting old wood—is quite common and expected when dealing with wood/charcoal samples (for an example from Troy II, see Kromer, Korfmann and Jablonka 2002:48 and Figure 4). Similar observations may be made about the datasets for: Step Pyramid of Djoser at Saqqara, Temple Complex associated with the Step Pyramid, Pyramid of Sekhemkhet at Saqqara (Bonani et al. 2001:1303), Pyramid of Khufu at Giza (p.1305), Pyramid of Djedefre at Abu Roash, Sphinx Temple of Khafre at Giza (p.1306), Pyramid of Menkaure at Giza, Mortuary Temple of Shepseskaf at South Saqqara (p.1307), Mortuary Temple and Pyramid of Sahure at Abusir (p.1309), and Pyramid of Teti at Saqqara (p.1310). In contrast, it is notable that the ^{14}C ages from a modern excavation at the Royal Production Centre at Giza offer both a reasonably consistent set and calibrated ages more recent than the surrounding Old Kingdom datasets from the monuments (Bonani et al. 2001:Figure 1, object 12, contrasted with other objects 10–19). Similarly, the Pyramid of Snefru at Meydum offers interesting evidence (Bonani et al. 2001:1304). Six of the 7 dates are closely comparable (SMU-1412 on a "log" is either aberrant or very old wood nothwithstanding the stated dating of its "outer rings") and 5 of the determinations date outer rings from wood from the burial chamber or shaft thereto. The calibrated age range of the average of these 6 similar ^{14}C ages is entirely compatible with the historical age estimate (Bonani et al. 2001:1314). Thus, with appropriate samples or good contextual association, there is no evidence of any systematic ^{14}C offset of 100–300 yr as argued by Keenan (2002).

Meanwhile, Keenan has carefully avoided citing any of the other studies that have found that, in general, Mediterranean region ^{14}C dates usually agree perfectly well with the relatively secure early historic dates (e.g. Bruins et al. 2003; Hassan and Robinson 1987; Weninger 1990, 1997; Betancourt and Lawn 1984). Thus, for example, in the 14th–12th centuries BC, when vast numbers of material culture linkages tie the east Mediterranean regional chronologies together very tightly with a fairly solid Egyptian proto-historical chronology, ^{14}C evidence yields wholly compatible and mutually reinforcing data (e.g. Manning et al. 2001; Manning and Weninger 1992). Similarly, where there is reasonable to good proto-historical evidence for the date of the destructions in Palestine at the close of the Middle Bronze Age, a significant set of data (Jericho) yields consonant data (Bruins and van der Plicht 1995). Nor does Keenan note that detailed studies of ^{14}C evidence from, for example, the 3rd millennium BC Aegean region yield dates both consistent with conventional views and, in fact, sometimes younger than pre-existing archaeological opinion (e.g. Korfmann and Kromer 1993; Kromer, Korfmann and Jablonka 2002; Manning 1995, 1997). In contrast, the couple of well-known "problem" areas where ^{14}C and previous archaeological interpretation disagree, such as the start of the Aegean Late Bronze Age, are notable as periods where the conventional archaeological evidence for chronology is widely recognized as ambiguous and capable of alternative interpretations (e.g. Kemp and Merrillees 1980; Betancourt 1987, 1998; Hallager 1988; Manning 1999; Manning et al. 2002). These debates offer no support to the hypothesis of Keenan.

The test for Keenan's hypothesis would be ^{14}C data on independently and securely dated samples. Are they too old as he suggests, or not? Such material is not plentiful. Egypt is the obvious place to look, as here there is an historical chronology, with mutually reinforcing linkages with the independent Assyrian chronology, known within small errors back to the mid-2nd millennium BC, at least (Kitchen 1996a, 1996b, 2000, 2002; von Beckerath 1994, 1997). However, although analyses of available ^{14}C data from the 2nd millennium BC have found that dates are generally compatible with historical chronology (Shaw 1985; Hassan and Robinson 1987; Weninger 1990, 1997), much of the data employed is less than ideal or even appropriate. Most of the samples employed did not derive from modern archaeological excavation or they derived from monuments or objects not necessarily offering biological ages contemporary with the supposed historical connection. However, 1 suite of data from Egypt demands attention. These are 5 dates on a range of materials (bone, horn, skin, wood, and charcoal) collected specifically and carefully for a high-quality program of ^{14}C dating (Switsur, in Kemp 1984:178–188) from modern excavations at Tell el-Amarna (Akhetaten) (Kemp 1984). Amarna was the short-lived capital of Egypt during the "Amarna Age." Construction began

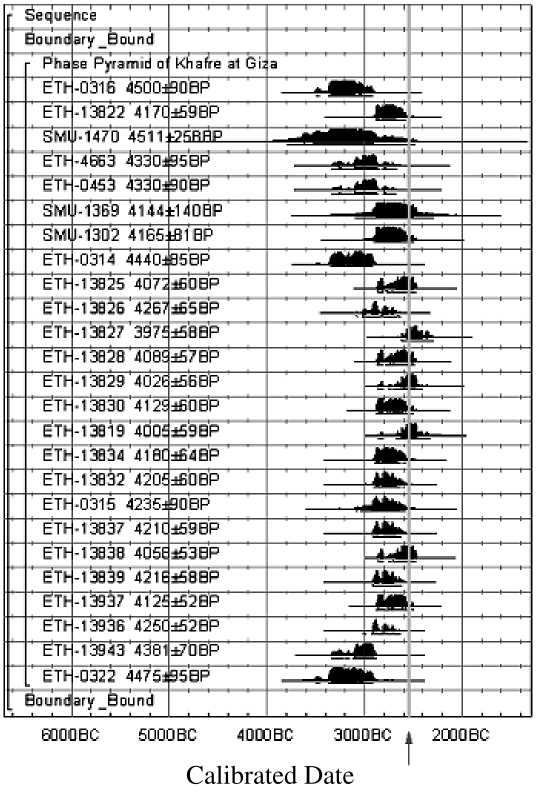

Figure 1 Calibrated age ranges for the ^{14}C ages reported from the Pyramid of Khafre at Giza by Bonani et al. (2001). The historical age estimate employed by Bonani et al. (2001) is 2558–2532 BC, indicated by the grey bar above the arrow. Samples are all of charcoal; they offer *termini post quos* ranges for human use. Eleven of the 25 samples—the more recent ones—offer ages compatible with this historical age estimate within their 2σ calibrated age ranges. The other older ages may, in most cases, be considered likely to reflect "old wood" or re-used material. The upper and lower lines under each histogram indicate, respectively, the 1σ (68.2%) and 2σ (95.4%) calibrated age ranges. Calibration and analysis employing OxCal 3.5 (Bronk Ramsey 1995; 2001 and later versions, with curve resolution set at 4) and INTCAL98 (Stuiver et al. 1998).

in year 4 of Amenhotep IV (Akhenaten) and the city became the capital by year 9; it was then no longer capital from about year 2 of Tutankhamun, and was being destroyed by the reign of Haremhab (Kemp 1984, 1987; Murnane 1995; Aldred 1988). The accession of Amenhotep IV is dated at about 1355–1351 BC and the accession of Haremhab about 1323–1319 BC by Kitchen and von Beckerath (Kitchen 1996a, 1996b, 2000, 2002; von Beckerath 1994, 1997). Letters preserved on clay tablets from the site (Moran 1992) provide synchronisms with Assyria and Babylonia and these confirm and require the dates given above within very narrow margins (Kitchen 1996a, 1996b, 2000, 2002; von Beckerath 1994, 1997). The specific context of the dated samples was a midden probably deposited early within the site's (very short) history and "thus during the reign of Akhenaten rather than that of Tutankhamun" (Switsur, in Kemp 1984:182–183). Hence, the historical date range might be narrowed to between about 1351/47 BC to 1338/34 BC. The Amarna ^{14}C ages on both known shorter-lived samples (skin, bone, and horn) and on the wood and charcoal samples tested, offer a tight and coherent set of results entirely consistent with the historical dates and very clearly provide no evidence at all for any systematic bias towards 100–300 yr too old ^{14}C ages as proposed by Keenan (2002) (see Figure 2).

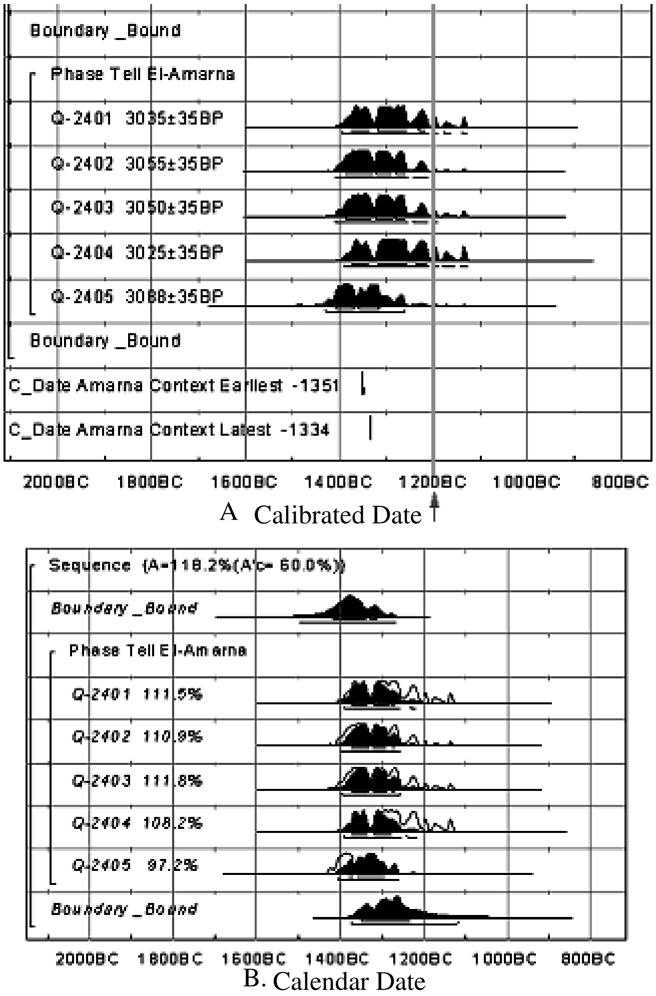

Figure 2 A) Calibrated calendar ages for the ^{14}C data reported from Tell el-Amarna, Egypt (Switsur in Kemp 1984:178–188) compared to the historical date for the context (see text—indicated by grey bar). The upper and lower lines under each histogram indicate, respectively, the 1σ (68.2%) and 2σ (95.4%) calibrated age ranges. B) Sequence analysis (solid histograms) of the Amarna data (with the individual probabilities from (A) indicated by the hollow histograms) as a phase within calculated boundaries. The Amarna data are entirely consistent with the historical age for the context and exhibit no evidence for any systematic bias for ^{14}C ages 100–300 yr older than real age as asserted by Keenan (2002) (indeed, if there is any scope for movement, it is in the opposite direction). Calibration and analysis employing OxCal 3.5 (Bronk Ramsey 1995; 2001 and later versions, with curve resolution set at 4) and INTCAL98 (Stuiver et al. 1998). Q-2401, wood; Q-2402, charcoal; Q-2403, skin; Q-2404, horn; Q-2505, bone. Weighted average of all 5 data: 3050 ± 16 BP (1), weighted average of just the 3 definitely shorter-lived samples 3054 ± 20 BP (2), 2σ (95.4%) confidence calibrated ranges respectively (1) 1388–1331 BC (46.6%), 1322–1260 BC (48.8%), and (2) 1393–1260 BC (94%), 1228–1222 BC (1.4%).

In sum, there is no body of evidence indicating systematic significantly too old ^{14}C ages compared with any robust historical dates for the east Mediterranean (and there is no proto-historic evidence prior to the mid-1st millennium BC for the central-west Mediterranean). But, rather than merely continuing to cite examples from the vast archaeological and archaeometric literature where the evidence is heavily weighted against Keenan's assertions, we instead offer a clear empirical test for his claim, and thereby, demonstrate that it is incorrect: see Section 4 below.

2. OLD SEA AND OLD AIR? REALITY

No one doubts that the reservoir age of the Mediterranean surface water has changed over time, nor that surficial sediments in deltaic plains, including in the Mediterranean, can yield significantly old ^{14}C ages due to erosion and transport of old carbon-bearing materials (Stanley 2000; Stanley and Hait 2000). The reservoir age of the modern pre-bomb Mediterranean, based on ^{14}C measurements of known-age shells, is on the order of 400 yr (Siani et al. 2000; Reimer and McCormac 2002). Unfortunately, there are currently no measurements of the marine reservoir age for the Mediterranean between the 19th century AD and about 3800 ^{14}C yr BP. Comparison of ^{14}C ages of planktonic foraminifera to those of associated tephra layers and of paired shell and charcoal samples support a reservoir age comparable to that of the modern pre-bomb measurements from about 3800–6000 ^{14}C yr BP (Facorellis et al. 1998; Siani et al. 2001). Between about 7400–8800 ^{14}C yr BP reservoir ages were larger at around 515 ± 22 ^{14}C yr (Facorellis et al. 1998). These increased reservoir ages are coincident with the S1 sapropel formation (Siani et al. 2001). Sapropel events are observed in sediment cores throughout the Mediterranean as 1 or 2 dark bands of high organic carbon content, which are formed during periods of summer insolation and monsoon intensification. These wet periods may increase water column stability, increase surface productivity and decrease ventilation of the deep water, which could result in increased surface reservoir ages (Mercone et al. 2000). Ba/Al ratios provide a more persistent criterion than organic carbon content or color for defining productivity pulses (Thomson et al. 1999). Ba/Al in 7 cores taken throughout the Mediterranean increases from background levels starting around 10,000 ^{14}C yr BP (marine, uncorrected) with peak levels between ~9000 to 6500 ^{14}C yr BP and ending ~5300 ^{14}C yr BP (Mercone et al. 2000). After that, Ba/Al ratios remain near background levels to the present day and no sapropel event more recent than S1 is observed in the Eastern Mediterranean cores (Mercone et al. 2000). The Mediterranean stagnation ended by ~5000 ^{14}C yr BP with increased overflow to the Atlantic as observed in the sedimentology and in the planktonic $\delta^{13}C$ of a series of cores east and west of the Gibraltar sill (Vergnaud-Grazzini et al. 1989), not the 1000–0 BC quoted from this same study by Keenan, and surface reservoir ages returned to near modern values by 6000 ^{14}C yr BP (Siani et al. 2001). All available evidence indicates approximate equivalency of the Mediterranean surface reservoir with the mid-Atlantic reservoir (Siani et al. 2001:1918 and refs.) with the exception of the sapropel event ~8500 yr B.P.

However, even if the Mediterranean surface reservoir age had been older than has been observed, there is little evidence that a large ocean reservoir age translates into a large air reservoir age. We presently lack recent marine-terrestrial data from the Mediterranean to demonstrate this, but an analogy exists from the North Atlantic. Here, we may compare data on the sea surface ^{14}C reservoir from sea shells against ^{14}C ages for tree rings growing "downwind" in the British Isles from the 19th–20th centuries AD (Figure 3). It is apparent that changes in sea surface reservoir age do not translate into changes in air reservoir ^{14}C ages as recorded by oaks in the British Isles. In general, regional differences have been difficult to observe in tree rings because they are of the order of the measurement error and may be masked by laboratory differences (McCormac et al. 1995). For instance, if we compare decadal ^{14}C measurements of tree rings from the northwest coast of the United States with those from the British Isles, we find an average offset from AD 950–1850 of just 4 ± 2 ^{14}C yr despite the upwelling of old water along the west coast of the United States (Stuiver et al. 1998; Hogg et al. 2002). Other regional differences of up to a few tens of yr in multi-ring and single-ring samples are summarized by Stuiver et al. (1998) and Stuiver and Braziunas (1998).

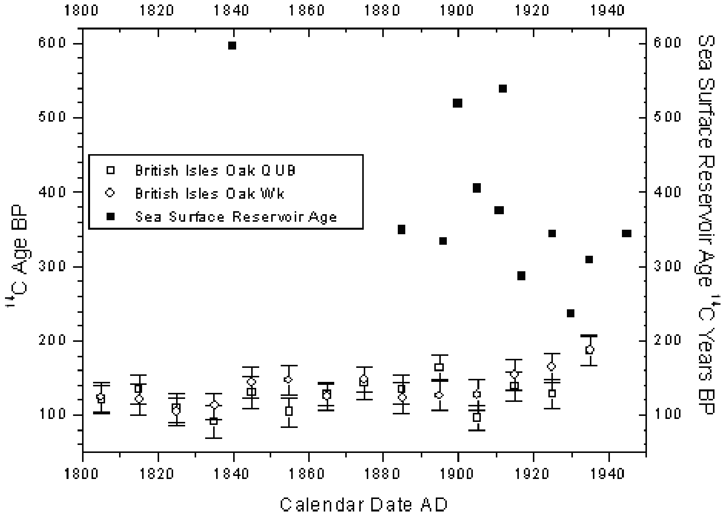

Figure 3 Comparison of ^{14}C ages for decadal samples of oak from the British Isles measured at the Queen's University Belfast (QUB) and Waikato (Wk) Laboratories (McCormac et al. 1998) versus the North Atlantic marine reservoir age as determined from measurements of sea shell data from <65 °N (and hence not potentially affected by changing ice cover) as tabulated from cited sources by Siani et al. (2000:Table 2 with refs. in text p 276). There is no correlation of sea surface reservoir age and air reservoir age as recorded in these downwind trees.

Turning now to the 5 specific examples cited by Keenan, we find that they appear to be highly selective and none of them actually provides support for systematic offsets of 1–3 centuries.

Keenan incorrectly states that trees in the northwestern United States, Olympic Peninsula, increased in ^{14}C age by 125 yr during 1868. This jump in ^{14}C age was observed in single-ring samples from trees growing on thawing permafrost in the MacKenzie River area of the Northwest Territories of Canada, in a particularly warm summer (Damon et al. 1996), not in trees from the Olympic Peninsula (Stuiver and Braziunas 1993; Stuiver et al. 1998). Thawing may have released CO_2 from centuries-old reservoirs of organic matter in close proximity to the location of tree uptake. This situation is not applicable to the scenario proposed by Keenan.

Hua et al. (2000a) gave $\Delta^{14}C$ results for single-ring samples from a cross-dated *Pinus kesiya* tree in northwestern Thailand which indicated depletions equivalent to 100–200 yr in 1953 and 1954, and stressed the need for confirmation of those results. Preliminary results from a longer series of data (1938–1951) were subsequently presented in a poster by Hua et al. (2000b) and showed depletions no greater than those for northwestern USA (Stuiver et al. 1998). This small depletion was observed despite air mass movement during the monsoon growing season from a potentially significant source of oceanic CO_2 outgassing in the Indian Ocean between 20° N and 5° S, where excess partial pressure of CO_2 in the surface ocean is up to 30 μatm (Keeling 1968) and the $\Delta^{14}C$ of surface water is low (~100‰ in 1977–1978, compared with ~140‰ at 30°S: Stuiver and Östlund 1983; 19th–early 20th century AD coastal reservoir ages of about 400–650 yr, equivalent to depletions of 50–80‰: Dutta et al. 2000a, 2001; Southon et al. 2002).

Bhushan et al. (1997) found an old ^{14}C age of air collected during the season of maximum upwelling in the Arabian Sea in only 1 of 8 sampling sites and concluded that "upwelling effects have to be very localized and time specific." Dutta et al. (2001b) reported variable ^{14}C in maritime air over the Bay of Bengal, but the abstract does not give details of locations and times.

We observe some further issues with respect to Keenan (2002). In Levin et al. (1987) the difference in atmospheric Δ^{14}C between the Northern Hemisphere and Neumayer Station on the Antarctic coast of the Weddell Sea was reported as –11‰ (equivalent to 88 yr, not 175 as claimed by Keenan). It was hypothesized that, based on unpublished South African data, the offset from equatorial latitudes could be greater. Meanwhile, however, a rich dataset exists and has been published (Levin and Hesshaimer 2000) but ignored by Keenan. The recent data result in a difference of Δ^{14}C between subtropical and Southern Ocean/Antarctic stations of about 3–5 ‰ (Levin and Hesshaimer 2000: Figure 3b), despite a more than 200‰ difference in surface water Δ^{14}C (Levin and Hesshaimer 2000:Figure 3c) (see Figure 4). We consider the Southern Ocean, and especially the Weddell Sea, the closest modern analogue of the scenario proposed by Keenan, as the surface waters are substantially depleted in ^{14}C and wind speeds are high, leading to enhanced gas exchange. Yet, the atmospheric memory of the old CO_2 is barely measurable (in fact, part of the difference may be caused by remnant bomb (^{14}C still being released during the 1990s from the tropical biosphere, as the difference has decreased in more recent years). On the other hand, Northern Hemisphere Δ^{14}C was higher in pre-industrial times and has been depressed relative to the Southern Hemisphere in the 20th century due to fossil fuel burning (Stuiver and Braziunas 1998; McCormac et al. 1998); the difference between the Southern Ocean and the subtropical Northern Hemisphere may, therefore, have been a little more than the current value of 3–5‰.

We disagree with Keenan's interpretation of the Rozanski et al. (1995) data as showing an atmospheric response to outgassing of old Pacific waters during an El Niño event. The very transient depletion occurred over a period of July to September in 1992. One of us operates a $^{14}CO_2$ sampling station in the equatorial region at Llano del Hato, Merida, Venezuela (early data shown in Rozanski et al. 1995), which, after more than 6 yr of monitoring to date, has not shown any ^{14}C depletion when compared with subtropical sites. One may argue that atmospheric diffusion acting in the transport of air from Ecuador to Venezuela masks the ^{14}C depletion, yet based on our measurement precision we would expect to be able to detect any significant systematic large-scale signal if there was one (the Ecuador site is about 250 km from the coast, and about 3000 m altitude; the Venezuela site is about 1000 km from the coast, and about 3600 m altitude).

In summary, there is currently little evidence anywhere for a sustained large-amplitude regional depletion of ^{14}C in terrestrial samples due to the influence of old CO_2 from the surface ocean and maritime air carried onshore. A limited number of measurements directly on maritime air show highly localized and variable results (Bhushan et al. 1997; Dutta et al. 2000b); such small-scale depleted air parcels would be expected to dissipate rapidly over short distances with atmospheric mixing, as is observed in air-sampling stations in the Southern Ocean/Antarctica (Figure 4). Where differences of up to a few ‰ (or a few tens of ^{14}C yr) do occur in tree-rings, they appear to vary on a relatively short timescale and may be partly or wholly due to other causes (McCormac et al. 1995; Damon 1995; Stuiver et al. 1998; Knox and McFadgen 2001; Kromer et al. 2001; Hogg et al. 2002; Hua et al. 2002). Even in extreme instances, such as in the Southern Ocean, where deep ventilation does occur, and some effect is observed in the air reservoir ^{14}C age as noted above, the terrestrial impact is nonetheless significantly less than required for Keenan's hypothesis. There is no evidence for such processes in the Mediterranean since the S1 sapropel episode.

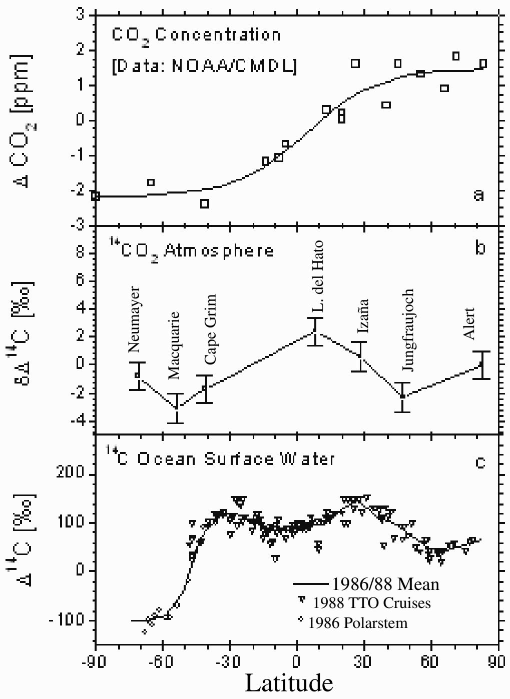

Figure 4 After Levin and Hesshaimer (2000:Figure 3). Mean meridional profiles 1993–1994 of a) CO_2 concentration (data from the NOAA/CMDL global network [Tans et al. 1996]) and b) $\Delta^{14}C$ in CO_2 in the atmosphere (Heidelberg unpublished data). Plotted in (a) and (b) are the deviations ΔCO_2 and $\delta\Delta^{14}C$ from the global mean values; (c) $\Delta^{14}C$ of CO_2 (Dissolved Inorganic Carbon) in surface ocean water derived from cruises of the TTO experiment (Broecker et al. 1995) together with unpublished Heidelberg data collected in 1986 in the South Atlantic Ocean during the Polarstern cruise ANT III. The solid line represents a spline through the 1986/1988 data.

3. ANATOLIAN DENDROCHRONOLOGY

Keenan states that there is "no dendrochronology for the region downwind from the Mediterranean" (2002:232)—exactly where such a "downwind" area lies is not defined, and it should be noted that his diagram (2002:Figure 1) reflects winter wind directions and not those for the key spring-summer growing season. He then turns to what he describes as "nearby" Anatolia—surely as Mediterranean as anywhere else he lists! Here there is an extensive dendrochronological record: the Aegean Dendrochronology Project (Kuniholm 1977, 1993, 1994, 1996; Kuniholm and Striker 1982, 1987; see also annual reports 1990-2001 at <http://www.arts.cornell.edu/dendro/>). This ADP work comprises absolute sequences from the present backwards (longest to the 4th century AD) for several tree species, then various floating sequences backwards over parts of 9 millennia, also in several tree species. Although the ADP began with the study of junipers from Anatolia, and in particular Gordion (Kuniholm 1977), for many years it has also investigated other species from much of the central and eastern Mediterranean and the Near East. In particular, and noted but essentially dismissed by Keenan, there is an extensive 1500-yr floating dendrochronology covering the late 3rd through earlier 1st millennia BC (Figure 5 and see Section 4 below). The core chronology comprises juniper (contra his assertion that different species are mixed); sequences for several other tree species also exist and correlate well to offer independent verification for much of this period. All crossdating employs established dendrochronological techniques (Cook and Kairiukstis 1990); the ADP in published reports has followed the European standards established by the laboratories in Belfast, Birmensdorf, and Hamburg. The statistics used include the standard student's t-test as modified by Baillie and Pilcher (1973) and trend coefficient (cf. Eckstein 1969:38-55), though, again following standard practice, priority is given to visual matching based on experience with given groups of samples (by species). Although Keenan devotes much of his "excursus on dendrochronology" to a critique of the exploratory D-value (Schmidt 1987), he mischaracterizes any use of this value in determining accepted crossdates.

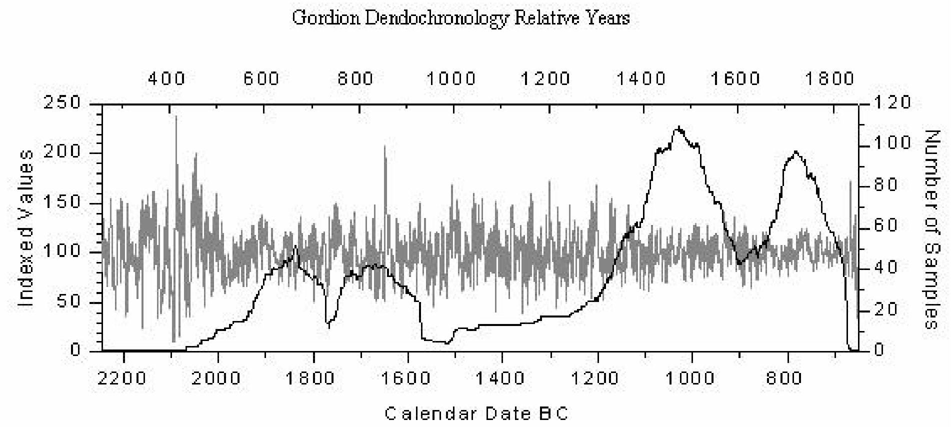

Figure 5 Aegean Dendrochronology Project Bronze-Iron Master Chronology as of AD 2002, shown in terms of the 20-yr moving average of the percent variation in ring-widths around normal (defined as 100) from all constituent data by yr (the "Index Values"—grey line). The number of securely cross-dated samples, an average of 32 trees per yr, which comprise this chronology is shown by the black line. The calendar date scale shown is the near-absolute dating proposed in Manning et al. (2001). For the specific trees from this chronology employed in the ^{14}C wiggle-match dating, see Figure 6. Although sample numbers are not especially large in the mid-16th century BC, we note that for the ^{14}C wiggle-match we employed a long-lived tree, GOR-161 with 861 tree rings, which grew from the 18th–10th centuries BC. It is securely cross-dated on the early end against dozens of juniper trees from Porsuk (Kuniholm et al. 1992 and on-going work since), and then against, progressively, dozens, scores, and finally over 100 trees from Gordion and environs. In addition to the data summarized above, newly developed juniper and pine dendrochronologies from the Hittite site of Kuşaklı match and so reinforce the earlier 17th to later 16th century BC interval. There is, thus, no possibility of dendrochronological error in the placement of the data shown in Figure 6.

In his "Excursus" Keenan purports to throw considerable doubt on the validity of the 30 yr of ADP work and sequences (of >10 million measurements from 9 millennia) through reference to the dating of 1 case—a "gateway." Keenan does not name the site—it is Tille Höyük—and he merely repeats previous misinformed claims by Porter, and repeated by Rohl (1985:389, with citations). Keenan fails to display a reading of the text by Kuniholm et al. (1993), where they explain what the samples comprise, and the other factors apart from simple statistics—the standard student's t-test and trend coefficient in addition to an excursus on the exploratory D-value—that were taken into account when offering a most likely fit for these undated samples against the Master Chronology. No one claimed this was an exact "scientific" fit for these samples—rather a best interpretation given all the available evidence. But, the fundamental point is that this discussion (Keenan:232, paragraphs 2–4) has nothing to do with invalidating the underlying Master Chronology, contrary to his assertion. At this time the ADP Bronze-Iron Age Master Chronology is a solid strongly-replicated set of—in total

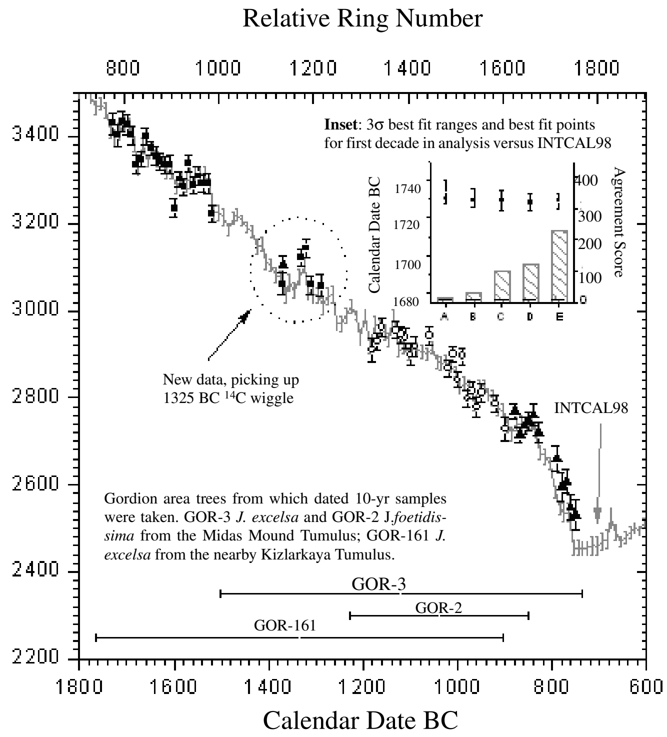

Figure 6 High-precision ^{14}C data, including 6 new data centered around the 1325 BC "wiggle" in the ^{14}C calibration curve, from 10-ring samples of the Aegean Dendrochronology Project Bronze-Iron tree-ring series (Manning et al. 2001 and refs.; Manning et al. 2003) compared at best fit placement against the current internationally recommended INTCAL98 ^{14}C calibration dataset (Stuiver et al. 1998). Samples were taken from 3 of the constituent trees of the well-replicated Gordion area dendrochronology forming 1 of the ADP floating sequences for the prehistoric Mediterranean and Near East. Data indicated by solid squares come from tree GOR-161, data indicated by hollow circles come from tree GOR-2, and data indicated by solid triangles come from tree GOR-3. All ^{14}C measurements were made at the Heidelberg ^{14}C laboratory (see Kromer et al. 2001; Manning et al. 2001 for details). The Heidelberg data include an error enlargement to allow for the likely maximum unexplained inter-laboratory error for the Heidelberg measurements versus Seattle data on similar German oak (Kromer et al. 2001:2530). Inset shows the derivation of the best fit placement for the data series shown under analysis using OxCal (Bronk Ramsey 1995; 2001 with curve resolution set at 1) versus the INTCAL98 dataset (Stuiver et al. 1998). The 3σ fit ranges and specific best fit points are shown versus the quality of fit (Agreement Score, with the horizontal bar across each column indicating the minimum 95% confidence threshold value). A: all data, n = 58. B: set with no 9–8th C BC data (see Kromer et al. 2001; Manning et al. 2001), n = 53. C: set excluding significant outliers from B (values under half the 95% agreement score), n = 49. D: set excluding the one significant outlier in analysis C, n = 48. E: set excluding all data from D, not exceeding an individual 95% agreement value, n = 42. The real errors on the fit described should also include a decade mis-matching allowance (estimated at an additional 2 calendar yr in Manning et al. (2001:2535 n.17), and an additional error for the likely average range of differences between relevant Northern Hemisphere ^{14}C calibration datasets (and possible other such datasets, were they in existence).

(*continued*) Data on Douglas-fir from the prevailing leeward side of the North Pacific Ocean versus British Isles oak from the prevailing leeward side of the North Atlantic Ocean should plausibly indicate a likely maximum factor (e.g. average difference AD 1720–1940 is calculated at 19 ± 3 ^{14}C yr by Knox and McFadgen 2001:98); of available individual datasets the bi-decadal British Isles oak data of Pearson et al. (1986) yields the largest divergence of best fit: +14 calendar yr (all data, n = 58, but poor agreement) or +12 calendar yr (n = 44 with no 9–8th century BC data (see Kromer et al. 2001; Manning et al. 2001) and significant outliers excluded—values under half the 95% agreement score). For the present case, however, comparison of much more proximate central European wood versus Turkish wood is likely to be rather closer in the absence of major ocean input or extreme altitude difference (e.g. for German oak versus Turkish pine the mean absolute difference over 23 paired data from AD 1420–1649 is only 1.4 ^{14}C yr: Kromer et al. 2001:2530). Two-thirds of the relevant part of the INTCAL98 calibration curve already consists of such wood. If the one-third Belfast component is removed, not surprisingly the wiggle-match range against just the Seattle laboratory data for oak from southern Germany (Stuiver, Reimer and Braziunas 1998) offers very similar best fits and total error ranges: the best fit across the same analysis models A-E above varies from +1 to +2 calendar yr and the overall 3σ fit ranges are within ±1–2 calendar yr. Thus, it is likely that overall real total errors will be only a little larger than those indicated in the inset. The choice of wiggle-matching approach employed (here Bayesian using OxCal) is not a significant variable as all current methods for fixed sequence ^{14}C curve fitting determine very similar to identical results (Bronk Ramsey et al. 2001)—demonstrated for the data in Figure 6 in Manning et al. (2001) and Manning et al. (2003).

at present—444 trees. The chronology is based around a core of many dozens of trees from the Gordion area, supported and verified by good juniper, pine, and cedar sequences from other sites.

4. ^{14}C AND ANATOLIAN DENDROCHRONOLOGY

We have an empirical test for whether there are systematic offsets to older ^{14}C ages for the east Mediterranean. We took an internally secure and extensively replicated long tree-ring record from the Mediterranean region covering the 2nd through earlier 1st millennia BC (Figure 5), and determined ^{14}C ages for long sequences of decadal samples from this chronology. The data closely match the standard international calibration dataset (Stuiver et al. 1998) comprised of analyses of German and Irish wood for this period, and do not indicate disparities of 100–300 yr (Kromer et al. 2001; Manning et al. 2001). Subsequent work further confirms these findings, notably picking up the sharp mid-14th century BC "wiggle" in the INTCAL98 calibration dataset (Stuiver et al. 1998), and, overall, offering a strong correlation for a total span of nearly 1000 calendar yr: see Figure 6. These data—58 high-precision ^{14}C determinations on wood from 3 securely cross-dated trees selected from a robust dendrochronology of 444 trees and 56,232 annual rings—and derived dendrochronological dates coordinate well with available proto-historical information (Manning et al. 2001; Veenhof 2000)—with any range for debate an order of magnitude less than the claimed 100–300 yr disparity asserted by Keenan. It is, thus, not possible that we have found a statistically "viable", but incorrect, wiggle-match. In further support of this assessment, we may note that the quality of fit achieved between the ^{14}C series from the BC period Bronze-Iron dendrochronology (Figure 6) is very similar to the fit observed when comparing ^{14}C measurements on known-age AD period Anatolian wood versus INTCAL98 (Figure 7). Thus, if there is no 100–300 yr disparity in the AD period (Keenan admits this, and plentiful evidence confirms this view), then there also cannot have been one in the 2nd through 1st millennia BC either, given both the quality and constancy of the fit, and the agreement of the BC period Bronze-Iron fit with secure historical dating at the recent end (especially 9th–7th centuries BC: see summary in Manning et al. 2001:2534).

In conclusion, available data from a variety of sources are incompatible with claimed systematic regional disparities of 100–300 yr. The only, and interesting, attested offset for the east Mediterranean is a short-lived, and much smaller one (albeit significant), in the 9th–8th century BC during a dramatic solar irradiance minimum (Kromer et al. 2001; Manning et al. 2001; van Geel et al. 1998). But this in no way supports the theory of Keenan (2002), and, in fact, rather demonstrates the opposite.

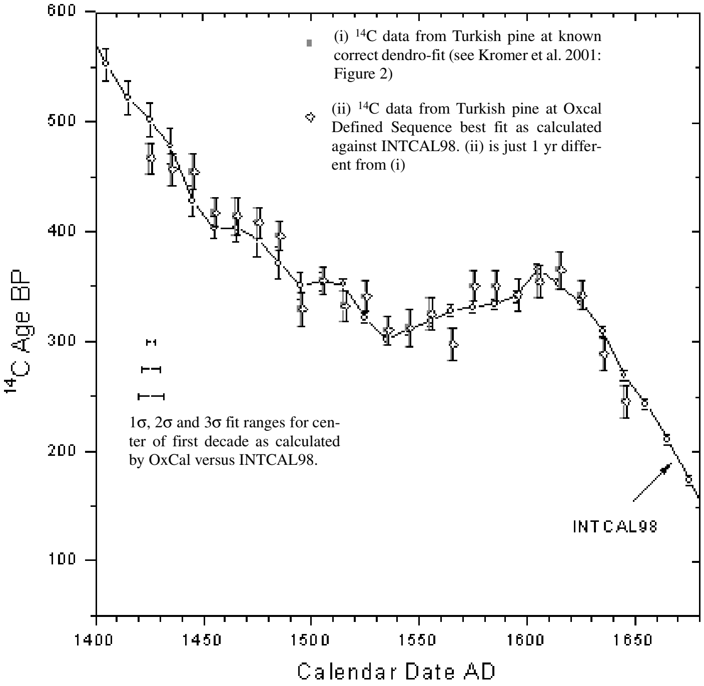

Figure 7 "Wiggle-match" fit of the AD period ^{14}C series on decadal samples of Turkish pine (Kromer et al. 2001:Fig.2) versus the INTCAL98 ^{14}C dataset using OxCal (Bronk Ramsey 1995; 2001, with curve resolution set at 1), compared with the verified/absolute tree ring ages. The ^{14}C wiggle-match best fit is just 1 calendar yr different from the correct date. Very similar results occur if the separate Douglas-fir dataset of Stuiver, Reimer and Braziunas (1998) or the separate Belfast British Isles oak dataset of Pearson et al. (1986) are employed, with the best fits again at AD 1426, just 1 year from the known dendro age. The 1σ, let alone the 2σ and 3σ, ranges around the best fit point include the correct age. Since the Turkish pine decades were cut to match INTCAL98, decade mis-matching is not an issue in this case. We observe a broadly similar quality of fit for the wiggle-match of the floating BC period Turkish wood against the INTCAL98 dataset in Figure 6.

ACKNOWLEDGMENTS

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ON CORRECTING ^{14}C AGES OF GASTROPOD SHELL CARBONATE FOR FRACTIONATION

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ABSTRACT. Correcting the ^{14}C age of a sample for fractionation is straightforward if the measured carbon was derived entirely from the atmosphere, either directly or through chemical and/or biological reactions that originated with atmospheric carbon. This correction is complicated in the case of gastropods that incorporate carbon from limestone or secondary carbonate (e.g. soil carbonate) during shell formation. The carbon isotopic composition of such gastropod shells is determined by fractionation, as well as mixing of carbon from sources with different isotopic values. Only the component of shell carbonate derived from atmospheric carbon should be corrected for fractionation. In this paper, the author derives a new expression for correcting the measured ^{14}C activity of gastropod shells for fractionation, and describe an iterative approach that allows the corrected ^{14}C activity and the fraction of shell carbonate derived from atmospheric carbon to be determined simultaneously.

INTRODUCTION

Carbon isotopes (^{12}C, ^{13}C, ^{14}C) fractionate along biological pathways because of differences in the rates of reaction for different molecular species (Clark and Fritz 1997). Typically, the lighter carbon isotope is taken up preferentially to the heavier isotope and, therefore, the stable and radiogenic isotopic ratios of biological products are lower than that of the reactants. For example, the isotopic composition of carbon in C_3 plants is nearly 20‰ lower than the atmospheric CO_2 from which it is derived. If the effects of fractionation were ignored, the ^{14}C age of material with a low δ^{13}C value would appear older than contemporaneous material with a high δ^{13}C value.

The effect of fractionation on ^{14}C is about twice the effect on ^{13}C (Wigley and Muller 1981), and therefore the δ^{13}C value of a sample can be used to correct the measured ^{14}C/^{13}C ratio ($A_{measured}$). The $A_{measured}$ value is determined by accelerator mass spectrometry (AMS), and is standardized to a δ^{13}C value of $-25‰_{pdb}$ (by convention) using the equation

$$A_{corrected} = A_{measured}\left(\frac{1000-25}{1000+\delta^{13}C}\right) \tag{1}$$

(Linick et al. 1986; Donahue et al. 1990). Note this equation is the correct form for AMS measurements of the ^{14}C/^{13}C ratio. The term in parentheses on the right side of the equation should be squared for AMS measurements of the ^{14}C/^{12}C ratio. The corrected ratio ($A_{corrected}$) is used to calculate the ^{14}C age of a sample.

Equation 1 is valid only if the measured carbon was derived entirely from the atmosphere, either directly or through chemical and/or biological reactions that originated with atmospheric carbon. For example, carbon fixated in plants by photosynthesis or in skeletal material by metabolic processes originates in the atmosphere. Similarly, carbon in shells of gastropods that do not incorporate limestone during shell formation is derived from the atmosphere, either directly or via consumption of plants. Any difference between the isotopic composition of these materials and atmospheric carbon is due entirely to fractionation, and Equation 1 can be applied.

CORRECTING FOR FRACTIONATION AND SOURCE MIXING

Correcting the $A_{measured}$ value of shell carbonate for gastropods that incorporate limestone or secondary carbonate is more complicated. Stable and radiogenic isotopic values of gastropod shell carbonate are determined by fractionation, as well as mixing of carbon from sources with different isotopic values (Goodfriend and Hood 1983). Only the component of shell carbonate derived from atmospheric carbon should be corrected for fractionation. Goodfriend and Hood (1983) previously derived an expression to correct the ^{14}C activity of this component for fractionation. However, their initial correction equation was in error (Equation 14; Goodfriend and Hood 1983), and, thus, their derived expression was also incorrect. The proper form of the correction equation for shell carbonate is

$$A_{corrected} = A_{measured}\left(\frac{1000-25}{1000+\delta^{13}C_{nlc}}\right) \tag{2}$$

where the subscript *nlc* denotes the component of shell carbonate derived from non-limestone carbon. $\delta^{13}C_{nlc}$ is related to the δ^{13}C value of the shell carbonate ($\delta^{13}C_{shell}$) by

$$\delta^{13}C_{shell} = f_{lc}\delta^{13}C_{lc} + f_{nlc}\delta^{13}C_{nlc} \tag{3}$$

where *f* is the fraction of shell carbonate derived from limestone (*lc*) and non-limestone (*nlc*) carbon. Goodfriend and Hood (1983) stated that the $\delta^{13}C_{lc}$ value for gastropods should be 0‰ (identical to limestone), and, thus, the $f_{lc}\delta^{13}C_{lc}$ term could be dropped. This is incorrect because carbon isotopes are fractionated during dissolution of limestone, which is composed of calcite, and the subsequent precipitation of shell carbonate, which is composed of aragonite. Laboratory experiments have shown the δ^{13}C value of synthetic aragonite precipitated from a bicarbonate solution is enriched by ~1.8‰ (1.8 ± 0.2‰—Rubinson and Clayton 1969; 1.7 ± 0.4‰ —Romanek et al. 1992) relative to calcite precipitated from an identical solution. The magnitude of enrichment is independent of temperature between 10 and 40°C (Romanek et al. 1992). If these experimental results can be applied to biogenic aragonite, and assuming equilibrium conditions prevail (i.e. no vital effects), the $\delta^{13}C_{lc}$ term in Equation 3 equals the δ^{13}C value of limestone (typically 0‰) plus the amount aragonite is enriched relative to calcite (denoted as $\Delta_{a\text{-}c}$) during the dissolution-precipitation process, or simply $\Delta_{a\text{-}c}$. Substituting $\Delta_{a\text{-}c}$ for $\delta^{13}C_{lc}$ and $(1\text{-}f_{nlc})$ for f_{lc} in Equation 3 gives

$$\delta^{13}C_{shell} = \left[\left(1-f_{nlc}\right)\Delta_{a-c}\right] + f_{nlc}\delta^{13}C_{nlc} \tag{4}$$

which can be simplified and solved for $\delta^{13}C_{nlc}$

$$\delta^{13}C_{nlc} = \left(\frac{\delta^{13}C_{shell}-\Delta_{a-c}}{f_{nlc}}\right) + \Delta_{a-c} \tag{5}$$

Equation 5 can then be combined with Equation 2 to give

$$A_{corrected} = A_{measured}\left[\frac{1000-25}{1000+\left(\dfrac{\delta^{13}C_{shell}-\Delta_{a-c}}{f_{nlc}}\right)+\Delta_{a-c}}\right] \tag{6}$$

Equation 6 cannot be solved directly because the f_{nlc} value cannot be determined independently of the $A_{corrected}$ value. However, an iterative approach described below can be used to solve for both parameters for live and fossil gastropods.

Application to live gastropods

The uncorrected ^{14}C activity of gastropod shell carbonate (A_{shell}) is related to the ^{14}C activity of the non-limestone carbon source by

$$A_{shell} = f_{lc} A_{lc} + f_{nlc} A_{nlc} \tag{7}$$

where the non-limestone component consists of carbon obtained via the atmosphere and live plants (they can be grouped together because their ^{14}C activities are identical). The $f_{lc}A_{lc}$ term can be dropped because the ^{14}C activity of limestone (A_{lc}) is zero. Rearranging gives

$$f_{nlc} = {}^{A_{shell}}\!\big/_{A_{nlc}} \tag{8}$$

For gastropods that consume only live vegetation, the A_{nlc} value can be quantified either through measurement of the atmosphere or live plants. In 2001, the ^{14}C activity of live plants was 1.0919 ± 0.0039 (n = 2; Pigati et al. submitted). Equations 6 and 8 can be solved iteratively as follows:

1. Begin by assuming $f_{nlc} = 1$ and solve Equation 6 for $A_{corrected}$.
2. Substitute the calculated $A_{corrected}$ value from step 1 for A_{shell} in Equation 8, and solve for f_{nlc}.
3. Substitute the calculated f_{nlc} value from step 2 into Equation 6 and solve for $A_{corrected}$.
4. Repeat steps 2 and 3 until the $A_{corrected}$ and f_{nlc} terms converge. This is usually achieved in 3–4 iterations.

An additional step is required for gastropods that include plant detritus (decaying leaves, wood, etc.) in their diet. The ^{14}C activity of plant detritus may be different from that of live vegetation because the ^{14}C activity of the atmosphere has decreased exponentially since cessation of aboveground testing of nuclear weapons (Manning et al. 1990; Meijer et al. 1995). The ^{14}C activity of homogenized plant detritus can be measured by AMS, which can then be used to estimate the mean ^{14}C activity of all plants (live and detritus) consumed by the gastropod by using

$$A_{plants} = A_{liveplants} f_{liveplants} + A_{detritus} f_{detritus} \tag{9}$$

The calculated A_{plants} value can then be combined with the ^{14}C activity of the atmosphere (A_{atmos}) to calculate the A_{nlc} value in Equation 8 using

$$A_{nlc} = A_{plants} f_{plants} + A_{atmos} f_{atmos} \tag{10}$$

There is disagreement regarding the relative contribution of plants (f_{plants}) and the atmosphere (f_{atmos}) toward the carbon isotopic composition of shell carbonate. Goodfriend and Hood (1983) suggested that 25–40% of inorganic carbon in gastropod shells is derived from plants, and 30–60% is derived from the atmosphere. Stott (2002), however, found that atmospheric CO_2 plays little to no role in determining the isotopic composition of gastropod shell carbonate. Until this is resolved, Equation 10 may be solved using a range of atmospheric (0–60%) and plant (25–100%) values to determine a range of corrected ^{14}C activities.

Application to fossil gastropods

There are 2 approaches for correcting ^{14}C ages obtained from fossil gastropod shells for fractionation. One is to measure the ^{14}C activity of live specimens collected from geologic settings that maximize the potential for ingestion of limestone. Equations 6 and 8 can then be solved to determine a species-specific, worst-case f_{nlc} value for the live specimens, which is assumed to be invariant through time. This should be done using the same species that are to be used for ^{14}C dating in the fossil record. Equations 6 and 8 can then also be solved using an f_{nlc} value of 1 (i.e. no limestone correction) to determine a range of possible $A_{corrected}$ values.

For cases in which measuring the ^{14}C activity of live specimens is not possible, Equations 6 and 8 can be solved using f_{nlc} values of ~0.6 and 1 to calculate a range of possible $A_{corrected}$ values. The lower f_{nlc} value is the minimum observed by Goodfriend and Hood (1983) and Pigati et al. (submitted). The upper value assumes limestone and/or secondary carbonate are not incorporated by the gastropod during shell formation. The difference in the $A_{corrected}$ values between those corrected using a f_{nlc} value of 0.6 and those corrected using a f_{nlc} value of 1 is 1.1% for gastropods feeding exclusively on C_3 biomass, and 0.1% for those feeding on C_4 biomass. These corrections are quite small compared to age anomalies due to incorporation of limestone during shell formation.

PROPOGATION OF UNCERTAINTIES

The uncertainty associated with the $A_{corrected}$ term in Equation 6 is given by

$$\Delta A_{corrected} = \left(A^2 + B^2 + C^2 + D^2\right)^{1/2} \tag{11}$$

where

$$A = \left[\frac{975}{1000 + \Delta_{a-c} + \left(\frac{\delta^{13}C_{shell} - \Delta_{a-c}}{f_{nlc}}\right)}\right]\Delta A_{measured}$$

$$B = \left\{\frac{-975A_{measured}\left(1 - \frac{1}{f_{nlc}}\right)}{\left[1000 + \Delta_{a-c} + \left(\frac{\delta^{13}C_{shell} - \Delta_{a-c}}{f_{nlc}}\right)\right]^2}\right\}\Delta\left(\Delta_{a-c}\right)$$

$$C = \left\{ \frac{-975 A_{measured}}{f_{nlc} \left[1000 + \Delta_{a-c} + \left(\frac{\delta^{13}C_{shell} - \Delta_{a-c}}{f_{nlc}} \right) \right]^2} \right\} \Delta\delta^{13}C_{shell}$$

$$D = \left\{ \frac{975 A_{measured} \left(\delta^{13}C_{shell} - \Delta_{a-c} \right)}{f_{nlc}^2 \left[1000 + \Delta_{a-c} + \left(\frac{\delta^{13}C_{shell} - \Delta_{a-c}}{f_{nlc}} \right) \right]^2} \right\} \Delta f_{nlc}$$

where the delta (Δ) symbol at the end of each equation denotes the uncertainty associated with the individual parameter.

The uncertainty associated with the f_{nlc} term in Equation 8 is given by

$$\Delta f_{nlc} = \left[\left(\frac{\Delta A_{shell}}{A_{nlc}} \right)^2 + \left(\frac{-A_{shell} \Delta A_{nlc}}{A_{nlc}^2} \right)^2 \right]^{1/2} \tag{12}$$

where A_{shell} is the same as $A_{measured}$ in Equation 11. Equations 11 and 12 can be solved iteratively in a spreadsheet simultaneously with Equations 6 and 8. The magnitude of these uncertainties is certainly small compared to other sources of error in the ^{14}C age calculation, but nonetheless can be propagated for completeness.

SUMMARY

Correcting the measured ^{14}C activity of gastropods that incorporate limestone or secondary carbonate during shell formation is complicated by the fact that the isotopic composition of the shell carbonate is a function of fractionation and mixing of carbon from sources with different isotopic values. Only the component of shell carbonate derived from atmospheric carbon should be corrected for fractionation. The equations presented here can be used to determine the corrected ^{14}C activity of gastropod shell carbonate, as well as the fraction of the carbonate derived from non-limestone sources. For gastropods that do not incorporate limestone during shell formation (i.e. $f_{nlc} = 1$), Equation 6 simplifies to the standard correction equation (Equation 1).

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RADIOCARBON UPDATES

Glenn Goodfriend Symposium

A symposium to honor Glenn Goodfriend has been approved for inclusion in the Geological Society of America's (GSA) annual meeting in Seattle, 2–5 November 2003.

Details about the symposium are forthcoming. Further information is available at http://www.geosociety.org/meetings. The session is jointly sponsored by GSA's Archaeological Geology Division, GSA's Quaternary Geology and Geomorphology Division, the Geochemical Society, the Paleontological Society, and the people who vet the Marine geoscience abstracts.

Interested participants are invited to submit abstracts for their posters or talks using the abstract submission system on the GSA website. This system will be operational from late May until the abstract deadline, which is usually in late June. For more details on abstract submission, please check the GSA website. If you have other questions, please contact symposium organizer Bonnie Blackwell (bonnie.a.b.blackwell@williams.edu).

New CalPal Edition

A new edition of CalPal (Cologne Radiocarbon Calibration & Paleoclimate Research Package) can be downloaded from http://www.calpal.de. Along with a number of refinements in graphic output, there are two main new features in the new edition, both supporting archaeological and palaeoclimate research in the Holocene and Glacial periods.

First, the CalCurveComparer is completed. This is a twin-window dialog with easy-to-use functions (e.g Add & Remove files) to study the properties of all ^{14}C data sets and climate proxies that may be of interest in refinement and Glacial extension of the ^{14}C-age calibration curve. A climate box supports the synchronisation and visual fine-tuning of climate proxy age models (e.g. ice cores) and corresponding (e.g. marine, lacustrine) ^{14}C-data sets.

Second, beginning with this edition of CalPal, all calibration programs are equipped with a slider, by which we have fingertip control over the shape of the calibration spline. This feature will be useful when studying the influence of the calcurve shape on radiocarbon age models.

Additional details can be taken from the update-log: http://www.calpal.de/calpal/update.htm.

RADIOCARBON LABORATORIES

This is *Radiocarbon*'s annual list of active radiocarbon laboratories and personnel known to us. Conventional beta-counting facilities are listed in Part I, and accelerator mass spectrometry (AMS) facilities in Part II. Laboratory code designations, used to identify published dates, are given to the left of the listing. (See p 787 ff. for a complete list of past and present lab codes.)

Please notify us of any changes in staff, addresses, or other contact information.

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AA	NSF-Ariz. AMS Facility	USA
AAR	University of Aarhus	Denmark
AC	Ingeis	Argentina
AECV	Alberta Environmental Center of Vegreville	Canada
AERIK*	Atomic Energy Res. Inst.	Korea
ALG*	Algiers	Algeria
ANL*	Argonne Nat. Lab., Ill.	USA
ANTW	Antwerp	Belgium
ANU	Australian National University	Australia
ANUA	ANU Accelerator	Australia
AU*	University of Alaska	USA
B	Bern	Switzerland
Ba	Bratislava	Slovakia
BC*	Brooklyn College	USA
Beta	Beta Analytic	USA
BGS	Brock University	Canada
Birm	Birmingham	UK
Bln	Berlin	Germany
BM	British Museum	England
BONN*	Universität Bonn	Germany
BS	Birbal Sahni Institute	India
C*	Chicago	USA
CAMS	Center for Accelerator Mass Spectrometry	USA
CAR*	Univ. College, Cardiff	Wales
CENA	Centro Energia Nuclear na Agricultura	Brazil
CG	Institute of Geology	China
CH	Chemistry Laboratory	India
CRCA	Cairo	Egypt
CSIC	Geochronology Lab, IQFR-CSIC, Madrid	Spain
CSM*	Cosmochemistry Lab. USSR Academy of Sciences	USSR
CT*	Caltech, Calif. Inst. Tech.	USA
CU	Charles University	Czech Republic
D*	Dublin, Trinity College	Ireland
Dak*	Univ. de Dakar	République du Sénégal
DAL*	Dalhousie University	Canada
DE*	USGS, Denver	USA
Deb	Debrecen	Hungary
DEM	NCSR Demokritos	Greece
DIC*	Dicar Corp and Dicarb Radioisotope Company	USA
DRI*	Desert Research Institute	USA
ENEA	ENEA, Bologna	Italy
ETH	ETH/AMS Facility	Switzerland
F	Florence	Italy
Fr	Freiberg	Germany
Fra	Frankfurt	Germany
FSU*	Florida State University	USA
FZ	Fortaleza	Brazil
G*	Göteborg	Sweden
GAK	Gakushuin University	Japan
Gd	Gliwice	Poland
GD*	Gdansk	Poland
Gif	Gif sur Yvette	France
Gif A	Gif sur Yvette and Orsay	France
GIN	Geological Institute	Russia
GL*	Geochronological Lab.	England
Gro*	Groningen	The Netherlands
GrN	Groningen	The Netherlands
GrA	Groningen AMS	The Netherlands
GSC	Geological Survey	Canada
GU	Scottish Universities Research & Reactor Centre (formerly Glasgow University)	Scotland
GX	Geochron Laboratories	USA
H*	Heidelberg	West Germany
HAM	Hamburg	Germany
HAR*	Harwell	England
Hd	Heidelberg	Germany
Hel	Helsinki	Finland
HIG*	Hawaii Inst. of Geophys.	USA
HL	Second Institute of Oceanography	China
HNS*	Hasleton-Nuclear, Palo Alto, California	USA
Hv	Hannover	Germany
I	Teledyne Isotopes	USA
IAEA	International Atomic Energy Agency	Austria
IAEA-MEL	Marine Environmental Laboratory	Monaco
ICEN	Instituto Tecnológico e Nuclear	Portugal
IEMAE	Institute of Evolutionary Morphology and Animal Ecology	Russia
IGAN	Institute of Geography	Russia
IGS*	Inst. of Geological Sci.,	Sweden
II*	Isotopes, Inc., Palo Alto	USA
IOAN	Institute of Oceanography	Russia
IORAN	Institute of Oceanology	Russia
IRPA	Royal Institute of Cultural Heritage	Belgium
ISGS*	Illinois State Geological Survey	USA
IVAN	Institute of Volcanology	Ukraine
IVIC*	Caracas	Venezuela
IWP	Institute of Water Problems	Russia
J	Max-Planck-Institut für Biogeochemie, Jena	Germany
JGS	Geological Survey of Japan	Japan
JUBR	Biren Roy Research Laboratory	India
K	National Museum	Denmark
KAERI*	Korean Atomic Energy Research Institute	Korea
KCP	National Cultural Property Research Institute	Korea
KEEA	Kyushu Environmental Evaluation Association	Japan
KI	Kiel	Germany

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* Indicates laboratories that are closed, no longer measuring ^{14}C, or operating under a different code designation.

KIA	Kiel AMS	Germany
Ki (KIEV)	Institute of Radio-Geochemistry of the Environment	Ukraine
KN	Köln	Germany
KR	Krakow	Poland
KRIL	Krasnoyarsk Institute	Russia
KSU	Kyoto Sangyo University	Japan
L*	Lamont-Doherty	USA
LAR	Liège State University	Belgium
LE	St. Petersburg	Russia
LIH	NCSR Demokritos	Greece
LJ*	Scripps (UCSD) La Jolla	USA
LOD	Lodz	Poland
LP	La Plata	Argentina
Lu	Lund	Sweden
LU	St. Petersburg State Univesity	Russia
Lv*	Louvain-la-Neuve	Belgium
Ly	University of Lyon	France
LZ	Umweltforschungs-zentrum Leipzig-Halle	Germany
M*	University of Michigan	USA
Ma*	University of Winnepeg	Canada
MAG	Quaternary Geology and Geochronology Laboratory	Russia
MC*	Centre Scientifique de Monaco	Monaco
METU	Middle East Technical University	Turkey
ML*	Miami	USA
Mo*	Verdanski Inst. of Geochemistry, Moscow	USSR
MOC*	Archaeological Institute, Czechoslovak Acad. of Sci.	Czechoslovakia
MP*	Magnolia Petroleum	USA
MRRI*	Marine Resources Research Institute	USA
MSU	Moscow	Russia
N	Nishina Memorial	Japan
NIST	National Institute of Standards and Technology	USA
NPL*	National Physical Laboratory, Middlesex	England
NS*	Nova Scotia Research Foundation	Canada
NSRL	INSTAAR – University of Colorado	USA
NSTF*	Nuclear Science and Technology Facility, State Univ. of New York	USA
NSW*	U. of New South Wales	Australia
NTU	National Taiwan University	Republic of China
NU	Nihon University	Japan
Ny*	Nancy, Centre de Recherches Radiogéologiques	France
NZ	New Zealand	New Zealand
NZA	New Zealand	New Zealand
O*	Humble Oil & Refining	USA
OBDY	ORSTOM Bondy	France
ORINS*	Oak Ridge Institute of Nuclear Studies	USA
OWU*	Ohio Wesleyan Univ.	USA
OX*	USDA Oxford, Mississippi	USA
OxA	Oxford Radiocarbon Accelerator Unit	England
P*	Univ. of Pennsylvania	USA
Pi*	Pisa	Italy
PI	Permafrost Institute	Russia
PIC*	Packard	USA
PITT*	University of Pittsburgh	USA
Poz	Pozan	Poland
Pr*	Prague	Czechoslovakia
PSU*	Pennsylvania State Univ.	USA
PKU	Peking University	China
PL	Purdue Rare Isotope Measurement Laboratory	USA
PRL	Physical Research Laboratory	India
Pta	Pretoria	South Africa
Q	Cambridge	England
QL*	Quaternary Isotope Laboratory	USA
QC*	Queens College	USA
QU*	Centre de Recherches Minérales, Québec	Canada
R	Rome	Italy
RCD	Radiocarbon Dating	England
RI*	Radiochemistry, Inc.	USA
RIDDL*	Simon Fraser Univ.	Canada
Riga	Institute of Science	Latvia
RL*	Radiocarbon, Ltd.	USA
Rome	Department of Earth Sciences, Rome	Italy
RT	Rehovot	Israel
RU*	Rice University	USA
S*	Saskatchewan	Canada
Sa*	Saclay, Gif-sur-Yvette	France
Sac	Instituto Tecnológico e Nuclear	Portugal
SFU*	Simon Fraser Univ.	Canada
Sh*	Shell Development Co.	USA
SI*	Smithsonian Institution	USA
SL*	Sharp Laboratories	USA
SM*	Mobil Oil Corp., Dallas (formerly Magnolia & Socony Mobil Oil)	USA
SMU*	Southern Methodist Univ.	USA
SNU	Seoul National Univ.	Korea
SOAN	Institute of Geology and Geophysics	Russia
SR*	Salisbury, Rhodesia	Rhodesia

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Su	Finland	Finland
SUA	Sydney University	Australia
SWA	Swansea	Wales
T	Trondheim	Norway
TA	Tartu	Estonia
TAM*	Texas A & M University	USA
TB	Tblisi	Georgia
TBNC*	Kaman Instruments (formerly Texas-Bio-Nuclear)	USA
TEM*	Temple University	USA
TF*	Tata Institute of Fundamental Research	India
TK	University of Tokyo	Japan
TKU	Turku	Finland
Tln	Tallinn	Estonia
TO	IsoTrace Laboratory	Canada
TUNC*	Tehran University Nuclear Centre	Iran
Tx*	Texas	USA
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Ua	Uppsala AMS	Sweden
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UBAR	University of Barcelona	Spain
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UCLA	University of California, Los Angeles	USA
UCR	University of California, Riverside	USA
UD	Udine	Italy
UGa	University of Georgia	USA
UGRA	University of Granada	Spain
UM*	University of Miami	USA
UQ	University of Quebec at Montreal	Canada
URCRM	Ukrainian Research Ctr. for Radiation Medicine	Ukraine
URU	University of Uruguay	Uruguay
USGS*	USGS, Menlo Park	USA
UtC	Utrecht van de Graaff Laboratorium	The Netherlands
UW*	University of Washington	USA
V*	Melbourne, Victoria	Australia
VRI	Vienna Radium Institute	Austria
W*	USGS, National Center	USA
WAT	University of Waterloo	Canada
WHAMS	National Ocean Sciences AMS Facility	USA
WIS	Wisconsin	USA
Wk	University of Waikato	New Zealand
WRD*	USGS Washington, D.C. Water Resources Division	USA
WSU*	Washington State Univ.	USA
XLLQ	Xian Laboratory of Loess and Quaternary Geology	China
X*	Whitworth College	USA
Y*	Yale University	USA
Ya*	Yale University	USA
Z	Zagreb	Croatia

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