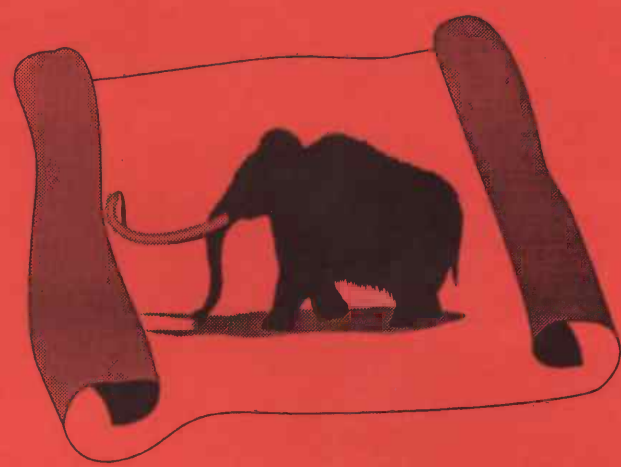
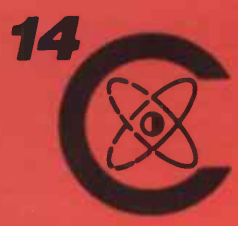
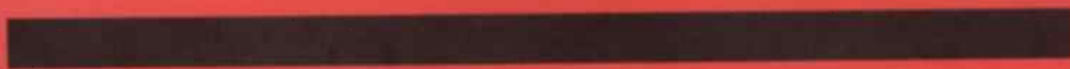


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Holocene Mammoths, Dead Sea Scrolls

Editor
AUSTIN LONG

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A. J. T. JULL

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Assistant Editor
DAVID R. SEWELL



Department of Geosciences
University of Arizona
East Ft. Lowell Road
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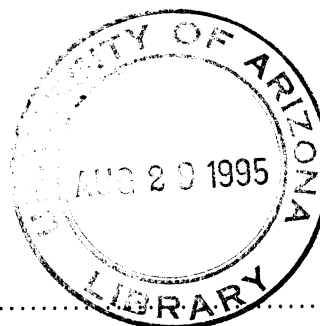
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List of laboratories. Our comprehensive list of laboratories is available upon request. We are expanding the list to include additional laboratories and scientific agencies with whom we have established contacts. The editors welcome information on these or other scientific organizations. We ask all laboratory directors to provide their laboratory code designation, as well as current telephone and fax numbers, and e-mail addresses. Changes in names or addresses, additions or deletions should be reported to the Managing Editor. Conventional and AMS laboratories are now arranged in alphabetical order by country and we include laboratories listed by code designation.



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FROM THE EDITOR

If your university, institution or laboratory does *not* subscribe to *RADIOCARBON*, this is an excellent issue to show to your local librarian as an argument for their subscribing. The regular-issue articles on Wrangel Island mammoths and new direct dates on Dead Sea Scrolls are topical and have been the focus of recent news releases. Moreover, this issue contains some of the papers presented at the PAGES Chronologies Workshop at the 15th International Radiocarbon Conference in Glasgow. These papers focus on the important interface between calibrated radiocarbon dates and other radiometric methods, and on the transition between glacial and postglacial times. Enjoy.

Good-bye to Pee Dee Belemnite and Standard Mean Ocean Water

The time has come for *RADIOCARBON* to deal with the problem of standardized reporting of stable isotope data: specifically, $\delta^{13}\text{C}$, $\delta^{18}\text{O}$ and δD values. For all new manuscripts we will ask authors to discontinue the use of PDB and SMOW, and to report carbon, oxygen and water stable isotope values with respect to VPDB and VSMOW, as recommended by the Commission on Atomic Weights and Isotopic Abundances of the International Union of Pure and Applied Chemistry (Coplen 1994). $\delta^{13}\text{C}$ in organic material and carbonates will be reported with respect to VPDB. The preferred reference for $\delta^{18}\text{O}$ in marine carbonates is VPDB. Authors may also prefer to report freshwater carbonate oxygen isotopes with respect to VPDB. δD and $\delta^{18}\text{O}$ in water will be reported with respect to VSMOW. Thus *RADIOCARBON* will join other journals in adopting this standard nomenclature. For laboratories that have already standardized their reporting of $\delta^{13}\text{C}_{\text{PDB}}$, $\delta^{18}\text{O}_{\text{PDB}}$, $\delta\text{D}_{\text{SMOW}}$ and $\delta^{18}\text{O}_{\text{SMOW}}$ with respect to NBS-19 and VSMOW, this may not require recalculation of data. Accepted relative values (Coplen 1994) are:

$$^{13}\text{C}/^{12}\text{C} \text{ of NBS-19} = 1.00195 \times ^{13}\text{C}/^{12}\text{C} \text{ of VPDB}$$

$$^{18}\text{O}/^{16}\text{O} \text{ of NBS-19 (carbonate)} = (1/1.0022) \times ^{18}\text{O}/^{16}\text{O} \text{ of VPDB} .$$

For reporting $\delta^{18}\text{O}$ and δD of water, laboratories must make sure that they have calibrated with VSMOW (the "zero" reference point) and SLAP ($\delta\text{D}_{\text{VSMOW}} = -428\text{‰}$; $\delta^{18}\text{O}_{\text{SMOW}} = -55.5\text{‰}$), which are available from the International Atomic Energy Agency (IAEA) in Vienna (International Atomic Energy Agency, Isotope Hydrology Section, Wagramerstrasse 5, P.O. Box 100, A-1400, Vienna, Austria.) In these cases also, laboratories are likely already tied in to these references. The addition of the "V" to the reference nomenclature will assure those using the data that they are tied to a common reference material.

We expect that this new nomenclature will help relieve the uncertainties created by the use of the terms PDB and SMOW. PDB was exhausted many years ago, and SMOW has three independent definitions (Coplen 1994). We regret any inconvenience to our authors, but we believe that this elimination of ambiguity and improved comparability of data makes the change worthwhile.

NIST Standard Nomenclature

In the same vein, *RADIOCARBON* will also now require authors to use the oxalic acid standard nomenclature defined and recommended by Currie *et al.* (1989), Currie (personal communication, 1995) and Klinedinst *et al.* (1994). This is, for NIST Oxalic Acid Standard Reference Materials:

Official NIST Designation	Traditional Designation	New Designation
SRM 4990 B	Oxalic Acid I	HOxI
SRM 4990 C	Oxalic Acid II	HOxII

Austin Long

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ROBERT STUCKENRATH, 1927–1994

Acerbic, abusive, brilliant, brusque, eclectic, exasperating, insightful, irritating, patient, patronizing, gentleman—these are but a few of the terms I have heard applied to Bob Stuckenrath over the 24-year period that I knew him. Their contradictory diversity serves to capture, if not the essence, at least some of his many different facets as a man and scholar.

Bob came from a highly educated background. In another time, some would have said he had “learning” in the genes. His father was a lawyer and many others in his immediate and extended family had extensive college training. In such a milieu, and with his great store of native intelligence, it is not surprising that, from an early age, his own interests were broad and remarkably varied. Humorously, but ruefully, he would note that, except for Latin (which he learned as a law student), the linguistic talents that characterized his family did not extend to him. Indeed, in self-deprecating fashion, he described himself as “p***-poor” in languages. Accurate assessment or not, his career as a student followed many paths—which superficially may have appeared unrelated—as did his later professional life. Yet a pattern can be discerned beneath the seemingly disparate threads. He began his higher education at Swarthmore as a mechanical engineer and, in fact, worked briefly as a junior engineer for the Baldwin-Lima-Hamilton corporation. Shortly thereafter, he transferred to Allegheny College and changed his major to history. His long-abiding interests in the pre-Revolutionary War United States and in contemporary European history took formal shape during this period, and he graduated with a degree in history in 1952.

At this juncture, his education took another turn: perhaps influenced by his barrister father, Bob entered the University of Pennsylvania Law School, where he ultimately earned an LL.B. (later converted to a J.D.). Within the pursuit of his law degree, he not surprisingly enjoyed the historical ele-

ments in title searching—"digging in the past" in a different way, he later called it—and he also became even quicker on his feet, expressing himself nimbly, logically, directly, and not uncommonly, devastatingly. Much of this "spin-off" from his legal education would serve him well and, indeed, distinguish him in his later years.

While at Penn Law School, he met his wife-to-be, Barbara, and through her was introduced to Elizabeth Ralph. Though he briefly served as a law clerk between 1955 and 1957, his legal career would end prematurely when his educational trajectory made its final shift. Uniting his historical interests with his not inconsiderable engineering talents, Bob served as a research assistant, research associate, and then research specialist in the Penn Radiocarbon Lab while completing his M.S. in Archaeology (1963) and his Ph.D. in Anthropology (1969). During his years at Penn, he had the opportunity to study within a milieu of dynamic scholars who included Robert Dyson, Carlton Coon, Loren Eiseley, Henry Fisher, J. Louis Giddings, James Pritchard, Linton Satterthwaite and both Kidders. Indeed, it was A. V. "Alfie" Kidder II who urged Bob to go to Debert, Nova Scotia, in 1963 with Douglas Byers. Widely regarded as a highly significant archaeological site from the time of its discovery, the careful excavation and dating program carried out for this fluted-point locality resulted in the recovery of a full suite of Paleo-Indian lithic artifacts firmly dated to 10,600 BP. Bob would return to Debert in 1964 and 1966, and also participate in a wide variety of other field projects, not just in archaeology but also in what now is usually called geoarchaeology. Following his M.A. project (on fluted-point chronologies in North America), Bob embarked on a study of sea-level changes in Chesapeake Bay. He would subsequently further develop and sustain an interest not only in the actors of antiquity but also in the stage upon which they performed. Indeed, from a very early point in his archaeological career he appreciated and championed a multidisciplinary approach to the systematic study of the human and natural record and their points of intersection. To his pride and credit, he *never* set aside or compromised that multi-field perspective.

During all of his formal academic training and fieldwork at Penn, he progressed in the Radiocarbon Lab from an "apprentice" scrubbing test tubes to a veritable master of his then still somewhat arcane craft. Indeed, his knowledge of all aspects of the dating process was encyclopedic—though he never deluded himself into believing that by being scientific, he actually practiced "science". For him, radiocarbon dating was no more and no less than a tool. It was not without reason that he referred to himself as Merlin, and when he finally had his own lab dubbed it Merlin's Lair.

At some point in 1967 or 1968, Betty Meggers and the late Clifford Evans heard Bob present a talk in Pullman, Washington, which ultimately led to Bob's appointment as Director of the Smithsonian Institution's Radiocarbon Laboratory, a position he would hold until 1986. Initially based in the basement of the "castle", then moved to the Radiation Biology Laboratory in Rockville, Maryland in 1970, the Smithsonian Radiocarbon Lab would become one of the leading archaeometric facilities in the world.

It was shortly after the establishment of the Rockville version of Merlin's den that I first met Bob. At that time, I was pursuing a postdoctoral project at the Smithsonian Institution that included, among other things, an attempt to unravel the evolution of basketry manufacture in prehistoric northern Mexico as reflected in Walt Taylor's Coahuila collections. With some trepidation, I approached Cliff Evans and Betty Meggers about having some of Taylor's perishables dated and was subsequently introduced to Bob. Many months and many odoriferous sandals, baskets, bags and even human coprolites later, I had a marvelous series of over 30 dates and, more importantly, had begun to appreciate Bob's incredible patience, attention to detail, and rigid insistence on the highest quality laboratory methods, from pretreatment through combustion, counting and ultimately the

generation of a “date”. I also quickly appreciated his equally rigorous insistence on the importance of well-defined stratigraphy, firm context and clear and unequivocal association relative to samples sent to him, and his utter contempt for archaeologists or other field workers whose data collection or documentation processes were not up to his standards. On many occasions, when asked about this putatively old site or that apparently anomalous date, or this archaeologist or that geologist, Bob would furrow his brow and bark “garbage in, garbage out!” Since the reliability of any date is only as good as its context, and its context only as good as its excavator, it is small wonder that when Bob weighed excavators on the scales of balance he found many wanting.

He frequently developed lifelong working relationships with those whose work he trusted, and it is not surprising that his circle of closest colleagues included a cadre of scholars well known for careful and meticulous field and lab practices. Interestingly, this group—which included geologists like Hal Borns, paleogeographers like Karl Butzer and archaeologists and geoarchaeologists like Bob Ackerman and Vance Haynes—all had a uniformly high opinion of Bob’s work even when they disagreed with his professional views on certain subjects.

Nowhere was this more personally apparent to me than when he undertook in 1973–1974 the assay of the first of some 50 radiocarbon samples from Meadowcroft Rockshelter. In what was to become a nearly two-decade-long project, Bob produced a 12,000–16,000-yr occupational chronology, the very earliest phases of which have been hotly debated almost *ab initio*. With Meadowcroft critics for whom he had little respect—or, sometimes, more accurately, little patience—he was indeed acerbic, abusive, brusque and caustic. But for others whose disagreements were well reasoned and carefully advanced, he had the greatest personal respect, whatever he might say (or, rarely, publish) in a moment of vexation.

My long experience with Bob during the Meadowcroft years led me (then Chair of Pitt’s Anthropology Department and Director of the Cultural Resource Management Program) to offer Bob a “home” for his lab and a new position when the Smithsonian closed his facility for financial reasons in 1986. Once ensconced and fully operational in Pittsburgh, Bob continued to take on a myriad of archaeological, geological, geoarchaeological and paleoenvironmental dating projects from sites and localities all over the world. He would continue this pattern until and even after his lab was again closed for budgetary reasons in 1992. Indeed, shortly before his death, Bob had acted as a consultant to Exxon on possible damage to archaeological sites caused by the *Valdez* oil spill and, characteristically, brought his usual rigor to bear in that unfortunate situation.

Recounting the dozens of projects with which Bob was engaged throughout his long and productive career reflects the great breadth and depth of his scholarly interests but does not, in my mind, fully underscore his unique qualities as a nonpareil scientist or a man. Unfortunately, I can only touch on some of these.

His insistence on multiple dates (preferably in long stratigraphic series) was evident from his Debert dissertation days and would characterize his dating philosophy to the end. Whether it was Meadowcroft, the Lower Vaal River in South Africa, Baffin Island, coastal Maine or anywhere else, “more” was *always* better—if the excavator could be trusted. His willingness to experiment with new sample materials (from whale blubber to ostrich eggshells) or to refine methods appropriate to more “traditional” dating media like charcoal, paleosols or coprolites is also well documented in his career. In the ostrich egg “case study”, another aspect of his character or personality was revealed when he and Karl Butzer collected a modern eggshell that Bob hoped to compare to archaeological specimens he was about to date. To preserve the shell for transport, they decided to hard-boil it, and while awaiting the results, became sufficiently engrossed in other matters to forget that they had not

punctured the specimen before immersion. Blown back to reality by the exploding egg, they had plenty of time to consider their error while laughingly cleaning up the debris.

This incident reflects another side of Bob and one that many who did not know him well seldom saw—his great humor and wit. Even when he was caustic, he was *elegantly* caustic; and though he may have used humor as a weapon, he did it with dash, flair and inimitable style. Though he was not a misanthrope, I am sure Bob would have gotten on quite well with Jonathan Swift.

Characteristically, at the time of his death, Bob had a number of ongoing projects and obviously hoped to continue both his research and his lifelong “affair” with the outdoors, especially through the venue of fly-fishing. No one—least of all me—can summarize a life, a career and a person like Bob Stuckenrath. He was one of those very rare persons who, once met, was not easily forgotten or readily dismissed. Like his chosen namesake, the Merlin of Arthurian romance, some took him well, others took him badly, but no one took him lightly.

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J. M. Adovasio

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Radiocarbon

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RADIOCARBON DATING EVIDENCE FOR MAMMOTHS ON WRANGEL ISLAND, ARCTIC OCEAN, UNTIL 2000 BC¹

S. L. VARTANYAN

Wrangel Island State Reserve, 686870 Ushakovskoye, Magadan Region, Russia

KH. A. ARSLANOV, T. V. TERTYCHNAYA and S. B. CHERNOV

Geographical Research Institute, St. Petersburg State University, Sredniy Prospect 41
199004 St. Petersburg, Russia

ABSTRACT. Radiocarbon dating results of mammoth tusks, teeth and bones collected on Wrangel Island between 1989 and 1991 reveal a unique mammoth refugium during the Holocene. We used an improved chemical procedure to obtain and purify collagen from bone. Benzene synthesized from the samples was measured using a liquid scintillation counter. The validity of our data has been confirmed by the results of our measurements on two international control sample series (IAEA and TIRI) and by parallel measurements of Wrangel Island mammoth remains at other laboratories.

INTRODUCTION

The systematic ¹⁴C dating of woolly mammoth (*Mammuthus primigenius*) remains, carried out in our laboratory for many years, should help determine the cause or causes of the mammoths' extinction. Among the remains found well preserved in permafrost are those of the mammoths known as Shandrín, Terekhtyakh, Magadan, Khatanga and Yuribei (Arslanov *et al.* 1980, 1982). Many scientists assume that the most probable cause of extinction was an abrupt change in climatic conditions during the Pleistocene/Holocene transition, a period corresponding to the ¹⁴C dates of the youngest mammoths of Siberia. Others invoke an anthropogenic agency, or a combination of cultural and paleoclimatic forces. An exhaustive explanation of the cause is not yet possible. In the Late Pleistocene, the range of mammoths shifted northward. In the Crimea and the Caucasus, mammoths became extinct >20–30 ka ago; on the Russian plain they were still present *ca.* 13 ka ago (Arslanov *et al.* 1972; Lavrov and Sulezhytsky 1992). Based on ¹⁴C ages, the latest mammoth remains found in western Europe (northern France, Switzerland and Great Britain) also date to 12–13 ka ago, when their remains become relatively uncommon (see Stuart 1991).

The last refugia of the mammoth were thought to be the Siberian Arctic and the Arctic islands, including Gydan and Taimyr Peninsulas and the Severnaya Zemlya Archipelago. The youngest ¹⁴C date our laboratory obtained for the Yuribei mammoth (Gydan peninsula, Siberia) was 10,000 ± 70 BP, based on stomach contents (plant debris). Similar results were later obtained by L. Sulerzhytsky (Geological Institute of the Russian Academy of Sciences, Moscow) for tusk and tooth remains of mammoths of the Taimyr Peninsula, Siberia (Lavrov and Sulerzhytsky 1992). ¹⁴C data thus indicated that mammoths became extinct, even in the refugia of the continental Siberian Arctic, *ca.* 9.7–10 ka ago. However, in 1990, our first five dates for mammoth remains from Wrangel Island were all of Holocene age, ranging from 7390–4740 BP (Vartanyan *et al.* 1992). This surprisingly young estimate prompted us to continue investigations at that location.

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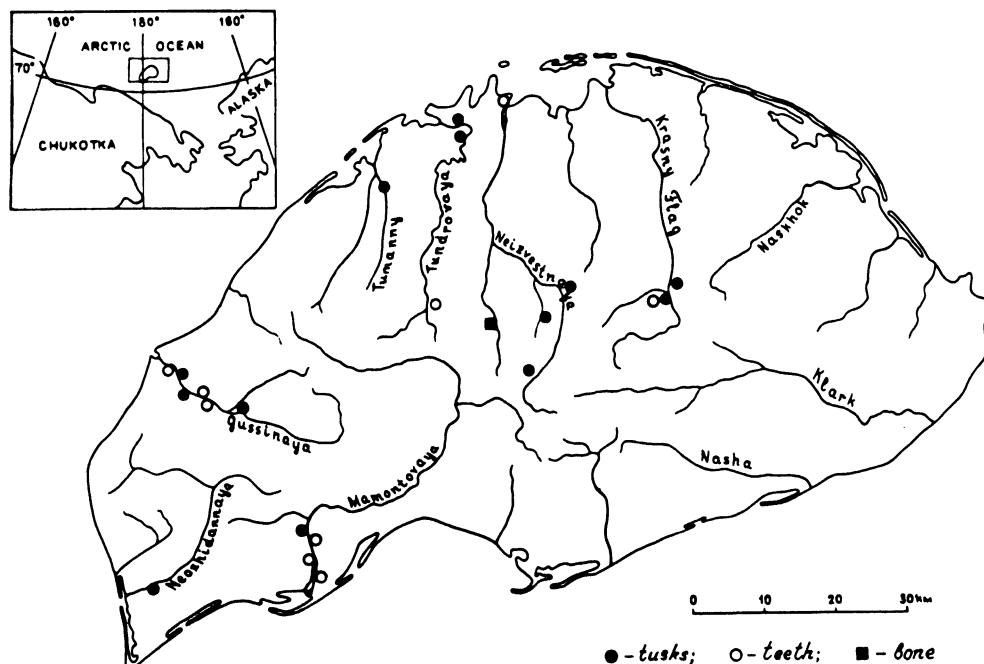


Fig. 1. Location map of Wrangel Island, showing sites of mammoth samples

GEOGRAPHY OF THE SITE

Wrangel Island is located on the border of the East-Siberian and Chukchi Seas, between 70° and 72°N, and 177°E and 176°W (Fig. 1). *Ca.* 8000 km², the island is separated from the continent by the Long Strait, with a minimum width of 140 km, and a depth of ≥ 45 m. The eastern, western, and central portions of the island have low mountain ranges (up to 1100 m asl) and bisected plateaus; the northern (“Akademia Tundra”) and southern portions are plains.

The Quaternary sediments of Wrangel Island are not very thick; they are primarily aleurite, sand, shingle and peat of Late Pleistocene and Holocene age. The coastal-marine sediments (in the Akademia Tundra) and the alluvium of the high terraces of large rivers, as well as the spare, heavily ice-bearing sand-shingle terraces of the lower hilly terrain, all seem to be of Pleistocene age. Numerous solifluction and other slope sediments of the mountainous part of the island, river-valley alluvium (floodplain and the first terraces of large rivers), and peat and lacustrine thermokarst sediments are widespread on the plains and intermontane depressions, and are of Holocene age.

Bone-rich horizons such as the “Edoma” strata in Siberia have not been found on Wrangel Island. All the bone specimens were collected from riverbed and slope sediments. Only well-preserved tusks (except for one split tusk), teeth and bones were collected. We selected teeth for dating from the collection of S. Vartanyan, made in 1991, which was previously studied by V. E. Garutt of the Zoological Institute of Russian Academy of Sciences.

METHODS

Although all samples were well preserved in frozen ground, we used stronger pretreatment than conventional to remove impurities. We extracted and purified bone collagen using a procedure devel-

oped in our laboratory to remove both easily soluble and relatively insoluble organic contaminants (Arslanov and Svezhentsev 1993). We found that the well-known HCl-NaOH and Longin's procedures by themselves did not sufficiently purify collagen.

We obtained carbon from the purified collagen by means of pyrolysis, then lithium carbide according to the reaction $2C + 2Li \rightarrow Li_2C_2$. Benzene synthesis was carried out using a high-efficiency vanadium-alumina-silica catalyst ($V_2O_5 \cdot Al_2O_3 \cdot SiO_2$) that enabled us to produce benzene with up to 96% yield. Sample activity was measured with a coincidence scintillation counter using quartz spherical vials (8.46 ml capacity); background and modern standard count rates were 1.9 cpm and 56.8 cpm, respectively. Our techniques for pretreatment and measurement are described in detail elsewhere (Arslanov, Tertychnaya and Chernov 1993).

RESULTS AND DISCUSSION

We dated 23 samples of *Mammuthus primigenius* remains from Wrangel Island. The youngest and the oldest presently known dates of mammoth remains from the Siberian Arctic and Severnaya Zemlya (Makeev, Arslanov and Garutt 1979; Arslanov *et al.* 1982; Lavrov and Sulerzhitsky 1992) are given in Table 1. Table 2 shows our Wrangel Island dates; 20 out of the 23 samples proved to be of Holocene age, falling within the range 3730 ± 40 to 7390 ± 30 BP. Three dates on teeth from Wrangel Island were Pleistocene in age: LU-2823, -2792 and -2807 at *ca.* 12, 13 and 20 ka, respectively (Table 2), when Wrangel Island was part of Beringia.

TABLE 1. Oldest and Youngest ¹⁴C Dates on Mammoths of Arctic Siberia (excluding Wrangel Island)

Material and location	Lab no.	¹⁴ C age (yr BP)
<i>Continent</i>		
Fragment of mammoth skin, Khatanga River, Taimyr Peninsula	LU-1057	≥53,170
Plant debris from mammoth stomach, Yuribei River, Gydan Peninsula	LU-1153	10,000 ± 70
Mammoth tusk, Nizhnaya Taimyra River	GIN-1823	9670 ± 60
Mammoth tooth, Nizhnaya Taimyra River	GIN-1495	9860 ± 50
<i>Arctic Islands</i>		
Mammoth tusk, October Revolution Island, Severnaya Zemlya Archipelago	LU-610	11,500 ± 60

Geomorphological reconstruction indicates that Wrangel Island formed a part of Beringia (an area that included Chukotka, Alaska, and the huge expanse of the surrounding shelf) during the Late Pleistocene, when the global sea level was *ca.* 100 m below the present level. At the end of the Pleistocene or beginning of the Holocene, Wrangel Island separated from the continent (Hopkins 1975), becoming a refugium for the mammoth population. Our data show that this population survived as long as 6000 yr after all mammoths on the continent were extinct. Morphological studies of mammoth teeth demonstrate that a previously unknown dwarf species of mammoth evolved on Wrangel Island (Vartanyan *et al.* 1993).

Three Wrangel Island mammoth teeth were found to be of Late Pleistocene age, with one sample (LU-2807) (Table 2) deriving from the period of the Late Glacial maximum (~20,000 BP). We obtained similar dates for a mammoth tusk collected on Severnaya Zemlya Archipelago (19,270 ±

TABLE 2. ^{14}C Dates of Mammoth Remains from Wrangel Island

Lab no.	Material, provenience	^{14}C age (yr BP)	Calibrated age (cal BC)*
<i>Tusks and Bone</i>			
LU-2741	Tusk (8 cm diameter (d)) from the bed of the lower Neozhydannaya River	3730 \pm 40	2192–2038
LU-2756	Tusk (11 cm d) from the bed of the lower Mamontovaya River	4400 \pm 40	3082–2924
LU-2768	Tusk (9 cm d) from the bed of Tumanny Creek	4410 \pm 50	3090–2924
LU-2556	Tibia bone (0.1–0.2 m) from the floodplain of the upper Lemmingovaya River	4740 \pm 40	3626–3382
LU-2740	Tusk (6 cm d) from the bed of the lower Tundrovaya River	4900 \pm 40	3706–3646
LU-2745	Tusk (9.5 cm d) from the bed of the lower Goosinaya River	5200 \pm 30	4036–3972
LU-2744	Tusk from the bed of the middle Goosinaya River	5250 \pm 40	4216–3990
LU-2742	Tusk from the bed of the lower Goosinaya River	5310 \pm 90	4232–4000
LU-2535	Tusk from left bank of the Red Flag River valley, 2 km upstream from the mouth of the Otrozhnaya River	5480 \pm 50	4440–4252
LU-2558	Tusk from diluvium-solifluction sediments on the western slope of Mount Kit, left side of Neizvestnaya River valley	6610 \pm 50	5567–5450
LU-2736	Tusk from creekbed on the left side of the upper Neizvestnaya River valley	6760 \pm 50	5666–5585
LU-2746	Tusk (7 cm d) from the bed of the lower Tundrovaya River	7040 \pm 60	5954–5816
LU-2559	Tusk from the bed of middle Vetvisty Creek	7360 \pm 50	6214–6062
LU-2444	Tusk from the right side of Red Flag River valley, area of the mouth of the Otrozhnaya River	7390 \pm 30	6216–6176
<i>Teeth</i>			
LU-2798	Last upper molar from the bed of the lower Mamontovaya River (N-MAM-6)	4010 \pm 50	2574–2464
LU-2808	Tooth fragment from the bed of the lower Mamontovaya River (N-MAM-2)	4040 \pm 30	2582–2492
LU-2794	Last lower molar from the bed of the lower Mamontovaya River (N-MAM-5)	5110 \pm 40	3966–3812
LU-2799	Last lower molar from the bed of the lower Goosinaya River (N-GUS-9)	6260 \pm 50	5262–5088
LU-2810	Tooth fragment from the bed of the lower Goosinaya River (N-GUS-9)	6890 \pm 50	5766–5672
LU-2809	Last lower molar from the bed of the upper Tundrovaya River	7250 \pm 60	6158–5988
LU-2823	Last lower molar from the bed of the lower Goosinaya River (N-GUS-8)	12,010 \pm 110	12,200–11,925
LU-2792	Last lower molar from the bed of the middle Red Flag River	12,980 \pm 80	13,580–13,325
LU-2807	Last lower molar found on a beach, 1 km from the mouth of the Neizvestnaya River	20,000 \pm 110	--

*Calibrated age calculated using CAL15 (van der Plicht 1993)

130 BP) (Makeev, Arslanov and Garutt 1979), and for a front leg-bone found at the mouth of the Lena River, on the Bykovsky Peninsula (21,630 ± 240 BP, LU-1328, Tomirdiaro *et al.* 1984). All of these data support the thesis that during the last glacial maximum, conditions in the East-Siberian Arctic and on the northern islands were adequate for habitation by mammoths.

ON THE RELIABILITY OF MAMMOTH DATES

How reliable are these Wrangel Island dates? The question is legitimate: bone is difficult to date, due to its high potential for absorbing external humid acids from groundwater. However, all our mammoth samples were well preserved and were collected from frozen ground. We used a reliable procedure for the chemical treatment of the bone (Arslanov and Svezhentsev 1993), which permitted us to obtain a collagen purified of organic contaminants, whatever their solubility. In addition, the viability of our methods and measurements is supported by our dating of a series of samples for inter-laboratory quality control supplied by the International Agency for Atomic Energy (IAEA) and by Glasgow University, the Third International Radiocarbon Intercomparison (TIRI) (Scott *et al.* 1992). All of our dates (Table 3) were in accordance with the control figures, within the limit of double-measurement uncertainties.

TABLE 3. Measurements of ¹⁴C Activity and Age Determination of TIRI-Control Samples

TIRI code	Material	St. Petersburg measurements	TIRI mean values
A	Grain	117.02 ± 0.57%	116.12%
B	Wood	4580 ± 40 BP	4486 BP
C	Cellulose	129.27 ± 0.58%	129.81%
D	Peat	3730 ± 40 BP	3799 BP
E	Humic acid	10,980 ± 70 BP	11,066 BP
F	Iceland double spar	≥50,000 BP	≥46,076 BP
G	Wood	≥51,560 BP	≥42,962 BP
H	Peat	11,130 ± 40 BP	11,115 ± 116 BP
I	Travertine	11,170 ± 80 BP	11,034 ± 127 BP
J	Wood	1590 ± 40 BP	1593 ± 50 BP
K	Carbonate	18,400 ± 140 BP	18,166 ± 238 BP
L	Whalebone	12,580 ± 60 BP	12,605 ± 127 BP

Later, 2 teeth and 1 tusk from Wrangel Island were dated at The University of Arizona Radiocarbon Laboratory (AA) (Long, Sher and Vartanyan 1994) and by L. Sulerzhitsky in the laboratory of the Geological Institute of the Russian Academy of Sciences (GIN). The data shown in Table 4 are in excellent agreement and, along with data from the Third International Radiocarbon Intercomparison (TIRI), these measurements confirm the reliability of our dates.

TABLE 4. Age determinations of Wrangel Island Mammoth Remains: Results of Three Laboratories

Sample	Age and sample number	
GUS-9, tooth	6260 ± 50, LU-2799	6360 ± 60, AA-11529
PIK-1, tooth	7250 ± 60, LU-2809	7295 ± 95, AA-11530
20-M, tusk	6760 ± 50, LU-2736	6750 ± 30, GIN-6990

CONCLUSION

During the last glacial maximum (*ca.* 20 ka ago), environmental conditions on Wrangel Island proved capable of sustaining habitation by mammoths. Our data show that woolly mammoths persisted on Wrangel Island in the mid-Holocene, from 7390–3730 yr ago. ^{14}C dating has shown that mammoths inhabited Wrangel Island for as long as 6000 yr after the estimated extinction of *Mammuthus primigenius* on the Siberian continent.

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MAMMOTH EXTINCTION: TWO CONTINENTS AND WRANGEL ISLAND

PAUL S. MARTIN

Desert Laboratory, The University of Arizona, Tucson, Arizona 85721 USA

and

ANTHONY J. STUART

Norfolk Museums, Norwich NR1 3JU England

A harvest of 300 radiocarbon dates on extinct elephants (Proboscidea) from the northern parts of the New and Old Worlds has revealed a striking difference. While catastrophic in North America, elephant extinction was gradual in Eurasia (Stuart 1991), where straight-tusked elephants (*Palaeoloxodon antiquus*) vanished 50 millennia or more before woolly mammoths (*Mammuthus primigenius*). The range of the woolly mammoths started shrinking before 20 ka ago (Vartanyan *et al.* 1995). By 12 ka BP, the beasts were very scarce or absent in western Europe. Until the dating of Wrangel Island tusks and teeth (Vartanyan, Garrutt and Sher 1993), mammoths appeared to make their last stand on the Arctic coast of Siberia *ca.* 10 ka BP. The Wrangel Island find of dwarf mammoths by Sergiy Vartanyan, V. E. Garrut and Andrei Sher (1993) stretched the extinction chronology of mammoths another 6 ka, into the time of the pharaohs.

Not since the early years of ^{14}C dating, when laboratory protocols for sample selection and pretreatment were not standardized or well understood by consumers of dates (see, *e.g.*, Martin 1958 and Hester 1960), has anyone seriously advanced the thought that mammoths or mastodons survived into the mid-Holocene. Those North American Holocene dates of yore were not replicated and could not be supported stratigraphically and geochemically. They moulder in the graveyard of unverified measurements. The 20 new dates on teeth and tusk from Wrangel Island range from 6148 to 2192 BC (Vartanyan *et al.* 1995). They are accompanied by standardized test measurements of the St. Petersburg Laboratory as well as independent replications (Long, Sher and Vartanyan 1994). Frozen ground at the latitude of Wrangel (70.5°N) is a very favorable environment for preservation of skeletal collagen, thus ideal for dating. The new data set appears unassailable. What does it mean?

For one thing, it appears discordant with the idea that a Younger Dryas (YD) cold snap or climatic upset roughly 11–10 ka BP forced megafaunal extinction (Berger 1991). While the YD falls hard on the heels of extinction of North American mammoths (*Mammuthus columbi*), horses, camels, ground sloths, extinct mountain goats, sabertooths and others, as well as Old World giant deer (*Megaloceros giganteus*) in Ireland, all other Old World extinctions precede or postdate the YD (Fig.1; Stuart 1991).

Ironically, the YD climatic signal is strongest in Western Europe. Even before ^{14}C dating, it was the basis for correlating pollen diagrams. On oceanic islands around the globe, most doomed genera of birds, mammals and reptiles would disappear, like the Wrangel dwarf mammoths, long afterward, in the Holocene (Steadman 1995).

Admittedly, the fossil record on many of the oceanic islands has yet to be traced into YD or earlier time. Late Glacial or earlier extinction pulses remain a possibility. However, when faunas of such an age are known, there is no extinction pulse at any time within the ^{14}C range, prior to the late Holocene (Steadman 1995). Neither the YD, nor the warm–cold oscillations in Greenland core ices known as Dansgaard-Oeschger events that coincide with ice transport of lithic material into the North Atlantic (Bond and Lotti 1995), nor any other independently established climatic perturbation

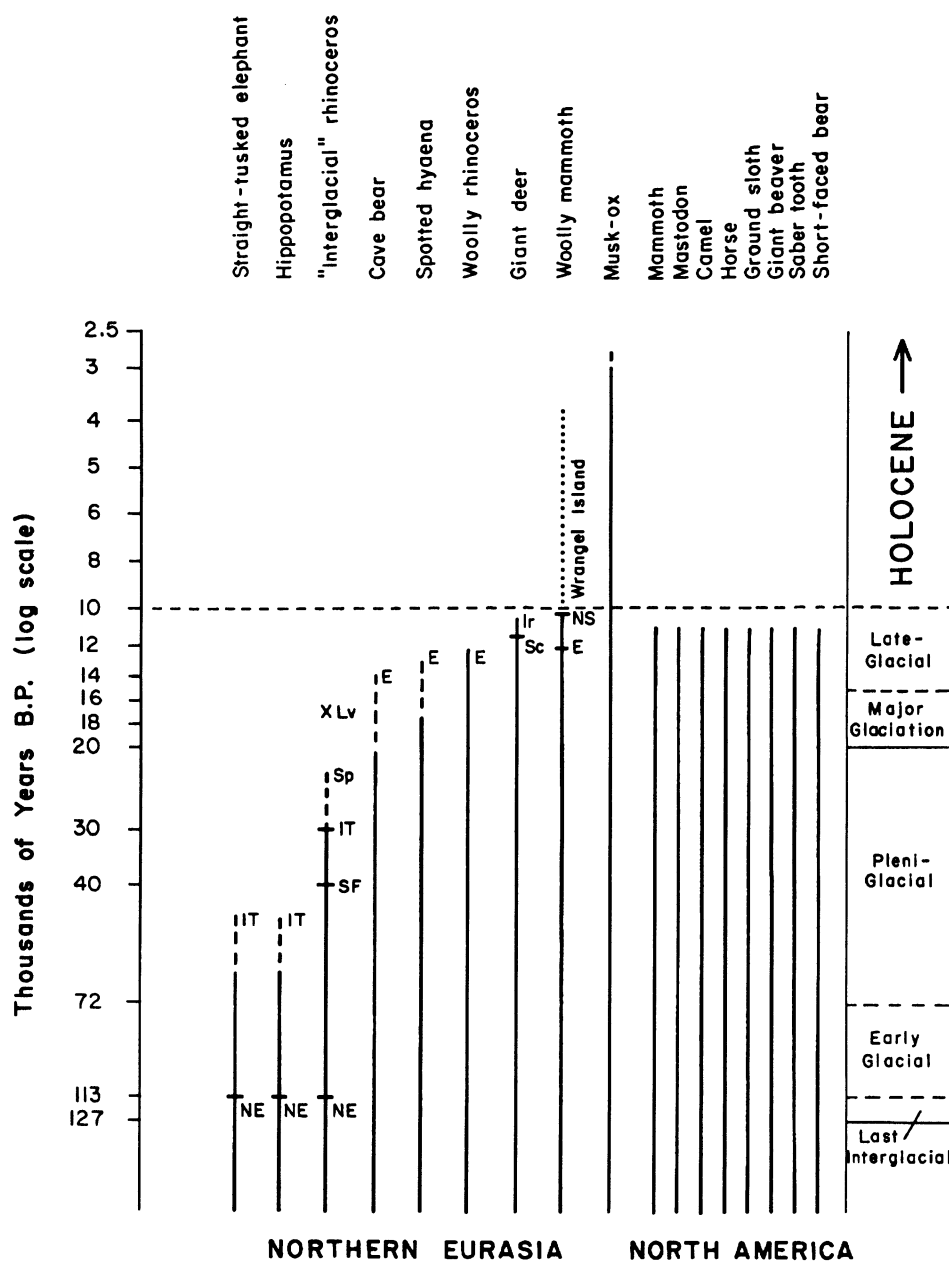


Fig. 1. Large mammals that vanished from northern Eurasia are compared with some of those that vanished from North America. Loss of the former was sequential over ~50 ka; the latter loss was catastrophic at 11 ka BP. NE = northern and central Europe; IT = Italy; SF = southern France; Sp = Spain; Lv = Levant; E = England; Sc = Scandinavia; Ir = Ireland; NS = north-central Siberia. Straight-tusked elephant = *Palaeoloxodon antiquus*; hippopotamus = *Hippopotamus amphibius*; "interglacial" rhinoceros = *Dicerorhinus* spp.; cave bear = *Ursus spelaeus*; spotted hyaena = *Crocota crocuta*; woolly rhinoceros = *Coelodonta antiquitatis*; giant deer = *Megaloceros giganteus*; woolly mammoth = *Mammuthus primigenius*; musk-ox = *Ovibus moschatus*. North American extinctions: mammoth = *Mammuthus* spp.; mastodon = *Mammuthus americanum*; camel = *Camelops hesternus*; horse = *Equus* spp.; ground sloth = *Nothrotheriops shastensis*; saber-tooth = *Smilodon fatalis*; and short-faced bear = *Arctodus simus*.

dated by ^{14}C coincides with the extinction pattern outlined above (Fig.1). Although further tests are needed, the extinctions that do not coincide with the YD are as noteworthy as cases that do (or might) fall within its time range.

In western North America, the source of the best ^{14}C dates on megafaunal extinctions in the New World (Stuart 1990), the event coincides with Clovis (Llano culture) origins (Haynes 1991, 1993). The number of sites in which mammoth bones were found with artifacts is small, and no bones of horse, camel and ground sloths (to cite a few examples) have been found in undeniable association with stone or bone tools. If the Paleolithic invaders of the New World forced the extinctions, as some propose, it was remarkably rapid, a “blitzkrieg” (Mosimann and Martin 1975), or a “mammoth undertaking” (Diamond 1992). Obviously, the catastrophic model will not work if all New World generic extinctions were not synchronous. Some believe that they were not (Grayson 1991). Critics, in turn, bear the burden of demonstrating how a climatic pulse could force the disparate extinction sequence between the New and the Old Worlds (Fig.1).

Another climatic model for mammoth extinction deserves mention. Beringian paleontologists, Russian, Canadian and Alaskan, have long viewed the coming of the Holocene in the far north, with its string bogs, sphagnum, ericads and other herbivore-resistant plants, and with deep snows in winter, as uninhabitable for mammoths and the extinct steppe fauna of Glacial times (Guthrie 1990 and references therein). In the case of Wrangel Island, even before the discovery of Holocene dwarf mammoths, Russian botanists had reported an unusually rich and palatable assemblage of steppe tundra plants, including 15 species of grasses, 4 of wormwood (*Artemisia*), 10 legumes, many Rosaceae and very few Ericaceae (Yurtsev 1982). Wrangel sounds like an ideal place to survive the loss of the mammoth steppe.

Nevertheless, Guthrie’s argument leaves unanswered questions of the suitability of other relict patches of grasses and palatable steppe plants elsewhere in Eurasia and North America. Were there no other Wrangel-size refugia for woolly mammoths? What of vast expanses of steppe with dozens of genera of grasses, legumes and Rosaceae and many species of *Artemisia* and palatable Chenopodiaceae in the high dry plateaus and valleys of eastern Eurasia and western North America? Would not western North America offer suitable continental climates for various species of mammoth? Special-case environmental arguments that serve to explain regional extinctions, such as the Beringian mammoth steppe, may sound plausible, until one attempts to generalize more widely.

On one matter all “extinctionists” interested in the fate of mammoths can agree: ^{14}C dating has done more to clarify events than any other analysis. The method offers a unique opportunity to evaluate extinctions in many corners of the earth over the last 50 ka. Bones of the youngest extinct large mammals (including mammoths) in the frozen ground of Arctic and subarctic latitudes may prove to be of any age. It takes a serious effort to wade through dozens of dates, most of which are too old by many thousands of years to approach the last millennium when extinction likely occurred. Russian paleontologists and Russian ^{14}C laboratories have worked hard on this problem. They are to be congratulated. Their work confirms dramatically an extinction chronology of mammoths in the Old World that was much more gradual than the one in the New World (Fig. 1). In the New World and increasingly in the Old, the chronology is in step with the spread of prehistoric humans.

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RADIOCARBON DATING OF SCROLLS AND LINEN FRAGMENTS FROM THE JUDEAN DESERT

A. J. TIMOTHY JULL,¹ DOUGLAS J. DONAHUE,¹ MAGEN BROSHI² and EMANUEL TOV³

ABSTRACT. We report on new ¹⁴C measurements of samples of 18 texts (scrolls) and 2 linen fragments from Qumran Caves 1, 2, and 4 and from Nahal Hever, both in the Dead Sea region. The radiocarbon results are in good agreement with estimates of age based on paleography.

INTRODUCTION

Various parchment and papyrus manuscripts found in caves in the area of Qumran and at other sites in the Judean Desert are known generally as the Dead Sea Scrolls. The Qumran scrolls are generally considered to have been hidden by the Qumran Community, identified by most scholars as the Essenes. The documents are usually regarded to have been copied between the mid-third century BC and AD 68, when the Qumran settlement was destroyed by the Romans.

Bonani *et al.* (1991, 1992) dated 14 texts, 8 of which came from Qumran. We present here new radiocarbon dates of 18 texts, including 3 date-bearing texts (3 from Qumran Cave 1, 12 from Cave 4, and 3 from other sites in the Judean Desert). We consider the importance of the ¹⁴C dates in relation to other age estimates and we also report on ¹⁴C examinations of linen fragments from the Judean Desert.

METHODS

All except three of the scroll samples were taken on 21 and 22 March 1994 by museum staff in the presence of the authors at either the Rockefeller or Israel Museums (see Table 1). Three additional samples (DSS-50, -52 and -53) were taken later at the Shrine of the Book and sent to Tucson for analysis. All samples were taken from ragged edges of top or bottom margins of the scrolls. No samples were taken that would have caused any significant damage to the scrolls themselves. The sizes of the samples are listed in Table 1. Most of the documents from the Judean Desert had been suggested to us by colleagues who had special interests in ¹⁴C analysis of particular texts.

Some samples from date-bearing documents were added as control texts (DSS-25, -52 and -53), and the identity and ages of these materials were unknown to the Arizona AMS laboratory at the time of measurement. One control sample had been dated previously at ETH-Zürich in 1990–1991 (DSS-50). The identity of this sample was also unknown to the Arizona laboratory at the time of measurement. Photographic records were made of the exact locations of the pieces subjected to examination. In some cases, scrolls suggested as important for dating had insufficient material available in the margins or the margins were too beautiful to be harmed. These samples were not taken and they account for the missing numbers in the lists (*e.g.*, DSS-2).

Sample Types and Treatment

Small samples of 5–15 mg of material were removed. Samples were studied under a binocular microscope and were divided into three types:

¹NSF Arizona Accelerator Mass Spectrometer Facility, The University of Arizona, Tucson, Arizona 85721 USA

²Shrine of the Book, Israel Museum, Jerusalem 91710 Israel

³Dead Sea Scrolls Publication Project, Department of the Bible, Hebrew University, Jerusalem, Israel

TABLE 1. Description and Weights of Dead Sea Scroll Samples and Related Materials

Sample	Q no.	Description	Museum no.*	Weight (mg)
DSS-1	4Q266 (D ^a)	Damascus Document, a	706	23.35
DSS-3	1QpHab	Commentary on Habakkuk	Shrine†	31.00
DSS-4	1QS	Community Rule	Shrine	14.20
DSS-5	4Q258	Community Rule, d	140	14.90
DSS-7	4Q171 (pPs ^a)	Commentary on Psalms, a	600	11.70
DSS-8	4Q521	Messianic Apocalypse	330	4.90
DSS-9	4Q267 (D ^b)	Damascus Document, b	107	7.80
DSS-10	4Q249	Midrash Sepher Moshe	590	6.20
DSS-11	4Q317	Phases of the Moon	899	19.30
DSS-13	4Q208	Astronomical Enoch ^a	823	19.20
DSS-19	4Q22	PaleoExodus ^m	661	6.75
DSS-19P	4Q22 patch	Patch on 4Q22	661-P	10.70
DSS-22	4Q342	Letter	602	23.70
DSS-23	4Q344	Debt acknowledgment	602	9.56
DSS-24	4Q345	Sale of land	602	14.40
DSS-25	Pap Yadin 21	Papyrus, AD 130	Shrine	5.24
DSS-26	Cave 4	Linen	1041	30.70
DSS-27	Cave 2	Linen	749	21.70
DSS-50	1QIsa ^a	Book of Isaiah ^a	Shrine	56.50
DSS-52	Kefar Bebayou	Papyrus	Shrine	20.20
DSS-53	Pap Yadin 19	Papyrus, AD 128	Shrine	18.20

*All items derive from the Rockefeller Museum, Jerusalem, unless otherwise indicated.

†Items from the Shrine of the Book, Israel Museum

Type 1. Parchment samples that appeared to be relatively clean

Pieces of *ca.* 2–10 mg were pretreated using procedures based on those reported by Bonani *et al.* (1991, 1992) with some modifications. Samples were washed in ~1N HCl for 10 min, rinsed in distilled water, washed in 0.1% NaOH for up to 10 min, rinsed again in distilled water, and finally reacidified with HCl, and cleaned with distilled water. Samples were dried in a vacuum oven and were removed as soon as they were dry. We found that some partially gelatinized samples were very easily dissolved by NaOH solutions (as previously reported by Bonani *et al.* 1991 and 1992), and all samples were monitored during this process. Samples that started to dissolve in NaOH were removed from the solution as quickly as possible.

Type 2. Parchment samples with glue contamination

These samples were contaminated with perspex glue, as they had been stuck to rice paper as a backing material. They included DSS-1, -5, -11, -22, -23 and -24. DSS-4 was difficult to clean, as it had been attached to a silk backing material and also appeared to be impregnated with a glue-like material. Pieces of 2–8 mg with adhering glue were washed in acetone in an ultrasonic bath for 30 min. This procedure worked well for most samples, but in the case of two sample pieces (DSS-23 and -24), this process had to be repeated for three hours. The samples were then subjected to the same pretreatment routine as the first batch.

Type 3. Papyri

Papyrus samples (DSS-10, -25, -52, -53) were generally very clean. Pretreatment was carried out easily using the standard methods of Type 1, above.

Dried samples were combusted with CuO to make CO₂ using the standard techniques at Arizona (Donahue, Jull and Toolin 1990). For most samples, sufficient CO₂ sample was available, and a split of up to 0.2 ml was taken for stable-isotope analysis of the δ¹³C of the carbon. This parameter is important to make accurate corrections to the ¹⁴C age, which are all quoted as normalized to -25‰ (Stuiver and Polach 1977). The remaining CO₂ was converted to graphite using standard procedures. The graphite powder so produced was pressed into an accelerator target holder, and the target was then analyzed by accelerator mass spectrometry (AMS). We loaded 24 sample targets with 8 standard targets (consisting of 4 standard graphites made from NIST HOxI and 4 of HOxII). In most cases, several separate preparations of samples were performed. A general description of the AMS measurements is given by Donahue, Jull and Toolin (1990). ¹⁴C results were calculated using the procedures reported by Donahue, Linick and Jull (1990).

RESULTS AND DISCUSSION

Written Texts

Table 2 presents the results of the ¹⁴C and δ¹³C measurements. The results are reported as conventional ¹⁴C ages in years before present (AD 1950), with errors on one standard deviation (1 σ), and calibrated ages obtained using both 1-σ and 2-σ confidence intervals, using the calibration of Stuiver and Pearson (1986). For samples with insufficient material for both ¹⁴C and δ¹³C measurements, an average value of δ¹³C was estimated from results of other scrolls, and this value is given in parentheses. Also included in Table 2 are ages determined by paleographic analysis. In Appendix 1, we present further information about the sources of the paleographic age estimates.

The dates reported in Table 2 were obtained in most cases by multiple measurements of several sub-samples. All ¹⁴C ages were corrected to a δ¹³C value of -25‰, from the values indicated (Donahue, Linick and Jull 1990). This small isotope correction is a standard convention of ¹⁴C measurements (Stuiver and Polach 1977). The best precisions are *ca.* ± 20 BP. For other samples where larger uncertainties are quoted, the precision was limited either by scatter in the individual measurements, or by the fact that only a few independent measurements were made, due to sample-size limitations. The calibration curve used to obtain the calendar age was the 20-yr average of Stuiver and Pearson (1986), although in some cases we also refer to the 10-yr average curve of Stuiver and Becker (1986). Calculations of probability are quoted for 2-σ ranges, where the calibration program (Stuiver and Reimer 1986) produces two ranges.

With one exception, the dates of the documents determined by the ¹⁴C agree well with the dates previously suggested on the basis of paleographical analysis. These results are summarized in Figure 1, which shows the calibrated ¹⁴C ages plotted against paleographic age estimates. The calibrated age ranges are derived by applying the ¹⁴C age with uncertainties of 2 σ to the calibration curve of Stuiver and Pearson (1986). One exception was the first set of dates on 4Q258 (DSS-5), which was anomalously young and difficult to explain in terms of the expected age of the material. A second and cleaner sample of material was removed for dating. This second sample was subjected to an extensive acetone cleaning as described for Type 2 samples, as well as the acid-base-acid treatment, and gave a ¹⁴C age comparable to the paleographic age.

Samples of Known Age

Samples in Table 2 listed as DSS-25, -52 and -53 are all papyri of precisely known age, since they bear written dates. For the two papyri, DSS-25 (pap Yadin 21) and -53 (pap Yadin 19), our results agree within 1 σ with known values. For DSS-52, the 2-σ range of our measurements fails by 10 yr

TABLE 2: Radiocarbon Dates on Dead Sea Scrolls and Related Materials Measured at The University of Arizona

Sample no.	Sample	No. of runs*	$\delta^{13}\text{C}$ (‰)†	^{14}C age (yr BP)	Calibrated age	Paleographic age
AA-13415	DSS-1 4Q266	5	-22.1	1954 ± 38	1 σ : AD 5–80 2 σ : 45 BC–AD 120	100–50 BC
AA-13417	DSS-3 1QpHab	8	-20.8	2054 ± 22	1 σ : 104–43 BC 2 σ : 153–143 BC (3%) 120–5 BC (97%)	30–1 BC
AA-13418	DSS-4 1QS	3	(-21.2)	2041 ± 68	1 σ : 159 BC–AD 20 2 σ : 346 BC–317 BC (2%) 206 BC–AD 111 (98%)	100–75 BC
AA-13419	DSS-5A 4Q258 (first sample)	5	-22.6	1823 ± 24	1 σ : AD 134–230 2 σ : AD 119–245	~100 BC
AA-16060	DSS-5B 4Q258 (second sample)	4	-21.4	1964 ± 45	1 σ : 11 BC–AD 78 2 σ : 95 BC–AD 122	~100 BC
AA-13420	DSS-7 4Q171	7	(-21.2)	1944 ± 23	1 σ : AD 22–78 2 σ : AD 5–111	
AA-13421	DSS-8 4Q521	4	(-21.2)	1984 ± 33	1 σ : 35 BC–AD 59 2 σ : 93 BC–AD 80	100–80 BC
AA-13422	DSS-9 4Q267	5	-21.6	2094 ± 29	1 σ : 172–98 BC 2 σ : 194–45 BC	50–0 BC
AA-13423	DSS-10 4Q249	6	-10.8	2097 ± 50	1 σ : 191–90 BC 2 σ : 380–354 BC (8%) 242 BC–AD 6 (92%)	
AA-13244	DSS-11 4Q317	4	-20.9	2084 ± 30	1 σ : 164–93 BC 2 σ : 191–36 BC	
AA-13245	DSS-13 4Q208	9	-21.0	2095 ± 20	1 σ : 166–102 BC 2 σ : 186–92 BC	~200 BC
AA-13246	DSS-19 4Q22	2	(-21.2)	2044 ± 65	1 σ : 159 BC–AD 16 2 σ : 207 BC–AD 89	100–25 BC
AA-13426P	DSS-19a 4Q22 Patch	4	(-21.2)	2024 ± 39	1 σ : 98 BC–AD 13 2 σ : 120 BC–AD 63	50 BC–AD 50
AA-13430	DSS-22 4Q342‡	4	-20.8	1934 ± 47	1 σ : AD 14–115 2 σ : 43 BC–AD 141	
AA-13431	DSS-23 4Q344‡	3	-20.4	1902 ± 39	1 σ : AD 72–127 2 σ : AD 26–195	
AA-13432	DSS-24 4Q345	5	-19.7	2185 ± 60	1 σ : 373–171 BC 2 σ : 390–100 BC	
AA-13433	DSS-25 5/6 Hev 21 (pap Yadin 21)	3	-12.0	1799 ± 57	1 σ : AD 130–321 2 σ : AD 80–380	AD 130
AA-14984	DSS-50 1QIsa ^a	5	-20.4	2141 ± 32	1 σ : 335–122 BC 2 σ : 356–291 BC (24%) 250–103 BC (76%)	150–125 BC
AA-14986	DSS-52 Kefar Bebayou	4	-10.0	1758 ± 36	1 σ : AD 231–332 2 σ : AD 144–370§	AD 135
AA-14987	DSS-53 5/6 Hev 21	4	-10.8	1827 ± 36	1 σ : AD 126–234 AD 86–314	AD 128

*The number of independent determinations of ^{14}C age

†Values in parentheses are estimated based on the mean values for Dead Sea Scroll parchments.

‡The documentary texts 4Q342 (letter in Judeo-Aramaic) and 4Q344 (debt acknowledgment) can be dated as late as the Bar-Kokhba period, and such a late date confirms doubts regarding the Qumranic origin of these texts. These fragments, bought from a Bedouin, were probably mixed up with the Qumran fragments by antiquity dealers (M. Broshi).

§The 10-yr average calibration curve of Stuiver and Becker (1986) gave AD 133–386 (2 σ) for this sample.

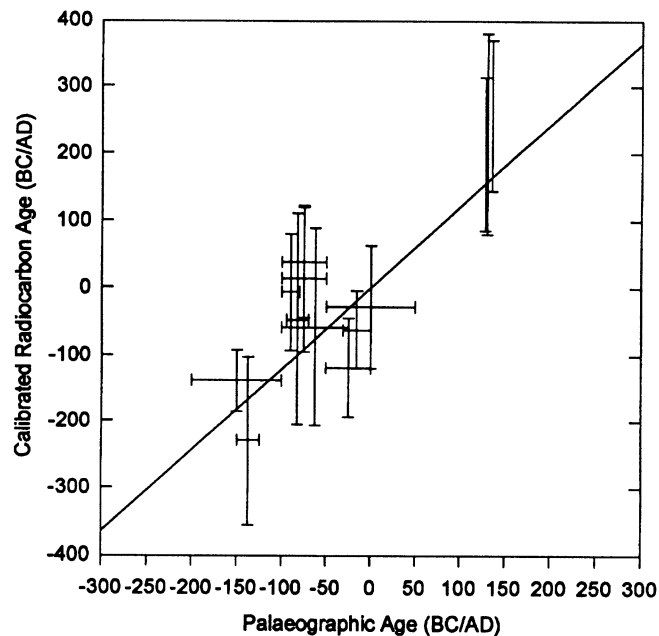


Fig. 1. Calibrated ¹⁴C age ranges vs. estimated paleographic ages of scroll samples. The calibrated ranges were deduced from measured conventional ¹⁴C ages, including 2 σ , using the tree-ring calibration curve of Stuiver and Pearson (1986). The ranges of paleographic estimates were chosen to include the range of the estimates reported in Appendix 1.

to include the known age. Interestingly, if the decadal tree-ring calibration curve of Stuiver and Becker (1986) is used, the 2- σ range of our measurement would be AD 133–386, and would include the known age. For a range produced by using the measured ¹⁴C age and 1 σ , the expectation is that there is a 68% probability that the range encompasses the correct age of the document. If 2 σ are used, the probability is 95%. It is also true that, in comparing known ages with a possible range of ages obtained from ¹⁴C measurements, the procedures for producing the calibrated age are such that the actual age can fall anywhere within the calculated limits.

Comparison to Zürich ¹⁴C Measurements

Sample DSS-50, which had been tested previously at the ETH Zürich Laboratory, was also measured in our study. This sample was taken from the same area of the scroll as the Zürich sample, from column XXXIX of the large Isaiah scroll from Cave 1. The ¹⁴C results of Bonani *et al.* (1991, 1992) yielded the result of 2128 ± 38 BP (ETH-6651), which is in excellent agreement with our value, 2141 ± 32 BP (AA-14984). We report the calibrated age range in Table 2. The weighted mean of the two measurements is 2136 ± 24 BP.

Comparison of Results to the Calibration Curve

All of the results discussed are presented graphically in Figure 2. The individual points are obtained by plotting the measured conventional ¹⁴C ages of the samples on the ordinate vs. the estimated paleographic ages of the samples on the abscissa. The fact that the individual points plot within 2 σ of the calibration curve indicates that the ¹⁴C and paleographic ages are in reasonable agreement. It

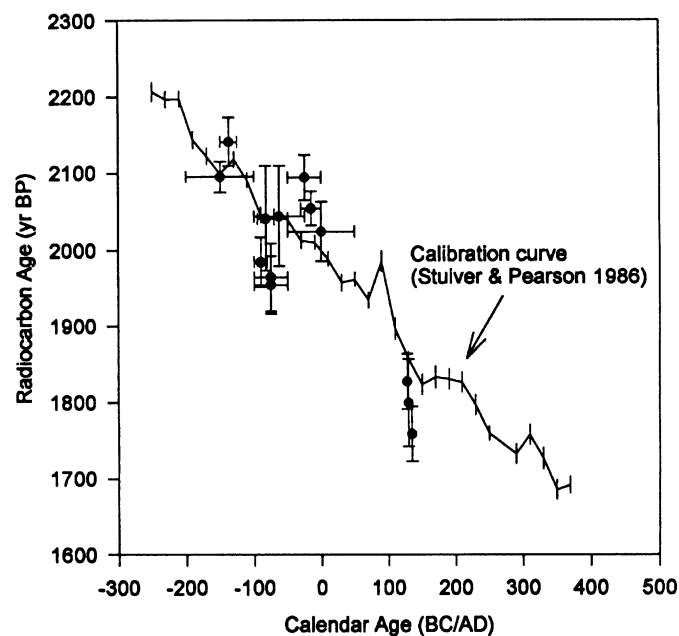


Fig. 2. Conventional ^{14}C age vs. calendar age. The solid curve shows the tree-ring calibration curve of Stuiver and Pearson (1986). The ordinate values for the data are conventional ^{14}C ages measured by AMS, as reported in Table 2. The error bars on the ^{14}C ages are 1σ . The abscissa values for the data are estimates of paleographic age from Appendix 1.

is interesting that the three date-bearing papyri are all of approximately the same age, and there is a tendency for our measured age ranges to be on the younger side of these known ages. It is possible that the calibration curve for AD 135 should be slightly lowered, as its position appears to be determined by a single point in the 20-yr calibration curve (Stuiver and Pearson 1986).

Linen Fragments

Two samples of linen, tested by AMS, yielded results in line with their anticipated dates based on context. These results are presented in Table 3. DSS-26 was a sample of cloth from Qumran Cave 4, to which a leather thong was attached, of the kind used to fasten the scrolls at Qumran (Carswell 1977). Significantly, the ^{14}C date for this sample fell solidly within the dating period established for the scrolls by both paleography and ^{14}C dating. DSS-27, a linen fragment with silk embroidery, was dated to the 12th–13th centuries AD. This sample was bought from antiquity dealers who represented it as material “from Qumran cave 2”; it most probably originated from Wadi Murabba‘at,

TABLE 3: Radiocarbon Dates on Linen Fragments from the Judean Desert

Sample no.	Sample identification	No. of runs	$\delta^{13}\text{C}$ (‰)	^{14}C age (yr BP)	Calibrated age
AA-13434	DSS-26, linen with leather thong Cave 4, inventory no. 1041	2	-26.5	2069 ± 40	160–41 BC (1σ) 193 BC–AD 11 (2σ)
AA-13435	DSS-27, linen, Cave 2 inventory no. 749	2	-26.3	664 ± 36	AD 1279–1376 (1σ) AD 1270–1392 (2σ)

where similar textiles were found (Crowfoot and Crowfoot 1961). Descriptions of these fragments are given in Appendix 2.

CONCLUSION

¹⁴C ages of 14 parchment and 4 papyrus samples found in caves in the Judean Desert have been measured by AMS. Measurements on samples of known ages are in good agreement with those known ages. Ages determined from ¹⁴C measurements on the remainder of the Dead Sea Scroll samples are in reasonable agreement with paleographic estimates of such ages, in the cases where those estimates are available.

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APPENDIX 1. DATES SUGGESTED FOR THE TEXTS ON THE BASIS OF PALEOGRAPHICAL ANALYSIS

The information given below for scroll samples includes the Q number, an abbreviated siglum following the name of the scroll, the PAM/Shrine photo number and the assigned paleographical date. The abbreviated sigla referring to the texts from the Judean Desert follow their conventional names. (For the most recent list, see Tov and Pfann (1995).) All photograph numbers are PAM (Palestine Archaeological Museum) numbers unless otherwise indicated.

- DSS-1. 4Q266 Damascus document, D^a 43.277 100–50 BC**
Comments: “The writing is in a semi-cursive Hasmonean hand which in Cross’ paleographic sequence may be dated to the first half of the first century B.C.E.” (Baumgarten 1992: 57). “The text is written in a semi-cursive Hasmonean hand which, in Cross’s paleographic typology, may be assigned to the beginning of the first century B.C.E.” (Baumgarten 1990: 153–165). 75–50 BC (Stegemann 1994: 166).
- DSS-3. Commentary on Habakkuk, 1QpHab col. XIII Shrine 7203-4 30–1 BC**
Comment: “The manuscript is written in an Early Herodian hand (ca. 30–1 B.C.), affecting the Palaeo-Hebrew script in a degenerate form when writing the Tetragrammaton” (Cross 1972: 4; Avigad 1965: 74).
- DSS-4. Community Rule, 1QS col. XI Shrine 7111 100–75 BC**
Comments: “As we have seen, it belongs to a special semi-formal tradition of the Jewish script, a Hasmonean exemplar of this style from about 100–75 B.C.” (Cross 1972: 4). “[A] date somewhat later than 1QIsa^a is to be preferred” (Avigad 1965: 71).
- DSS-5. 4Q258 Community Rule, S^d 43.244 Beginning of 1st century BC**
Comment: “The script of the oldest copy is dated by F. M. Cross to the beginning of the first Century B.C.” (Vermes 1991: 250; Cross 1956: 61).
- DSS-7. 4Q171 pPsa 41.303**
- DSS-8. 4Q521 Mess. Apoc. 43.604 100–80 BC**
Comment: “Cette écriture se placerait assez bien après celle de 1QIsa et 1QS, dans le premier quart de 1^{er} s., entre 100 et 80, plus ou moins contemporaine de SiraMas, mais après 4QDt^c” (Puech 1992: 480). (This text is the focus of much debate in research, in particular with regard to its possible connection with early Christianity.)

- DSS-9. 4Q267 Damascus Document, D^b 43.294 Latter part of 1st century BC**
Comment: “[A] formal hand of the latter part of the first Century B.C.E.” (Baumgarten 1992: 60).
- DSS-10. 4Q249 pap Midrash Sefer Moshe 43.409**
- DSS-11. 4Q317 Phases of the Moon 42.424**
- DSS-13. 4Q208 Astronomical Enoch, Enastr^a ar 43.210 End 3rd/beginning 2nd century BC**
Comment: “The handwriting of Enastr^a is rather unusual, but fairly archaic; it resembles ‘an archaic or early Hasmonaeon semi-formal script of ca. 175–125 B.C.’ (Cross [1961], p. 137, fig. I, line 6; cf. *ibid.*, line 7 and p. 138, fig. 2, line I). It seems to me, however, to be older than the alphabets discussed by Cross, and to be related, by many a detail, to the writings of fig. I, lines 2–5. As a result I would date 4QEnastr^a to the end of the third century or else the beginning of the second Century B.C.” (Milik 1976: 273)
- DSS-19. 4Q22 paleoExodus, paleoExod^m 42.582 100–25 BC**
Comment: “[MacLean] has dated this scroll along with 4QpaleoGen-Exod^l . . . and 4Q124 . . . within the period 100–50 or 100–25 BC, with this qualification: ‘Of these three contemporary manuscripts, I believe 4QpaleoExod^m to display the latest features and the greatest number of novel features which will see subsequent development’ (MacLean [1982], 78). On the basis of the extensive nature of MacLean’s study, as well as Cross’s endorsement of his conclusions, we accept his dating” (Skehan, Ulrich and Sanderson 1992: 62).
- DSS-19a. 4Q22 paleoExod^m col. VIII (patch) 42.648 50 BC–AD 50**
Comment: Skehan, Ulrich and Sanderson (1992: 85) state that “a patch was sewn from behind the leather to repair damage suffered by the MS after it had been inscribed.” The patch thus had to be later than the manuscript itself; E. Ulrich (personal communication, 1995) dates it to 50 BC–AD 50.
- DSS-25. 5/6Hev 21 (pap Yadin 21) purchase of crop in Greek Shrine 5195 11 September AD 130**
(Lewis 1989: pl. 26)
- DSS-50. Book of Isaiah, 1QIsa^a col. XXXIX Shrine 7039 150–125 BC**
Comments: “[T]he old Isaiah scroll . . . dates to c. 150–125 B.C.E.” (Cross 1992: 5). “A date somewhere in the second half of the second century B.C.E. for Isa^a, somewhat later than Nash, seems to be most reasonable and in keeping with the opinion of most scholars” (Avigad 1965: 69).
- DSS-52. XHev /Se 8a pap sale of a house, Kefar Bebayou 40.996 AD 135**
(Milik 1957; 1959: 138 and p. 25)
- DSS-53. 5/6Hev 19 (pap Yadin 19) deed of gift Shrine 5185 16 April AD 128**
(Lewis 1989: 83–87 and pl. 20)

APPENDIX 2. LINEN FRAGMENTS

Description by A. Baginski, Israel Antiquities Authority, Jerusalem.

DSS-26. Linen fragment with leather thong attached from Qumran Cave 4

Size: 3.0 × 2.5 cm. Warp: linen, cream S M 12/cm. Weft: linen, cream S M 12/cm. Technique: balanced tabby sewing threads: linen Z₂ S. Description: small triangular linen fragment, on one side remains of a rolled hem. A leather strip is sewn to one corner (0.7 × 4.0 cm). The same box contains another small fragment of the leather thong (0.6 × 2.7 cm) and a larger square piece of leather which is folded and has two slits (2.7 × 3.0 cm), through which the thong was probably inserted. The linen fragment is very similar to some textiles from Qumran Cave 1; the leather thong and pieces are similar to those exhibited currently in the Israel Museum (Carswell 1977: 23–28; Crowfoot 1955: 37 no. 56, 38 no. 77, pl VII no. 26; Sussmann and Peled 1993: 114–115; Sheffer and Granger-Taylor 1994: 176 no. 102[A]).

DSS-27. Linen fragment with remains of silk embroidery from Qumran Cave 2

Size: 6.0 × 4.5 cm. Warp: linen, cream Z M 18/cm. Weft: linen, cream Z M 18/cm. Technique: balanced tabby weave. Decoration: embroidered, silk I, buff, darning stitches. Description: worn linen fragment with weaving faults; remains of silk embroidery, possibly of geometric pattern, but unrecognizable due to poor preservation. Fragment is most likely medieval or originating from Wadi Muraba‘at, where similar textiles were found (Crowfoot and Crowfoot 1961: 51–63, pl. XVII no. 2; Lamm 1937: 65–76; Makie 1989: 81–101).

STORAGE CONDITIONS AND PHYSICAL TREATMENTS RELATING TO THE DATING OF THE DEAD SEA SCROLLS

NICCOLO CALDARARO

Department of Anthropology, San Francisco State University
1600 Holloway Avenue, San Francisco, California 94132 USA

ABSTRACT. The Dead Sea Scrolls have been analyzed by paleographic, non-destructive and destructive testing. The dates of their creation have been in dispute since their discovery. Research has established their authenticity, but a variety of conditions including the methods of skin preparation, variation in storage conditions and post-discovery restoration treatments could have introduced changes now affecting dating efforts. Comprehensive analyses were not possible until recently. Such analysis must be performed to establish a concrete framework for all the texts.

Professor R. B. Blake told a story in response to a question of why so little remained of writing on leather. He said that on one of his expeditions to Asia Minor, one of his native servants exhibited proudly some chamois trousers of his own manufacture, upon which Professor Blake detected with sorrow, traces of medieval writing (Reed 1972).

INTRODUCTION

A recent ^{14}C study of 14 Dead Sea Scrolls by Bonani *et al.* (1992) is a welcome addition to the analytical literature on the Scrolls. The authors have undertaken a more comprehensive sampling than any previous study, an effort that T. B. Kahle and I proposed in an article in *Nature* in 1986. In that article, we commented on amino acid racemization analysis of the Dead Sea Scrolls published by Weiner *et al.* (1980). Our comments then, as mine now, relate to the potential effects on dating results of prior storage conditions and restoration treatments. Other points raised in the *Nature* article referred to the different types of animal skin used and the methods of skin preparation to produce a useful writing surface. In this article, I present a general background of the Dead Sea Scrolls and a more detailed account of chemical treatments used on the Scrolls and other factors, including environmental conditions resulting from storage, which may be sources of variation in the physical properties of the Scrolls relating to dating efforts.

DESCRIPTION OF THE PROBLEM

The story in the epigraph emphasizes the importance that leather has always had to mankind. The meager remains of the vast parchment libraries of antiquity have perished mainly by fire, water and biological degeneration. The discovery of the Dead Sea Scrolls in 1947 focused the attention of the public on a few fragments of what was the great Semitic heritage of writing and scholarship. Since our cultural foundations in writing originate in this heritage, every discovery lifts the mists from the rich, and now largely lost, literature of the pre-Christian era. Each time caches of ancient books are found, some commentator asks if there are not more texts waiting to be found. Why have some ancient texts survived and others perished? In most archaeological excavations, the only traces of writing on leather are dark stains in the soil. Occasionally, in a bog or dry cave, given remarkable conditions for preservation, a fragment or scroll will “persist”. Skin is not only of great use to man for everyday use, but it is food for many life forms. It is because of its impermanence that we study its conditions of preservation. In this study, the age of the surviving objects is of great importance. Knowing the age of leather objects, one can determine a relative time frame for aging. Properly prepared parchment and vellum objects can survive in stable environmental conditions for several thou-

sand years (Reed 1972). Writing on perishable materials, such as paper and papyrus, and less perishable materials such as stone and clay, has survived, but only a sample of what existed remains.

When the sensational discovery of the Dead Sea Scrolls was first reported in 1947, most scholars responded with mistrust. Documents similar to the Scrolls had been appearing on the antiquities market for many years prior to 1947, without provenience and with mysterious “original owners”. As was obvious from the excavation of the caves (Barthelemy and Milik 1955; Benoit, Milik and de Vaux 1961; Baillet, Milik and de Vaux, 1962), most of the scrolls originally deposited in the caves had been removed long before the caves’ discovery. Kahle (1959) provides historical evidence that the Qumran caves were discovered in the 9th century AD. Earlier “discoveries” of scrolls were made in the first part of the second century (Driver 1965). The quantity of broken scroll cases and fragments found on the floor of Qumran I indicates that the original library was much larger than the amount found in 1947 and the following years. It is possible that the caves were “mined” of their valuable contents for some time by desert tribes. The story of the shepherd who followed a stray animal to a cave (Cross 1954) is a curiously common tale of “lost” treasure, such as the Avroman deeds (Minns 1915).

Creators of forgeries tend to fill known gaps in the archaeological record, as Kurz detailed in his classic study (1948) and I recently expanded (Caldararo 1992). In this context, the Temple scroll should be the most suspect, as it contains supposedly lost instructions on construction of the Temple of Jerusalem as well as the missing “Statutes of the King” (Yadin 1992).

The study of the Dead Sea Scroll fragments found in 1947 has been impeded by a division of the fragments among nations and scholars with restrictions on publication and translation rights. Recent events have eased this situation, and photographs of the Scrolls were circulated among scholars and students by the Biblical Archaeological Society (Shanks 1992). Misunderstandings and rumors regarding the content of the fragments were generated in the past 40 years because of the sequestration of the Scrolls and by the stalled translations (Wilson 1969).

With the release of the original photographs, the Scrolls were viewed for the first time in their entirety. However, during the 50-yr interval, the fragments had aged and deteriorated, either due to improper storage or to treatments for “improved” legibility. Comparison of the original photographs taken after discovery and the fragments now in the Rockefeller Museum in Jerusalem show, in many cases, vastly different objects. Microfilm archives of the Scroll fragments kept at the Huntington Library in San Marino, California—where the *ca.* 3000 photographs reside—seem to support the contention that the Scrolls have undergone aging expected of prepared skin exposed to normal environmental changes, hydrolysis and other agents of deterioration (Hansen, Lee and Sobel 1991). This information should be considered with caution, however, as no comprehensive comparative analysis of the Scroll material or the original Scrolls has been undertaken. Frank (1992) asserts that much of the Scroll material has faded from exposure to light. Weiner (personal communication, 1995) notes that, from his examination of a Scroll and comparison of it to a photograph taken in the 1950s, he sees no difference.

STATE OF PRESERVATION

A conference at Stanford University in 1992 focused on: 1) access to the Scrolls by scholars; 2) the relation of the Scrolls to the Essenes; and 3) the influence of the authors on the early Christian church. Little attention was paid to the physical condition of the Scrolls, their status as documents when compared to other parchment writings or to the body of the Semitic text. A brief study com-

paring the surviving Scrolls and the physical layout of the Qumran site to our knowledge of the libraries of antiquity (Pedley 1964) supported the idea of a library at the site.

Other fragments and manuscripts from the area do not conform to the manner in which the Qumran I objects are assumed to have been originally stored (Cross 1954, 1958). Kahle and Caldararo (1986) described how restoration treatment and parchment preparation methods affect the results of dating studies. Reed and Poole (1962) concluded from examining Scroll fragments from Cave 4 that they were not all made at precisely the same time or according to the same methods. Quite possibly these prior treatments or different histories of the body of materials termed "Dead Sea Scrolls" could affect ^{14}C dating. The testing procedures described by Bonani (1992), with samples taken from different parts of scrolls, would seem to mitigate contamination problems.

The wide range of treatments and exposures (Tables 1 and 2) might call for a controlled study of ^{14}C analysis applied to ancient samples with different preparations and modern samples exposed to conditions that degrade skin products and to artificially aged samples. Variables would then be: 1) preparation methods; 2) restoration chemicals and 3) chemical interactions during aging of skin components and residual reagents. Such an experiment would provide us with controlled results on contaminant removal from test samples. In the preceding article (Jull *et al.* 1995), the ^{14}C results of sample 4Q258 indicate the potential value of such a study. In this study (Jull *et al.* 1995, Table 2), the dates for their sample 1 of 4Q258 were *ca.* 300 yr younger than the paleographic dates. More intensive pretreatment of a second sample from 4Q258 fell within the paleographic range. Aging under natural conditions can show different results due to unforeseen variables (Table 3): for example, leather is unstable at a pH other than 3 to 6. A low pH can indicate an acid attack, perhaps a residue of a tanning process (Stambolov 1969). Poole and Reed (1962) found gallic acid in every sample they analyzed from Cave 4. Leather so degraded when in contact with moisture will darken, in many cases, irretrievably. In some of the Qumran caves, percolating water had so damaged scroll material that the floors of the caves were covered with gelatinous slime (Reed and Poole 1962). White ants damaged some Scrolls, and where fragments were found in a heap, covered fragments remained white whereas exposed fragments turned dark brown (Harding 1948/9).

How does one describe this darkening? Is there a comparable physical condition among objects prepared for writing in the Near East? Reed (1972) compared the surface of fragments of the Dead Sea

TABLE 1. Treatments Applied to Scrolls and Fragments

Scroll(s)	Material	Reference
Cave 1	Glycerine	Plenderleith (1955)
	Water	
Most scrolls	Water	Cross (1954)
	Moisture	Benoit (1956)
	Alcohol	Wilson (1969)
?	Oils	"Araldite" (1961)
	Araldite on Temple Scroll?*	
Copper scroll	Araldite celluloid in acetone Durofix	Baker (1962)

*The plastic substance reported by Bonani *et al.* (1992) on the Temple Scroll may be celluloid, which was noted in the literature by a reference to Scott's (1927) work on an Egyptian scroll. Scott used celluloid in acetone and amyl acetate 1:1.

TABLE 2. Tests on Scrolls and Fragments

Scroll	Test	Sample(s)	Reference	Results
Cave 4	For gallic acid	Numerous	Poole and Reed (1962)	Positive
?	For tannins	?	Reed and Poole (1964); Reed (1972)	Positive for ellagic acid*
Isaiah Cave 1	¹⁴ C	Linen wrapping	Libby (1955: 84)	2117–1717 BP
Fragment Wadi Murraba'at	Shrinkage	Scroll	Burton, Poole and Reed (1959)	Relative date correlated with other methods to 130 BP†
Fragment from Cave 4				Relative date similar to Murraba'at
Isaiah-B Thanksgiving	Effects of relative humidity	Unknown sample from Temple A was contaminated with what appeared to be PVA‡; it was not tested.	Schilling and Ginell (1993)	No overall consistency in response of Scroll fragments in relation to modern degraded parchment. Scroll fragments did respond more slowly to RH.
Genesis Apocryphon				
Temple A				
? Cave 4				

*Tested a variety of other leather objects from other locations and periods

†Tested by the same method as fragments of an Egyptian Aramaic letter (ca. 2500 BP). The results fell in an expected range of the Murraba'at fragment.

‡Polyvinyl chloride

TABLE 3. Observations Relating to Condition of Scrolls

Scroll	Observation	Citation
?	Scrolls attacked by white ants; covered Scroll fragments were white; exposed fragments were brown.	Harding (1948/49)
Cave 4	Electron microscopy shows collagen fibers in good condition, but with characteristics of aging. Light microscopy shows distribution from tannin stain.	Reed and Poole (1962) Reed (1972)*
18 Dead Sea Scroll samples; 5 from Murraba'at	Histological structures of follicles well preserved. Follicle grouping perfect only in 2. Dead Sea samples and 2 from Murraba'at. Fewer follicles found in modern parchment compared to ancient samples. In general, ancient follicles lost pigment.	Ryder (1958)† Ryder (1963)

*Samples from Wadi Murraba'at and Romano-British leather found at Catterick included.

†Samples from Aramaic Documents included.

Derrick (1991) studied fragments of the Scrolls using FT-IR spectroscopy. Her results were non-homogeneous. Test results varied from area to area within a fragment. Her samples were taken from the outer edges of Scrolls and may represent more degraded areas. Some fragments were less degraded than others: for example, Temple B and Cave IV 9A3 were most degraded, and those from Khirbet Hird were the least degraded. Sectioning and IR microspectroscopy and cross-polarized light microscopy showed that degradation products and inorganic components were concentrated on or near the surface. Alum was found only in the Khirbet Hird samples and the sample from the Temple Scroll. Silicates, presumably from pumice used as an abrasive in skin preparation, was found in all of the exterior surfaces of the Scroll pieces except for the backside of the Thanksgiving Scroll. Carbonates were found in all samples but were in greater concentrations on samples from Cave IV.

Scrolls with those of the Aramaic Documents (500 BC) and the Philistine Documents (900–700 BC). Apparently, the Dead Sea Scroll fragment and the Aramaic Documents had been treated with vegetable tannins. This was confirmed by sectioning and examination with light and electron microscopy.

The Philistine Documents were in better condition than the other two and, unless the skin was remarkably preserved, their authenticity is questionable. Other tests comparing the three Philistine Documents showed that the Philistine Documents were similar to modern parchment. For a description of the distinguishing characteristics of parchments and leather as they apply to Judaic religious uses, see Figure 1. Reed and Poole (1962) divided their samples from Cave 4 into three groups based on appearance, feel, microscopic examination and some chemical tests. These are:

Parchment-like fragments	57 samples
Gewil-like fragments	12 samples
Leather fragments	9 samples

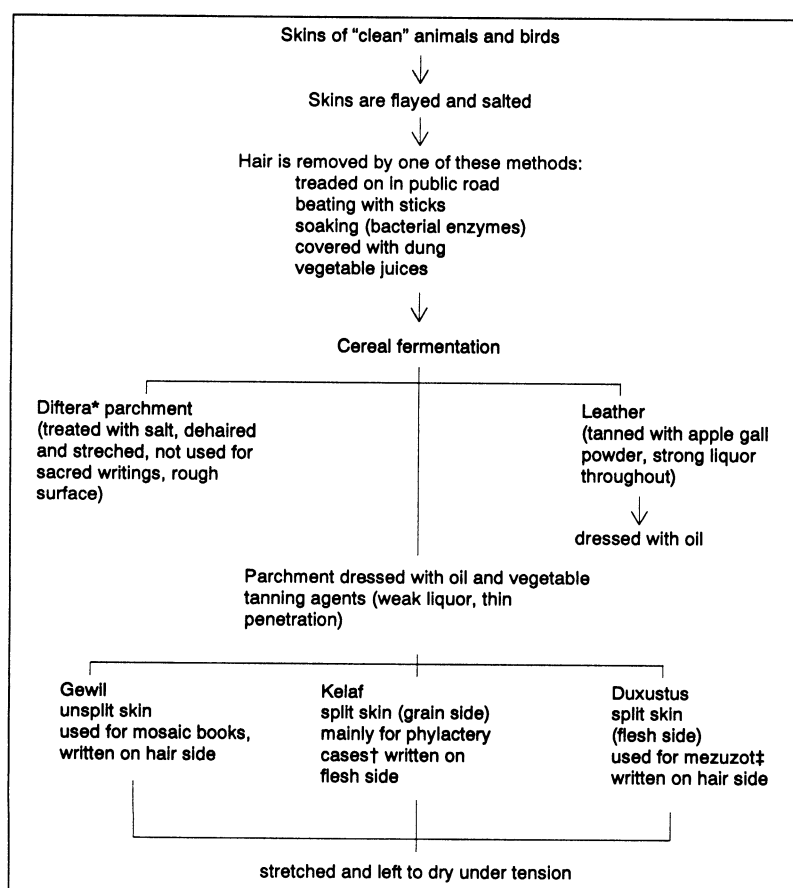


Fig. 1. Description of parchments and leather fabrication; data from Reed and Poole (1962).
 *Diftera: a rough parchment, suitable for everyday use; †Phylactery case: a case containing a strip or strips of parchment inscribed with passages of Scripture; ‡Mezuzot: pieces of parchment inscribed with texts from Deuteronomy on one side and the name of God on the other. The *mezuzah* is rolled and put into a case and attached to the doorpost of a residence.

Restorers have known for some time that faded ink can be regenerated by rubbing parchments and leather with tannic acid. Gallic acids—as well as other acids—were often applied hot and had remarkable results (Wachter 1981–1982). This may partly explain the blackened appearance of the Shapira Scrolls (Allegro 1965). Alum was often applied by restorers to increase the adhesion of ink and to prevent mold. Comparing the blackened and gelatinized areas of some of the Scrolls with parchment documents that have been exposed to acid treatments might identify which Scrolls have been treated. This might also explain some of the wide range of testing used by various researchers (*e.g.*, Weiner 1980; Schilling and Ginell 1993).

It would be interesting to continue these studies on more fragments from Dead Sea Scroll materials, the Damascus Document, the Avroman deeds (*ca.* 100 BC), Stein's leather writing fragments from Khotan, and some of the oldest surviving examples of writing on skin from Egypt (Driver 1959). Other skin documents that could provide samples for a varied series are from the Firkowitch Collection in the Russian Public Library in Leningrad. The conditions under which some of these objects were preserved are known and assumptions can be made for the others. Initial studies in this direction have had conflicting results.

Rebricova and Solovyova (1987) compared parchments with an electron microscope and subjected samples of modern, 18th-, 17th- and 12th-century parchment to enzyme attack, biochemical analysis and microorganism destruction. These results demonstrated the general stability of parchment to resist destruction, but soluble collagen varied among the samples unrelated to age, whereas sample hydrolysis by collagenase was consistent with age. Reed (1975) studied preparation methods for parchment and how these affect aging; Hansen, Lee and Sobel (1991) continued this research with vellum and parchment. Their results explain some of the mechanisms leading to variations in stability and resistance to degradation in skin products. Horie (1990) studied the deterioration of objects made of skin in museum collections, which detailed the problems in identifying ideal conditions to prevent deterioration. Hansen, Lee and Sobel (1991) found significant differences in parchment response to changes in humidity with the method of preparation, although their samples were limited to Medieval, Talmudic and modern objects. Comparing unidentified Dead Sea Scroll fragments, the authors report unpublished experiments showing that, in contrast to modern parchment, the Scroll fragments exhibited a state of deterioration “consistent with their age”. They base this conclusion on the reduced extraction of collagen (70% compared with 90% for modern parchment) and on the fact that the extent of degradation varied but that evidence of denaturation, hydrolysis and oxidation were associated to a greater extent with both brittle areas and darker, discolored areas.

However, considerable damage to the Scrolls resulted from their storage in the basement of a bank. The humidity caused extensive microorganism growth, rendering some of the Scrolls illegible. The Scrolls were cleaned but no report has been published. McCarter (1992) contends that the Copper Scroll is deteriorating and has actually lost mass along the edges of the saw cuts made by Baker. If corrosion is active, steps should be taken to stabilize the Copper Scrolls. This recalls Corwin's efforts to stabilize the Copper Scrolls by reduction in the 1950s (“Unrolling” 1956). Corwin experimented with replicas of the Copper Scrolls created to mimic the metallic composition and corrosion products and adhered debris of the original. These conservation experiments showed promise, but the treatment was never applied to the Scrolls.

DATING AND PROVENIENCE

The fact that the Scrolls have been substantially altered is not so disturbing to an archaeologist as that none of the Scrolls, as we have come to know them (Table 4), has provenience. Not only are

there few completed archaeological reports for the caves and Qumran, but what has been presented as preliminary reports is inadequate, inconsistent and lacking in scientific information (Caldararo 1984–1985). None of the original Scrolls was found *in situ*, and none has been linked to any of the caves by scientific evidence. Teicher (1963) and Davies (1988) criticized the archaeology related to Qumran and the caves related to the Scrolls and their fragments. Barthelemy and Milik (1955) showed drawings and photographs of Cave 1, but as the objects were found on the surface, the loci of their discovery were not shown, nor were stratigraphic notes made.

TABLE 4. The Original Dead Sea Scrolls

1.	St. Mark's Isaiah Scroll (1QIsaa) Associated with Cave 1
2.	Hebrew University Isaiah Scroll (1QIsab) Associated with Cave 1
3.	A Midrash on the Book of Habakkuk (1QpHab)
4.	Manual of Discipline(1QS)
5.	The War of the Sons of Light with the Sons of Darkness (1QM)
6.	Thanksgiving Psalms (1QA)
7.	Lamech (Genesis Apocryphon) (1QApoc)

This in contrast to Yadin's 1963 report, which clearly depicts locations and details the excavation. Attempts to associate writing materials removed from caves by the Ta'amireh tribe are horrifying in documenting their destructive methods (Lapp 1978).

As T. B. Kahle and Caldararo suggested in 1986, the Scrolls need to be put in context, which can be done, now that the Scrolls are available for examination. The Scrolls and the fragments found in the caves, as well as other related skin writing materials found elsewhere, such as the Damascus Document, should be examined using a variety of non-destructive testing methods to build a database with which to determine their associations.

A first step would be to photograph all the fragments that came from the caves since 1947, using infrared film, and then to compare the photos with those taken by Najib Albina of the original state of the Scrolls (Sanders 1992). Unfortunately, some of Albina's negatives are now ruined. Reed (1991) has undertaken a survey of the Scrolls, fragments and photographs. If this work can be correlated with the infrared imaging of Zuckerman and Bearman (Wilford 1993), reconstruction may be possible. The bulk of these data will enable the comparison with the Scrolls as they first appeared before handling, exposure and treatments resulted in their present, deteriorated condition. As the ¹⁴C data have been found to contain significant discrepancies (Rodley 1993), the Scrolls should be sampled by separate groups and assigned to independent testing sites as was done with the Shroud of Turin (Gove 1987, 1990).

One of the central controversies surrounding the Scrolls is when they were made and who made them. Linked to this is the question of whether they constituted a library, and if they were hidden or stored, as in a *geniza*.¹ The contents were later removed and buried (Bruce 1950). Much of the debate has centered around the history of Judea in the 200 yr before the current era (BCE), that is, before the birth of Christ, and the first 100 yr of the current era (CE). The Scrolls have been associated with the Jewish Essene and the Zakodite sects, among others, and their search for religious and cultural freedom from their Greek and Roman conquerors. The main theme of this association with

¹A *geniza* is a room in or near a synagogue into which all sorts of written and printed material are deposited. The materials placed into such a room were not intended to be kept as in an archive, but were intended to remain there undisturbed for a certain time (Kahle 1959).

various sects is the emphasis that the Scrolls were hidden to prevent their destruction. This is curious, as neither the Greek Selucids nor the pagan Romans had a record of destroying the written works of their subjects. The Romans suppressed the religious works of only one group up to the first century, the cult of Isis (Gibbon 1932). The Ptolemy's were known to seize the libraries of peoples to enhance their great collection at Alexandria.

Schiffman (1992) suggested that the Scrolls and the fragments from the caves were stored before the early Christian church had developed the New Testament. He also emphasized that the body of the Scrolls and the fragments from the caves constitute a variety of materials that do not fit the concept of the library of a narrow sect. This is a nebulous idea and introduces the prospect of an interesting study: to compare the contents of the Gnostic library discovered at Nag Hamadi and that of the Manichean writings found at Turfan with those of the Dead Sea caves and scrolls. Do they each represent a single type of collecting and purpose, or do they have similarities? Pedley's (1964) efforts provide some background information, but no firm conclusions.

Do the Dead Sea finds constitute a library or a geniza? First posed by Sukenik, this question was investigated in some detail by Del Medico (1958). Do the documents found in the Dead Sea caves compare with the remnants of the Cairo Geniza or the Islamic documents found in the walls of the Great Mosque of Sanaa (Dreibholz, 1983)? Here is a link to wider influences in the religious literature of the Middle East. We know that the Karaites were influenced by writings found in caves, and in the works of Ja'kub al-Kirkisani written *ca.* 937 are references to a Jewish sect he calls "cave people", who were active after the Sadducees and before the Christians. The Karaites refer to them as do Muslim writers of the same period. This leads to a third possibility, that the collection of writings in the caves were not deposited in haste, but were part of a community that lived in or near the caves where the writings were kept.

Kahle (1959) suggested, from considerable evidence, that the Karaites had removed several Scrolls from the caves near Qumran *ca.* 800 and that the fragments of Scrolls—especially the Testament of Levi—found in the caves probably belong to these Scrolls. It seems curious though, that they would remove only a few Scrolls. It seems more likely that the Scrolls were controlled by the desert tribes even then and that they sold these earlier few from their cache in the same manner as they did in the 1940s.

A fact that is only beginning to become clear is that the body of writings from the caves of the Dead Sea is varied and rich. This quality was also noted in the Gnostic library of Chenoboskion (Jonas 1963). Another striking similarity is that the teachers of the literature of Chenoboskion are never identified by name, as is true of the Dead Sea writings. Jonas (1963) noted the numerous similarities in the Dead Sea writings and those of the Gnostics and even conjectured a contemporary link between the Essenes and Gnostics. It is likely that we underestimate the number of desert religious communities of antiquity and their distribution and interaction. Gibbon remarks, quoting various contemporary sources, that a fugitive like Athanasius could elude capture by the Roman military and civil authorities in the wastes of Egypt and Palestine due to the community of ascetics who survived there.

SURFACE TREATMENTS

A treatment that would reveal much about the Scrolls is rubbing with oil of cedar. Thorough sampling of the original Scrolls and fragments with infrared spectroscopy (FT), emission spectroscopy and particle induced X-ray emission (PIXE) would establish a firm database for comparison. Kahle (1959) quotes from the pseudepigraphic work, *Assumption of Moses*, which dates from the begin-

ning of the current era, that one should anoint books with oil of cedar and place them into earthen vessels. This was advised to protect them for all times “to the end of the days” (Kahle 1959: 15). In Cave 1, two Scrolls were found still wrapped in linen and sealed in ceramic jars. Some of the Scrolls and fragments were covered with bat dung, especially the Book of Lamech (Genesis). Had they been removed from earthen vessels by looters? Can scrolls and jars be associated in cases where the jars exist only in fragments?

At separate intervals after discovery, the Scrolls and the numerous fragments were treated by various methods to unroll them, to clarify the writing and to preserve their surfaces or ink, increasing the likelihood of contamination. Wachter (1962a, b) and Stambolov (1969) describe the use of parchment size (a glue made from parchment scraps) sprayed or painted on to the surface as a consolidant, a practice well known and widespread by the 10th century. Reed (1972) noted the use of castor oil applied either “neat” (undiluted) or with an organic solvent to increase the flexibility of parchment documents. Plenderleith (1955) found that fragments were permeated with a black bituminous substance which he identified as a decomposition product of the skin. Wilson (1969) reported extensive treatment with moisture and oils, including castor oil, to remove clay from the surface or to relax the skin. Some of these uses may be derived from Wachter (1962b) who listed spermaceti, sperm oil, neatsfoot oil, lanolin, milk, unbleached beeswax, Japan wax (from sumac plants) and Zapon (nitro-cellulose applied in acetone).

Driver (1965) noted that some letters had been re-inked in the Isaiah A Scroll. Although contemporary repairs, corrections and marginal notations are common in the Scrolls, these re-inked letters appeared to Driver to be very recent. Milik, de Vaux and Baker (1962) reported the use of celluloid in acetone, which was a popular consolidant at the time (Caldararo 1987). Wachter (1962b) noted that the use of celluloid in organic solvents also provided sterilization against microbes. Baker (1962) described the use of Araldite 102 with hardener 951, and toluene for increased penetration of the Copper Scroll, and Durofix.

R. J. Gettens developed a method to unroll the Lamech Scroll (Genesis Apocryphon), humidifying it in a chamber at 50% relative humidity (RH) (“Unrolling” 1956). The Scroll was removed before he could try the method, but in 1955, N. Aviged, Yadin and James Bieberkraut used a similar method, at 75–78% RH at 17°C (“Unrolling” 1956; Yadin 1992). Benoit (1956) also reported a similar method. Plenderleith alternated humidity to relax and consolidate the skin with refrigeration to consolidate (Bruce 1950).

Stegemann (1991, 1992) developed an ingenious method for re-assembling scrolls from fragments based on wear and deterioration. He matches the pattern of both processes in the fragments as they would develop in a scroll. He uses clues such as color to associate missing pieces with acids. Color often reflects varied exposure to light (Harding 1948/9). Stegemann also uses ruled lines as clues to association. However, Driver (1965) believed that ruling was not consistently applied in the creation of the Scrolls, and that ruled texts are nearly absent in pre-Christian times.

Rabinovich (1994) reported that Scotch™-type, pressure sensitive tape, used to hold fragments of Scrolls together, darkened and stained the skin. Conservators used fuller’s earth (an impure hydrous aluminum silicate) to remove the tape and adhesive, sometimes restoring obscured writing. This is curious, as pressure-sensitive tapes like Scotch™ tape usually age in a fashion resulting in embrittlement of the adhesive and detachment of the carrier film (Feller and Encke 1982). At this point, the adhesive is so brittle that it must be removed with organic solvents or by mechanical means. In some cases, the related stains can be reduced on paper objects (Caldararo and Sheldon 1992). From tape that has not aged so much as to become embrittled, the carrier can be removed using directed hot air

or a hot spatula, and the remaining adhesive picked up with a granular material such as fuller's earth. This is probably what Rabinovich (1994) described.

CONCLUSION

A detailed and systematic study of the physical aspects of the Scrolls is clearly needed to analyze preparation residues, traces of ritual use, degradation products, aging characteristics and treatment residues and effects. Such an investigation will then lead to a body of knowledge with which one can compare the Scrolls to other samples of ancient writing from which an analytical context can be developed. This context will enable us to better understand how to associate fragments as well as how to regard variations in dating methods among associated and non-associated remains.

In this paper, I have surveyed the published literature and abstracted information on storage and restorative treatments used in preserving the Dead Sea Scrolls for further study. A more thorough and formal survey should be organized to query those who have executed treatments on the scrolls. T. B. Kahle and I have made numerous queries over the past ten years, only one of which was answered (by H. Plenderleith). Although many of these individuals have passed away, information may still be available in notebooks and other personal papers. It is time for a concentrated effort to gather this material and make it available to the laboratory researchers who are attempting to date the Scrolls.

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INTERCOMPARISON OF HIGH-PRECISION ^{14}C MEASUREMENTS AT THE UNIVERSITY OF ARIZONA AND THE QUEEN'S UNIVERSITY OF BELFAST RADIOCARBON LABORATORIES

R. M. KALIN,¹ F. G. McCORMAC,² P. E. DAMON,³ C. J. EASTOE³ and AUSTIN LONG³

ABSTRACT. High-precision measurements were completed concurrently at the University of Arizona and the Queen's University of Belfast on blind samples of Irish oak originally measured for the 1986 radiocarbon calibration curve. Subsequent single-year *Sequoiadendron* results were decadal averaged and compared with published results on decadal Douglas-fir samples. The results of these intercomparisons show that the Arizona high-precision results compare favorably with published values from the University of Washington, but show a systematic offset with published Belfast data.

INTRODUCTION

During the last four years, considerable discussion has focused on quality assurance/quality control (QA/QC) and intercomparison between radiocarbon laboratories (Long and Kalin 1990; Rozanski *et al.* 1992; Stuiver, Long and Kra 1993). The results of international intercomparison studies (Scott *et al.* 1992) allow laboratories to identify bias in their ^{14}C results, as compared with the geometric mean of all the participating laboratories. Few laboratories produce high-precision ^{14}C analyses due to the time, expense and effort required to determine natural ^{14}C activity to $\pm 0.2\%$ precision.

Two publications dedicated to the calibration of the ^{14}C time scale have resulted from high-precision ^{14}C research (Stuiver and Kra 1986; Stuiver, Long and Kra 1993). Studies of variations in the ^{14}C record (Jirikowic 1994; Stuiver 1993; Jirikowic and Kalin 1993; Damon and Jirikowic 1992; Stuiver *et al.* 1991) show that solar, geophysical, oceanographic and paleoclimate changes may account for variability in the record. Therefore, to study this variability with data that were measured at different laboratories on wood from different geographical settings, it is imperative that detailed and continuing intercomparisons between high-precision laboratories be conducted to determine any minor laboratory bias in the results.

METHODS

The University of Arizona ^{14}C laboratory began using liquid scintillation counting (LSC) in 1992 for high-precision measurement of natural ^{14}C variations. Bidecadal samples of Irish oak, originally measured by Pearson *et al.* (1986), were provided by Prof. Michael Baillie, Palaeoecology Centre, The Queen's University of Belfast to both Arizona and Queen's ^{14}C labs. The bidecade that each sample represented was not known until the results had been completed. As part of ongoing research at the University of Arizona, single-year *Sequoiadendron* samples were separated at the Laboratory of Tree-Ring Research. Due to limited sample size for single-year samples, we decided that sample size for LSC measurements at Arizona could not exceed 7 g carbon. Therefore, counting times would have to be extended to ensure that $\pm 0.2\%$ precision was attained.

A slightly modified treatment after Linick *et al.* (1986) was used to remove the non-cellulose matter from the wood, leaving only holocellulose. This new procedure involves the use of an ultrasonic bath to facilitate the removal of resins from the wood prior to soxhlet reflux treatment. This modifi-

¹Department of Civil Engineering, The Queen's University of Belfast, Belfast BT7 1NN, Northern Ireland

²Radiocarbon Laboratory, The Queen's University of Belfast, Belfast BT7 1NN, Northern Ireland

³Department of Geosciences, The University of Arizona, Tucson, Arizona 85721

cation was needed to remove the excessive resin in *Sequoiadendron* samples. The procedure listed in the ^{14}C laboratory QA/QC manual is:

1. Weigh sample and record all relevant sample information in notebook.
2. Cut sample into matchstick pieces with hammer and chisel.
3. Pulverize sample in the Wiley mill (20 mesh). **Make sure mill is spotless**, clean mill before and after sample with Kimwipes® and 95% ethanol to ensure sample integrity.
4. Resin extraction:
 - a. The sample is placed in a 1000-ml beaker and 500 ml of toluene is added. The beaker is placed in an ultrasonic bath for 30 min. The toluene is decanted off, and the process is repeated until the removal of resins ceases.
 - b. Clean soxhlet apparatus.
 - c. In the bottom of soxhlet, place *ca.* 2.5 cm glass wool, place the ground sample on glass wool, then cover with *ca.* 2.5 cm glass wool. Make sure the sample is below the siphon outlet. Put 400 ml of 95% ethanol into the boiling flask (be sure there are sufficient boiling stones).
 - d. Assemble the soxhlet apparatus and turn on cooling water to the condensing tower.
 - e. Turn on the heat, check the apparatus after 1 h to ensure proper function. Record the date and action in the lab book.
 - f. After 24 h, turn off the heat, let cool.
 - g. Remove the 95% ethanol solution and discard in an appropriate waste bottle.
 - h. Dry the sample by blowing air through it.
 - i. If the sample is not clean, refill the 500-ml bulb with 400 ml of 95% ethanol and restart extraction. Record the date and action in the lab book.
 - j. After 24 h, turn off the heat, let cool.
 - k. Remove the sample, place it in a 1000-ml beaker, and dry in the oven at low heat. Record the date and action in the lab book.
 - l. Discard the 95% ethanol in an appropriate waste bottle.
 - m. Clean soxhlet apparatus fully, rinsing with 95% ethanol.
5. Place the beaker on a hot plate, add *ca.* 500 ml of distilled water, cover with a watch glass and bring the sample to a boil for 6 h. Add distilled water as needed. Record the date and action in the lab book.
6. Remove from heat and decant distilled water from the sample. If unable to continue the treatment at this time, dry the sample in the oven and store. Record the date and action in the lab book.
7. In a 1000-ml beaker, add 500 ml of distilled water. To this add several drops of phosphoric acid, and at the same time, add *ca.* 0.5 g of sodium chlorite. Cover and place on a hot plate on low heat.
8. Repeat every 2 h for 8 h and leave on low heat overnight.
9. Remove from heat and uncover the beaker. If the sample is not paper white, repeat steps 7 and 8. Decant the liquid and rinse the sample 6 times with 1000 ml of distilled water. Dry the sample, record the date, weight and action and store the sample for combustion.

The samples were synthesized to benzene following the procedures outlined in Witkin *et al.* (1993) and Long and Kalin (1992), in which specific steps were determined to ensure the purity and reproducibility of synthesized benzene. Two Wallac Quantulus LS counters were used to measure the ^{14}C activity of the synthesized benzene. The vial selection, counting windows and manual high-voltage settings on these two counters were determined based on the results published by Pearson (1979,

1983) and McCormac (1992). The counters were modified at the Arizona laboratory to allow for manual adjustment of the high voltage applied to each photomultiplier tube. State-of-the-art equipment and the underground lab reduced our need to apply corrections originally used by Pearson (1983) with older LSC equipment (e.g., atmospheric pressure, diurnal effect and sample evaporation), and corrections for benzene purity (after McCormac 1992) are very small due to steps taken during benzene synthesis to ensure purity (Witkin 1992). The LS counters are housed in a stable underground counting laboratory (Kalin and Long 1989). Samples of Oxalic Acid I, background benzene and sample benzene are counted sufficiently long to attain better than $\pm 0.2\%$ precision (10 k min minimum for samples).

RESULTS AND DISCUSSION

The results of this intercomparison exercise are listed in Table 1 (Irish oak) and in Table 2 (*Sequoiadendron*). Long (1990) presented the consensus QA protocol for ^{14}C dating laboratories in which total analytical precision (TAP) was defined to be the total uncertainty applied to each laboratory result. Stuiver and Pearson (1986) showed that, for high-precision data, a laboratory multiplier (K) should be applied to results. The value of K represents uncertainty in the precision of results above that predicted by normal statistics. Therefore, the TAP presented by Long (1990) is the product of the laboratory error multiplier K and the counting statistics.

TABLE 1. Intercomparison of the First University of Arizona High-Precision ^{14}C LSC Analyses on Bidecadal Samples of Irish Oak

Sample site	Bidecadal center year	Pearson and Qua (1993)		Pearson <i>et al.</i> (1986)		Belfast University (this study)		Arizona (this study)	
		$\Delta^{14}\text{C}$ (‰)	$\delta^{13}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)	$\delta^{13}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)	$\delta^{13}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)	$\delta^{13}\text{C}$ (‰)
Blackwater	3450 BC	73.2 ± 2.3	N.A.*	75.0 ± 2.1	N.A.	77.6 ± 2.1	-25.7	77.3 ± 1.8	-24.4
Blackwater	3470 BC	75.5 ± 2.5	N.A.	78.0 ± 2.5	N.A.	77.2 ± 1.8	N.A.	81.5 ± 1.6	-24.5
Motorway	2490 BC	42.4 ± 1.7	N.A.	43.0 ± 1.7	N.A.	44.2 ± 2.1	-26.2	42.5 ± 1.4	-26.1
Garry Bog	390 BC	-2.6 ± 1.9	N.A.	-0.8 ± 1.9	N.A.	N.A.	N.A.	-0.5 ± 1.5	-25.3
Garry Bog	370 BC	1.8 ± 1.4	N.A.	3.9 ± 1.4	N.A.	1.2 ± 2.0	-26.1	6.5 ± 1.1	-24.8

*Not available

TABLE 2. Intercomparison of averaged single-year* high-precision ^{14}C LSC analyses with decadal samples (Stuiver and Becker 1993). Arizona samples are *Sequoiadendron*, California. Stuiver and Becker (1986, 1993) samples are Douglas fir, Vancouver Island, British Columbia. Pearson *et al.* (1986) and Pearson and Qua (1993) samples are *Quercus* spp. from the British Isles.

Decadal center year	Arizona (this study)		Pearson <i>et al.</i> (1986)	Stuiver and Becker (1986)	Stuiver and Becker (1993)	Pearson and Qua (1993)
	$\Delta^{14}\text{C}$ (‰)	$\delta^{13}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)
AD 1085	-10.5 ± 2.3	-20.5 ± 0.3		-8.6 ± 1.4	-10.8 ± 1.5	
AD 1095	-15.9 ± 2.8	-19.9 ± 0.3	-10.0 ± 1.4	-12.3 ± 2.0	-15.5 ± 2.1	-11.6 ± 1.4
AD 1105	-17.2 ± 1.4	-20.0 ± 0.4		-14.6 ± 1.8	-17.0 ± 1.9	
AD 1115	-17.8 ± 2.7	-19.8 ± 0.2	-13.3 ± 1.5	-15.7 ± 2.0	-17.9 ± 2.1	-14.8 ± 1.5
Avg. value	-15.35		-11.65	-12.80	-15.30	-13.20

*Arizona dates are the average of separate analyses on individual rings with standard deviation (σ). We did not divide σ by \sqrt{n} to obtain the standard deviation of the mean due to small but real variation between annual rings.

The data in Table 1 show that the initial high-precision results obtained by Arizona vary more than counting statistics alone would suggest. The calculated bias among these initial values (Table 1) and the results of Pearson *et al.* (1986) is 1.6‰. The offset of these data with Pearson and Qua (1993) is 3.4‰. The calculated bias between the new Belfast data (Table 1) and the results of Pearson *et al.* (1986) is 0.08‰. The offset of these data with Pearson and Qua (1993) is 1.8‰. It is interesting to note that, when compared to the mean of all three results, the mean bias of the published results of Pearson and Qua (1993) is -1.7‰, the mean bias of the new Belfast data is +1.5‰ and the mean bias of the Arizona data is +1.7‰ (Table 3). The nature of this bias changes substantially when the data published by Pearson *et al.* (1986) is used in the intercomparison. Here the mean bias of the published results of Pearson *et al.* (1986) is -0.4‰, the mean bias of the new Belfast data is -0.7‰ and the mean bias of the Arizona data is +1.0‰ (Table 3). This intercomparison would suggest that new analyses at the Belfast ¹⁴C lab are nearly identical to the results obtained in the same laboratory a decade ago, and that there may be a +1.8‰ offset between the Arizona and Belfast ¹⁴C laboratories. The data presented in Table 1 represent the first five high-precision LSC ¹⁴C analyses performed at the Arizona laboratory. Therefore, given the sparse data set, no exhaustive statistical treatment of the data in Table 1 is possible.

TABLE 3. Laboratory Bias (Offset) from Mean of Three High-Precision Results

	Bias (‰) 1993 average	Bias(‰) 1986 average
Pearson and Qua (1993)	-1.7	--
Arizona	+1.7	+1.1
Belfast	+1.5	-0.7*
Pearson <i>et al.</i> (1986)	--	-0.4

*Intercomparison of the new Arizona and Belfast data show little offset with the results of Pearson *et al.* (1986). Intercomparison of the new Arizona and Belfast data show significant offset with the revised ¹⁴C calibration data of Pearson and Qua (1993).

The intercomparison of routine high-precision results (Table 2) with published results of Stuiver and Becker (1986, 1993), Pearson *et al.* (1986) and Pearson and Qua (1993) may also be used to investigate routine laboratory bias at Arizona. Table 2 presents the decadal average of single-year samples analyzed at Arizona to study solar variability during the Medieval Warm period (provided by P. E. Damon, personal communication). These data compare favorably with the published data of Stuiver and Becker (1993) showing an average bias of only 0.05‰. Although the samples are not identical, they are from the same geographical region, thus spatial differences in atmospheric ¹⁴C (Jirikowic and Kalin 1993) probably do not affect the comparison of these two data sets. However, the data show a bias of 2.55‰ with the uncorrected data of Stuiver and Becker (1986). This weakens the case for a favorable comparison between the Seattle data and the Arizona data unless we endorse the corrected data of Stuiver and Becker (1993).

Comparison of the *Sequoiadendron* ¹⁴C results with published results on bidecadal oak from the British Isles for the same time period show a bias of 3.70‰ with the data of Pearson *et al.* (1986) and a bias of 2.15‰ with the data of Pearson and Qua (1993). The intercomparison of the new Arizona and Belfast data (Table 1) suggest a possible bias of +1.8‰. The nature of the divergence of the *Sequoiadendron* results and the oak results could represent evidence for a geographical effect or may represent a true bias between results of the two laboratories. There is clear evidence for a sys-

tematic offset between the data presented in Table 2, but it is difficult to endorse one data set (1986 or 1993) as offering the best intercomparison of results.

CONCLUSION

The results of intercomparison between published data (Pearson and Qua 1993) and new high-precision ^{14}C measurements from the University of Arizona and the Queen's University of Belfast ^{14}C laboratories show the data as originally published by Pearson *et al.* (1986) compare more favorably than the "revised" 1993 data. Statistically, no offset can be determined between new results at Belfast and those produced a decade before. Routine high-precision analyses at the University of Arizona on single-year *Sequoiadendron* samples have resulted in decadal averages that compare remarkably well with the published decadal results of Stuiver and Becker (1993), but do show bias with the uncorrected results of Stuiver and Becker (1986) supporting the use of the 1993 data. There is a systematic offset between the new Arizona and Belfast oak results and between the Arizona *Sequoiadendron* and published Belfast oak data (Pearson *et al.* 1986; Pearson and Qua 1993).

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COMPARING LONG-TERM ATMOSPHERIC ^{14}C AND ^3H RECORDS NEAR GRONINGEN, THE NETHERLANDS WITH FRUHHOLMEN, NORWAY AND IZAÑA, CANARY ISLANDS ^{14}C STATIONS¹

HARRO A. J. MEIJER, JOHANNES VAN DER PLICHT

Centrum voor Isotopen Onderzoek, University of Groningen, NL-9747 AG Groningen
The Netherlands

JORUNN S. GISLEFOSS and REIDAR NYDAL

Radiological Dating Laboratory, NTH, N-7034 Trondheim, Norway

ABSTRACT. We present the results of the $^{14}\text{CO}_2$ atmospheric monitoring station at the Smilde observation station, near Groningen, the Netherlands, a typical continental station. We compared these data, for absolute values and annual variation, with data from Fruholmen, Nordkapp, Norway and Izaña, Tenerife, Canary Islands, which are situated in areas less influenced by fossil-fuel CO_2 . The 20-yr Smilde record shows much seasonal variation (peak-to-trough variation is $\approx 30\%$ in contrast to $\approx 12\%$ for Fruholmen, and $\approx 5\%$ for Izaña) and a lower overall value due to fossil-fuel consumption, in accordance with findings from similar stations in continental Western Europe. The Fruholmen and Izaña data show fairly equal mean $\Delta^{14}\text{C}$ levels, but differ in seasonal amplitude. This difference could be due partly to the elevation difference between the stations. The Izaña station also has a slow exchange with the ground-level air because of an inversion layer. It is speculative whether annual injection of ^{14}C from the stratosphere also plays a role. We give the Groningen long record of tritium in precipitation, showing profound seasonality.

INTRODUCTION

The ^{14}C content of atmospheric CO_2 has been monitored in several places around the world for many years, in some cases dating back to the beginning of nuclear bomb testing (Münnich and Vogel 1958; Nydal and Lövseth 1983; Levin, Münnich and Weiss 1980; Vogel 1970; Manning *et al.* 1990). Most of these monitoring sites have been chosen for minimal direct continental biospheric and anthropogenic influences. These measurement series have shown their usefulness in supporting modeling of northern-southern hemisphere mixing in the atmosphere, as well as atmosphere-ocean CO_2 exchange rates (Nydal 1968; Broecker *et al.* 1985).

True continental stations, on the other hand, might shed more light on the (spatial and temporal) distribution of fossil-fuel emissions, mainly by providing information about summer vs. winter ^{14}C levels in comparison with “nearby” background sampling stations (Levin *et al.* 1989). Surprisingly little is known about the summer/winter difference in fossil-fuel consumption on the different continents. Here, we compare the continental Smilde observation station, Netherlands, to background stations: Fruholmen/Nordkapp, Norway, and Izaña/Tenerife, Canary Islands, in absolute value, long-term trends and in annual variation. Data from another station, Schauinsland, in the Black Forest region of Germany, which could be characterized as “continental background” (Levin, Graul and Trivett 1994), are also given for comparison.

METHOD

The Smilde station is a television broadcasting tower, *ca.* 40 km south of Groningen ($52^\circ 54'\text{N}$, $6^\circ 24'\text{E}$). Sample collection methods are similar to those described by Vogel (1970). All samples are collected from a height of ≈ 80 m above the ground (≈ 80 m above sea level), by gently bubbling air

¹This paper was presented as a poster at the 15th International ^{14}C Conference, Glasgow, Scotland, 15–19 August 1994.

through an NaOH absorption system. Direct contamination of the surroundings is considered to be small. The NaOH solution is (nearly always) replaced once a month, so that each measuring point is a true average of a 1-month period. The system is designed to absorb all CO₂ that passes through it.

In the laboratory, the CO₂ is liberated from the solution again by acidification and stripping, and the collected CO₂ is inserted into a proportional gas counter. Typical counting statistics allow for a precision of ~5‰. The δ¹³C values obtained for the CO₂ resemble the true isotope ratio in the atmospheric CO₂, although with considerable scatter for the period between 1974–1985. After 1985, no 100% stripping yield was reached. This effect gradually became worse until we discovered it to be the cause of the decreasing δ¹³C values. Since January 1993, recollection has occurred with 100% efficiency, and reliable δ¹³C values are being produced. Of course, this effect has no influence on the normalized ¹⁴C values.

Nydal (1966) previously described the Fruholmen and Izaña stations and the preparation technique. The main differences from the Smilde station are that the points are averages over a few days (7 days from 1962–1981, and 3–4 days later), and that considerable isotope fractionation occurs (leading to values of ~-25‰ for δ¹³C) due to using NaOH solution exposed to open air. The counting statistics for each Δ¹⁴C are *ca.* 10‰ from 1962–1981, and 5‰ thereafter. Of course, the shorter collection time span yields a greater number of points per year than the Smilde series.

RESULTS

Trends

Figure 1 is an overview of the data discussed in this paper. The ¹⁴C results are given in excess per mil relative to 1950 standard air, corrected for isotopic fractionation and radioactive decay, where

$${}^{14}\Delta = e^{-\left(F_{\text{modern}}/8033\right)} e^{\left(\left(1950 - \text{date}\right) / 8267\right)} - 1 \left(\times 1000\text{‰}\right)$$

(Stuiver and Polach 1977).²

The Smilde station began operation in the early 1960s, stopped in 1964, and resumed in 1974. Vogel (1970) published the early Smilde series. Nydal and Lövseth (1983) published the Izaña and Fruholmen series in both tables and graphs until 1980. Only graphs were published later on (*e.g.*, Nydal 1991; Gislefoss 1994). We give the Smilde series in Appendix 1. Tables of all data from the Trondheim laboratory will be available online in a Carbon Dioxide Information Analysis Center (CDIAC) database.

The curves plotted in Figure 1 are “weighted smooth fits”, also called “Loess fits” (Cleveland and McRae 1989). The smoothing factor of this fit procedure can be set so that the time response of the fit procedure is shorter than a year, thereby allowing the curve to follow slow, trend-like changes, but preventing it from following the intra-annual variations. The Fruholmen trend fit has been performed since July 1963. Because of its long duration, the trend yields important information about the troposphere-ocean exchange process. The main feature of the curve is the exponential decay of the atmospheric value caused primarily by uptake of ¹⁴C into the oceanic mixed layer (Nydal *et al.* 1984). Several other effects contribute to the shape of this exponential decay curve, however. At first, the disequilibrium between the northern and southern hemisphere during the 1960s (Nydal

² As it is given here, the Fruholmen signal has been corrected for radioactive decay. This is in contrast to other publications of these data. The differences, however, are small (6‰ in absolute value in 1992), and are most likely negligible.

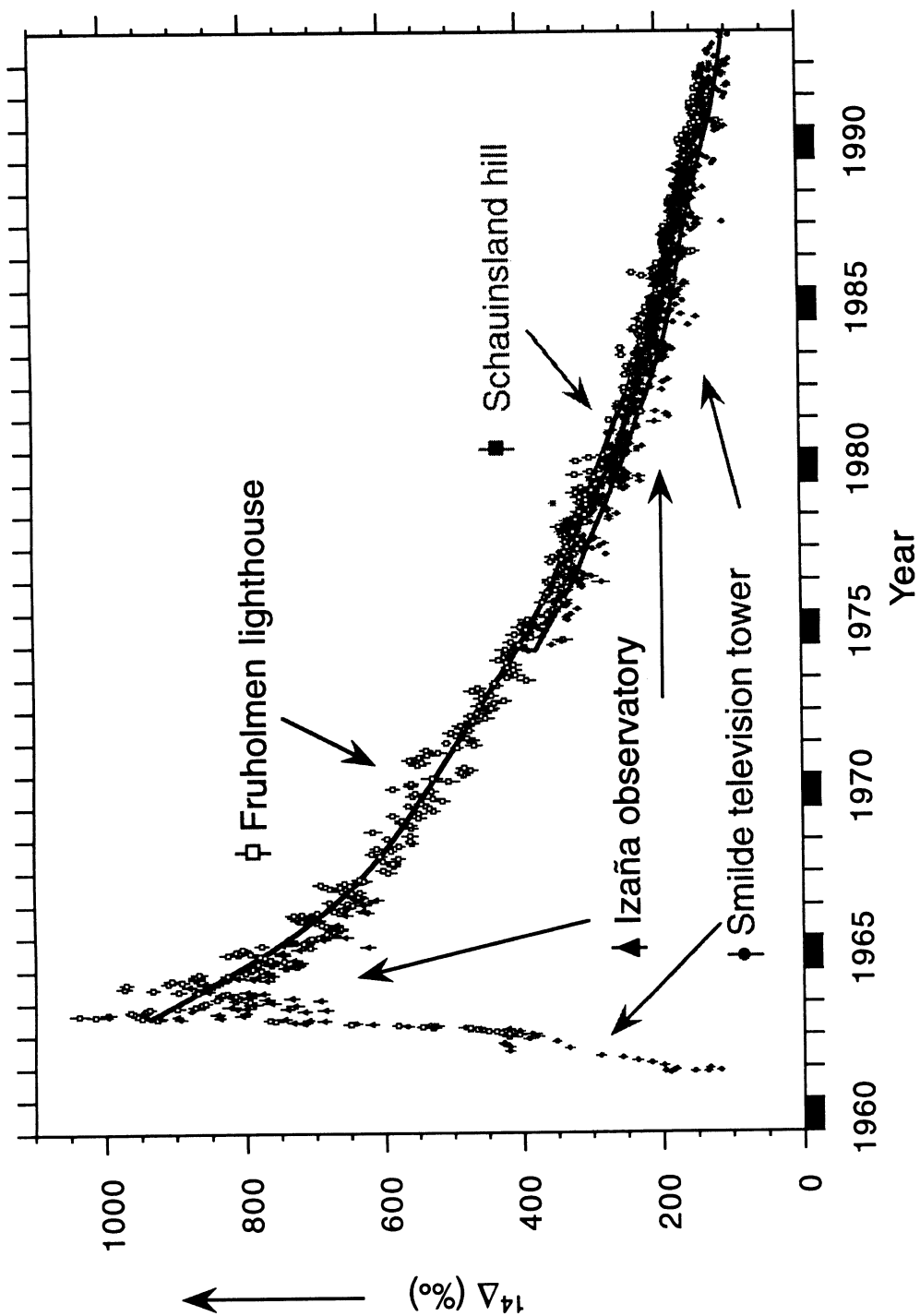


Fig. 1. Overview of our $\Delta^{14}\text{C}$ measurement series from four stations: 1) the Smilde television tower close to Groningen, Netherlands ($52^{\circ}54'\text{N}$, $6^{\circ}24'\text{E}$) 80 m asl; 2) the Fruholmen lighthouse, Nordkapp, Norway ($71^{\circ}06'\text{N}$, $23^{\circ}59'\text{E}$) 70 m asl; the Izaña observatory, Tenerife, Canary Islands, ($28^{\circ}22'\text{N}$, $16^{\circ}03'\text{W}$) 2376 m asl; and 4) the Schauinsland hill station, Black Forest, Germany (48°N , 8°E) 1205 m asl, data from Levin, Graull and Trivett (1994). The lines are so-called "weighted smooth" or Loess trend fits to the points.

1968) is the main cause for the relatively high decay during that period (characterized by a half-life of *ca.* 5 yr from 1963–1968 vs. *ca.* 8 yr thereafter). Then, atmospheric nuclear bomb tests by France (mainly 1966–1968) and China (mainly 1968–1972) led to smaller disturbances, as well as ^{14}C produced by nuclear power plants. At present, a significant part of the annual decrease (*ca.* 4‰ yr $^{-1}$) is due to the Suess effect (Tans 1978; Levin *et al.* 1989).

Considering all these effects, we believe the best way to understand the shape of the observed trend is modeling the abovementioned effects, combined with fit results. Examples of such efforts can be found in (Nydal 1968) for the period 1962–1967, or a more recent global approach (Hesshaimer, Heimann and Levin 1994).

Seasonality

We fitted the detrended (measurements minus the trend fit) Smilde signal with a single harmonic curve and treated the Schauinsland data in the same manner. In the case of the Fruholmen series, two curve fits were applied: one was similar to Smilde, for the period starting May 1977; the other was an exponentially damped harmonic function, fitted to all the data beginning July 1963. The seasonal fits are shown in Figure 2, whereas the fit results are shown in Table 1.

TABLE 1A. Characteristics of the fitted seasonal functions (single harmonic $A\cos(2\pi t/T + \varphi) + B$). For the Fruholmen curve, only the data since May 1977 were used. The Schauinsland curve (Levin, Graul and Trivett 1994) was treated similarly to the others, and is given for comparison.

Station	Peak-to-trough	Maximum	Residual standard deviation (‰)	Average measurement uncertainty (‰)
Fruholmen	12.4 ± 1.6	Aug. 29 \pm 7 d	10.0	6.0
Izaña	5.4 ± 2.0	July 14 \pm 20 d	7.6	5.0
Smilde	31.0 ± 3.0	July 11 \pm 5 d	11.6	6.0
Schauinsland	12.0 ± 1.2	Aug. 28 \pm 5 d	8.3	4.3

TABLE 1B. The full Fruholmen curve has been fitted here, to an exponentially damped harmonic curve ($A\cos(2\pi t/T + \varphi) + B$) $\times e^{-Ct/T} + D$. The damping characteristic time gives a good indication of the stratosphere-troposphere interaction time. In A and B, the remaining undamped part of the full fit curve is the same within the error bars as the single harmonic fit to the “damped” part of the Fruholmen data, both in phase and amplitude.

Station	Peak to trough (‰) (fully damped)	Maximum	Damping $t_{1/2}$	Residual standard deviation (‰)	Average measurement uncertainty (‰)
Fruholmen	12.4 ± 2.0	Aug 23 \pm 3 d	2.1 \pm 0.3 yr	16.2	8

The Fruholmen seasonality signal is important because it contains a record of very high amplitudes from the period of clear disequilibrium between stratosphere and troposphere, as well as the later record of stabilized amplitudes. Comparing Tables 1A and 1B, one observes that the remaining undamped part of the full-fit curve is the same within the error bars as the single harmonic fit to the “damped” part of the Fruholmen data. This suggests that a considerable part of the remaining seasonality in the signal is still due to stratospheric input. On the other hand, the Fruholmen seasonality resembles the Schauinsland signal in both phase and amplitude. This station, at an elevation of *ca.* 1200 m in the Black Forest region of southwest Germany, has been affected by anthropogenically induced emission (Levin *et al.* 1989), which causes the major part of its seasonality.

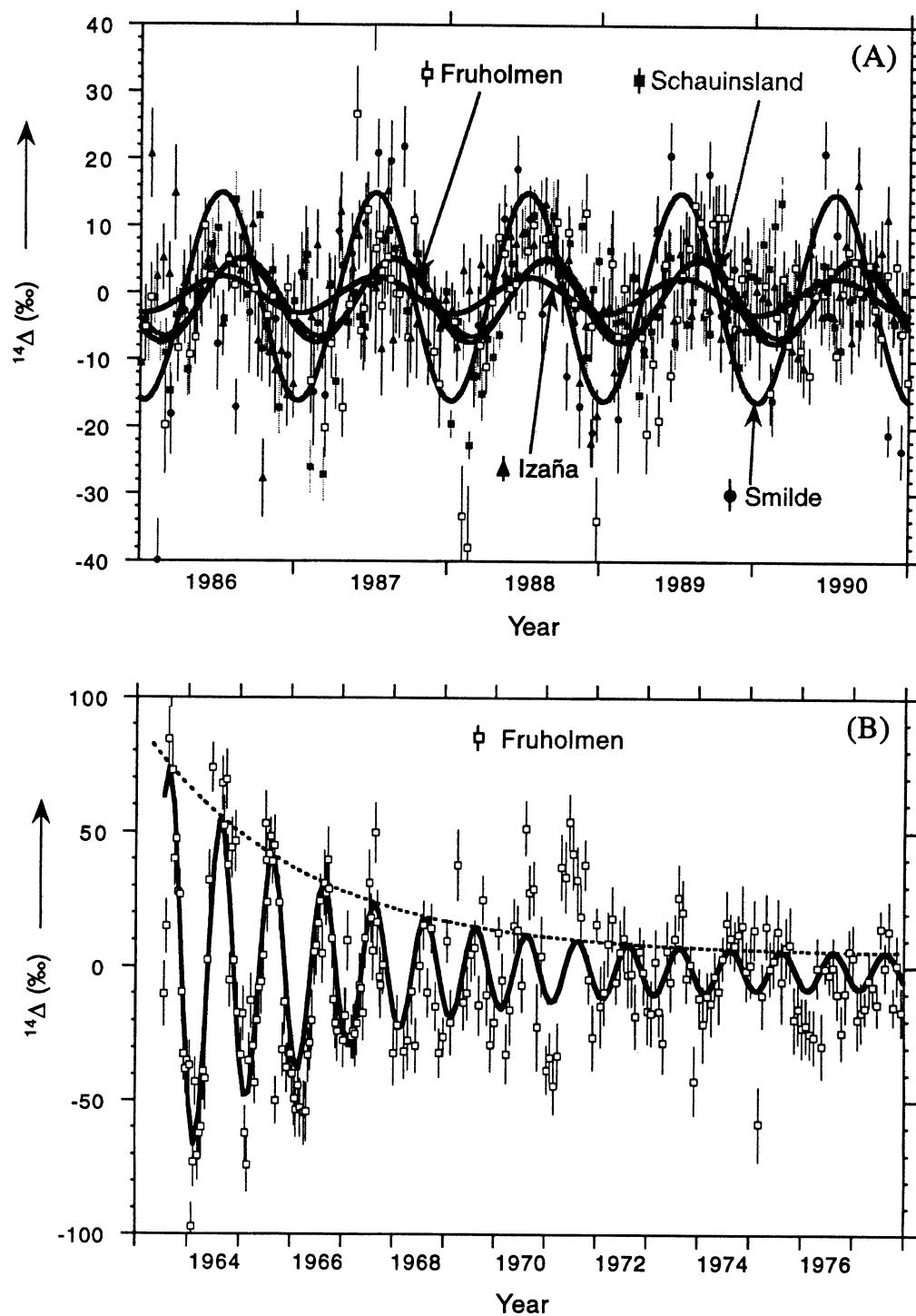


Fig 2. A. The detrended (measurements minus the trend fit) measurement series, fitted with a single harmonic curve, since May 1977. Shown is the part for the years 1986–1991. B. The detrended Fruholmen series, fitted with an exponentially damped harmonic function, fitted to all the data since July 1963. Shown is the part for the years 1963–1977.

The continental Smilde record shows relatively high seasonality, obviously caused by the summer-winter difference in fossil-fuel emissions (combined with the seasonal variation in average mixing layer thickness). The Smilde oscillation is somewhat out of phase with that of Fruholmen; its minimum at mid-January coincides with the coldest part of winter in northwestern Europe.

The comparison in Table 1A of the residual standard deviation to the average measurement uncertainty in the respective series shows that all series suffer from random fluctuations, most probably correlated with meteorological circumstances. Furthermore, the seasonality of the signal shows irregularities on a year-to-year basis.

Absolute Values

All the stations discussed here were operative during the period 1980–1990, which makes this period the most useful for comparison of absolute values. Figure 3 shows the weighted smooth fits of all four stations for this period. Here the time response of the fits allows the fit curves to follow intra-annual variations. To show the difference clearly, we subtracted an exponentially decaying curve with a half-life of 8 yr from all curves. This procedure should not be taken to represent a “best fit” of the measurements.

As expected, the Smilde values are generally lower than the Fruholmen values. In summer, however, the difference is much smaller than in winter. For the three background stations, the differences between Fruholmen on the one hand, and Izaña and Schauinsland, on the other, seem to decrease

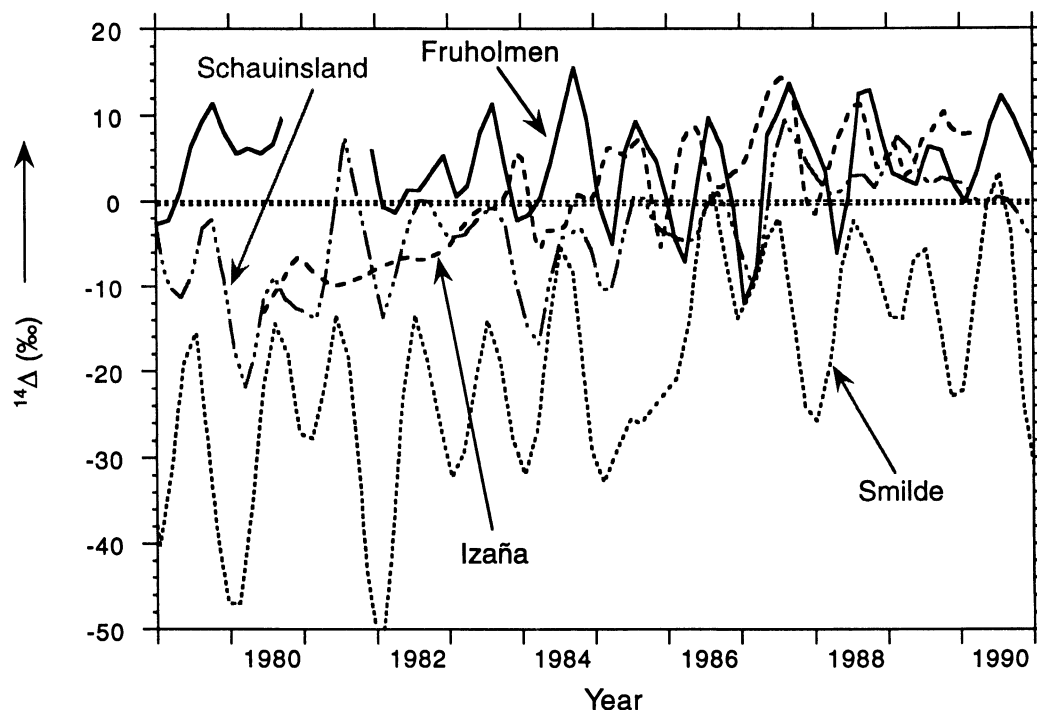


Fig. 3. Weighted smooth fits of all four stations between 1979 and 1990. The time response of the fits allowed the fit curves to follow intra-annual variations. To make the differences in absolute values more clearly visible we subtracted an exponentially decaying curve with a half-life of 8 yr. The zero on the y-axis is arbitrary.

between 1980 and 1990. It should be noted that these are relatively small effects, and that all measurement series show a considerable residual standard deviation (compare Table 1).

TRITIUM IN PRECIPITATION

Tritium provides additional information about the stratosphere-troposphere interaction. The Groningen laboratory has measured ^3H in precipitation continuously since 1972.

Precipitation water is collected monthly, reduced over hot magnesium, and its hydrogen transmuted into ethene. This acts as a counting gas in a proportional counter, belonging to the same low-activity setup as the CO_2 counters for ^{14}C . The data are available through the IAEA (1994). The activity is expressed in tritium units (TU). One TU is equivalent to a ^3H activity of $0.118 \text{ Bq liter}^{-1} \text{ H}_2\text{O}$. Activity and monthly amount of precipitation must then be combined to produce the monthly ^3H activity from precipitation per square meter. Figure 4 gives an overview of these results. We obtained average TU by dividing the activity numbers by the average monthly precipitation (62 mm/month), thus circumventing the convolution of the ^3H deposit with the seasonality of the amount of precipitation.

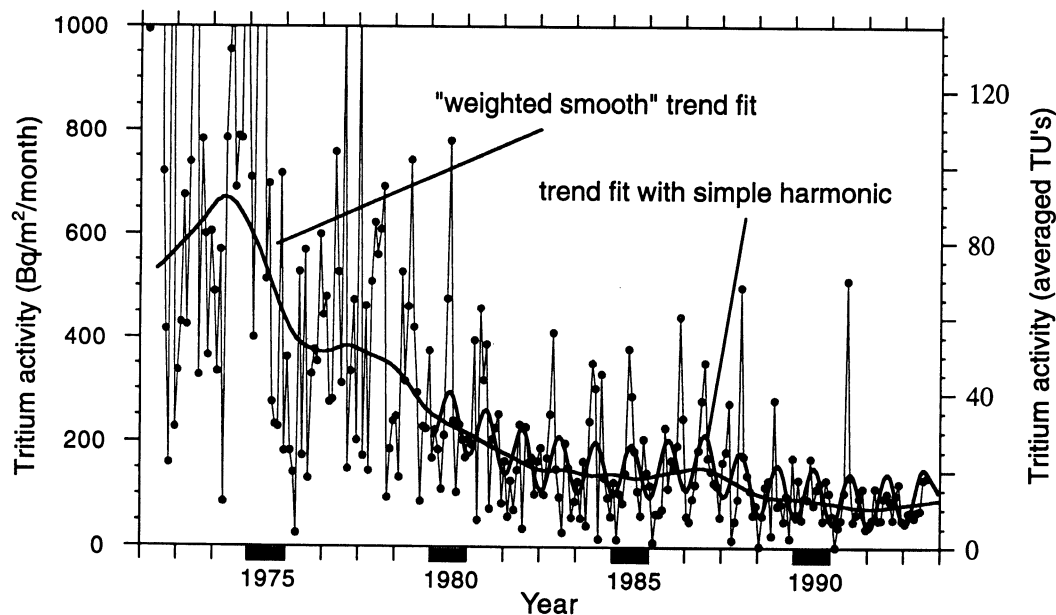


Fig. 4. The ^3H activity in precipitation in Groningen ($52^{\circ}15'\text{N}$, $6^{\circ}33'\text{E}$) 0 m asl. On the left-hand y-axis, the activity is expressed in Bq m^{-2} per month. These numbers are obtained by multiplying the measured activity of the precipitation (in TU or $0.118 \text{ Bq liter}^{-1}$ water) with the monthly amount of precipitation. The right-hand y-axis is expressed in average TU, obtained by dividing the left-hand scale activity numbers by the average monthly precipitation (62 mm/month). Two fits to the data are shown: a weighted smooth trend fit and a "full fit" (from 1981 onwards), consisting of the above trend with a single harmonic.

We show two fits to the curves: 1) a weighted smooth trend fit, showing an irregularly decaying trend. This irregularity is caused by the bomb tests in the 1970s; 2) a "full fit" (since 1981), consisting of the above trend with a single harmonic. The peak-to-peak value for the harmonic is $104 \pm 22 \text{ Bq m}^{-2}$ ($14 \pm 3 \text{ TU}$); the maximum occurs mid-July. A fit to the "direct" TU values yields the same amplitude within the uncertainty bars, but the maximum occurs mid-June.

In recent years the values generally move between $\sim 40 \text{ Bq m}^{-2}$ (5 TU) in winter and 140 Bq m^{-2} (20 TU) in summer. The average value is still decreasing, although in recent years at a slower rate. The amplitude of the harmonic seems to be constant over the last 10 yr.

DISCUSSION

Levin *et al.* (1989) have studied the influence of fossil fuel in the Schauinsland measurements by comparing the Schauinsland values to those of background station Jungfrauoch. Assuming that the Jungfrauoch series is fairly clean, the authors found 15‰ in winter, and 5‰ in summer. The average difference between the Smilde and Schauinsland full fits is 27‰ in winter and 4‰ in summer. Combining these numbers with the Levin *et al.* (1989) data, and taking into account the phase difference between the two stations, we conclude that fossil-fuel contributions for the Smilde series are *ca.* 40‰ or 14 ppm in winter, and 9‰ or 3 ppm in summer.

These computations cannot be applied to the Fruholmen and Izaña stations, because they are too far away. The differences in absolute values between the curves change considerably between 1979 and 1990, as indicated above. Using only the first 5 yr of Figure 3, one can conclude that Fruholmen is almost undisturbed, with higher values in winter than Schauinsland shows in summer. On the other hand, if one uses the amplitude of the seasonal cycle as an indicator of fossil-fuel influence, it would imply that Fruholmen also suffers from fossil-fuel influences in winter, of about the same size as Schauinsland. Absolute values in the Izaña curve from 1979–1984 indicate that the station is quite affected by fossil-fuel emissions throughout the year, whereas the small amplitude of the seasonal cycle indicates an undisturbed background station.

Clearly, the situation is more complicated. Apart from possible experimental problems (*e.g.*, sample site changes, pretreatment uncertainties, calibration problems), which only comparison with data of other northern hemisphere stations can elucidate, other factors such as enhanced (seasonal) stratospheric input at higher latitudes, or influences by the oceans must also play a role. Furthermore, it is quite conceivable that the part of the terrestrial biosphere (typically soils) that interacts slowly with the troposphere presently acts as a net ^{14}C source with a summer-winter periodicity (Hesshaimer, personal communication 1995). The main point of discussion is the amplitude of the natural seasonal effect, caused by the annual “injection” of ^{14}C from the stratosphere into the troposphere (Tans 1978). The periodicity of this effect is clearly illustrated in the Fruholmen series between 1963 and 1970, where a clear stratosphere-troposphere disequilibrium existed. Presently, at least in Europe, the periodicity of fossil-fuel emission seems to be the dominant cause of seasonality, even in such remote places as Fruholmen.

^3H is much more influenced by nuclear bomb testing than is ^{14}C . In summer 1963, the average precipitation activity was *ca.* 3800 TU (230 kBq m^{-2}), or *ca.* 700 times the pre-bomb activity (IAEA 1979). The fast decay of this activity is caused both by the short half-life of ^3H ($t_{1/2} = 12.3 \text{ yr}$), and the huge existing reservoir with short interaction times. The ocean mixing layer (taken here as 130 m) contains *ca.* $5 \times 10^7 \text{ GT}$ water compared to 13,000 GT water vapor in the atmosphere, resulting in a rapid 1:4000 dilution, whereas in the case of $^{14}\text{CO}_2$, the ratio atmosphere:(ocean mixing layer + land biosphere) is only *ca.* 1:2 (Schlesinger 1991, and references therein). A strong seasonal signal is clearly visible, although the scatter in the signal is considerable. This scatter is caused by the irregular character of rainfall.

Because of the high mixing rate, together with the short decay time, it is probable that at least the seasonal cycle observed in ^3H is close to its natural value. On the other hand, soil and groundwater reservoirs can be found with relatively high ^3H content, and, especially in summer, it could well be

that soil water evaporation acts as a net ^3H source, thereby enhancing the natural seasonality. It is most likely that considerable natural seasonality exists. Stratosphere-troposphere interaction has a pulsed character, whereas the troposphere mixing is very fast for water (the annual amount of precipitation is *ca.* 40 times as high as the water-vapor content of the troposphere (Schlesinger 1991)).

In the case of steady state, stratospheric production and stratosphere-troposphere exchange of cosmogenic isotopes must balance the loss of these isotopes in the troposphere caused by decay. Using as input the amounts of water for the oceanic mixing layer and total precipitation mentioned above, it is simple to compute that the measured seasonality is able to maintain a (mixing layer + troposphere) ^3H background level of *ca.* 1.5 TU. Unfortunately, we know of no reliable data for seasonality in precipitation and upper ocean natural ^3H values preceding the bomb tests.

For ^{14}C , a similar simple computation (using the fact that ^{14}C decays by 0.125‰ per year, and that the troposphere contains 1/55 of the total global reservoirs (Schlesinger 1991)) leads to the conclusion that a replenishment of 7‰ is needed for steady-state conditions. If we assume that the troposphere gets this total “refill” in a pulse-like manner (which is illustrated in the early Fruholmen measurements), the natural summer-winter difference cannot be more than this 7‰ (and is most likely considerably lower). This shows that, from the measurement series presented here, the seasonality of only the Izaña record can be totally natural.

A new air monitoring station close to Smilde (Kollumerwaard), where the Groningen group recently started $^{14}\text{CO}_2$ (as well as stable isotope) measurements with high temporal resolution (so-called “event-trapping”) could shed more light on the amount of fossil-fuel CO_2 admixtures and their seasonality (Zondervan and Meijer 1995).

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APPENDIX 1. SMILDE ^{14}C DATE LIST

Month	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ unc. (‰)	$\delta^{13}\text{C}$ (‰ VPDB)
Nov. 1974	389	9	- 8.55
Dec. 1974	393	10	- 8.48
Jan. 1975	356	6	- 8.51
Feb. 1975	342	7	- 9
Mar. 1975	391	9	-10.15
Apr. 1975	370	6	- 8.4
May. 1975	389	6	- 8.46
Jun. 1975	368	6	- 7.47
Jul. 1975	376	6	- 8.46
Aug. 1975	382	5	- 8.66
Sep. 1975	365	6	- 8.53
Oct. 1975	333	6	- 9.18
Nov. 1975	335	6	- 8.87
Dec. 1975	328	6	- 8.73
Jan. 1976	350	6	- 8.51
Feb. 1976	318	6	- 9.32
Mar. 1976	336	6	- 9.09
Apr. 1976	332	6	- 9.08
May. 1976	353	6	- 8.38
Jun. 1976	345	6	- 7.89
Jul. 1976	342	6	- 8.27
Aug. 1976	342	6	- 7.55
Sep. 1976	330	6	- 7.93
Oct. 1976	312	6	- 8.22
Nov. 1976	312	6	- 8.91
Dec. 1976	284	13	- 9.54
Jan. 1977	311	10	- 9.15
Feb. 1977	298	10	- 8.95
Mar. 1977	310	10	- 9
Apr. 1977	320	10	- 8.7
May. 1977	321	6	- 8.88
Jun. 1977	335	6	- 8.07
Jul. 1977	336	6	- 7.23
Aug. 1977	331	6	- 7.67
Sep. 1977	325	6	-13.08
Oct. 1977	277	6	- 9.48
Nov. 1977	310	6	- 8.39
Dec. 1977	296	5	- 9.86
Jan. 1978	286	4	- 9.53
Feb. 1978	279	7	- 9.67
Mar. 1978	298	6	- 8.63

Month	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ unc. (‰)	$\delta^{13}\text{C}$ (‰ VPDB)
Apr. 1978	292	9	- 9.54
May. 1978	309	4	- 9.8
Jun. 1978	331	7	- 8.08
Jul./Aug. 1978	334	6	- 7.75
Sep. 1978	330	6	- 8.07
Oct. 1978	298	6	- 8.32
Nov. 1978	272	6	- 9.11
Dec. 1978	271	6	- 8.9
Jan. 1979	260	6	- 9.55
Feb. 1979	274	6	-10.15
Mar. 1979	274	8	- 8.6
Apr. 1979	280	6	- 8.92
May. 1979	310	9	- 8.47
Jun. 1979	298	6	- 8.87
Jul. 1979	285	6	- 8.14
Aug. 1979	280	5	- 7.99
Oct. 1979	250	6	- 8.88
Nov. 1979	246	7	- 9.32
Dec. 1979	255	5	- 9.25
Jan. 1980	226	6	- 9.1
Feb. 1980	244	7	- 9.07
Mar. 1980	228	9	- 9.13
Apr. 1980	244	7	- 9.09
May. 1980	265	12	- 8.98
Jun. 1980	262	6	- 9.48
Jul. 1980	267	6	- 9.05
Aug. 1980	261	5	- 8.17
Sep. 1980	267	6	- 8.51
Oct. 1980	268	6	- 7.55
Nov. 1980	246	5	- 7.5
Dec. 1980	244	6	- 8.42
Feb. 1981	224	6	- 7.03
Mar. 1981	240	6	- 8.08
Apr. 1981	257	4	- 6.99
May. 1981	243	4	- 6.64
Jun. 1981	251	4	- 8.29
Jul. 1981	251	5	- 8.73
Aug. 1981	265	6	- 7.38
Sep. 1981	229	5	- 8.02
Oct. 1981	232	6	- 8.82
Nov. 1981	205	10	- 8.52

Month	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ unc. (‰)	$\delta^{13}\text{C}$ (‰ VPDB)
Jan. 1982	185	6	-10.71
Feb. 1982	188	6	- 9.02
Mar. 1982	204	6	-10.24
Apr. 1982	243	12	- 7.7
May. 1982	235	6	- 8.57
Jul. 1982	227	6	- 7.44
Aug. 1982	238	6	- 8
Sep. 1982	220	5	- 8.87
Oct. 1982	220	5	- 8.84
Nov. 1982	194	6	- 6.34
Dec. 1982	210	6	- 8.5
Jan. 1983	212	5	- 8.69
Feb. 1983	193	6	- 8.15
Mar. 1983	186	6	- 8.06
Apr. 1983	207	6	- 8.88
May. 1983	215	5	- 8.27
Jun. 1983	208	6	- 8.05
Jul. 1983	225	6	- 8.29
Aug. 1983	217	6	-10.72
Sep. 1983	208	6	-10.48
Oct. 1983	194	5	- 9.07
Nov. 1983	190	6	- 9.58
Dec. 1983	191	6	- 9.26
Jan. 1984	191	6	- 8.86
Feb. 1984	180	6	- 9.11
Mar. 1984	183	6	- 9.1
Apr. 1984	196	5	- 8.79
May. 1984	200	6	- 8.57
Jun. 1984	210	6	- 8.21
Jul. 1984	214	6	- 8.15
Sep. 1984	202	5	- 7.97
Oct. 1984	204	5	- 8.31
Nov. 1984	156	5	- 9.19
Dec. 1984	177	5	- 9.03
Feb. 1985	145	6	- 9.11
Mar. 1985	166	6	- 9.03
Apr. 1985	181	5	- 8.75
May. 1985	182	7	- 8.14
Jun/Jul. 1985	174	7	-10.15
Aug. 1985	163	4	- 8.51
Sep. 1985	177	9	-10.36
Oct. 1985	171	5	-10.35
Nov. 1985	174	6	- 9.36
Dec. 1985	168	5	-10.49
Jan. 1986	180	6	- 8.81
Feb. 1986	161	8	- 9.47
Mar. 1986	160	6	- 9.81
Apr. 1986	184	5	- 8.88
May. 1986	180	6	- 9.2
Jun. 1986	184	6	- 9.75
Jul. 1986	193	5	-10.13
Aug. 1986	191	6	- 9.62
Sep. 1986	193	6	- 8.55
Oct. 1986	176	6	- 9.92
Nov. 1986	168	6	- 9.51
Dec. 1986	164	6	- 9.2
Mar. 1987	163	6	- 9.59
May. 1987	178	5	-10.14
Jun. 1987	184	5	- 9.36
Jul. 1987	176	5	- 8.77

Month	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ unc. (‰)	$\delta^{13}\text{C}$ (‰ VPDB)
Aug. 1987	161	6	- 8.38
Sep. 1987	172	6	- 9.86
Oct. 1987	151	5	-10.17
Nov. 1987	145	6	-11.55
Dec. 1987	141	5	-10.63
Jan. 1988	106	5	- 9.66
Feb. 1988	141	8	- 9.41
Mar. 1988	151	6	-10.42
Apr. 1988	149	5	- 9.71
May. 1988	166	5	- 9.86
Jun. 1988	176	5	- 9.09
Jul. 1988	157	6	-10.15
Aug. 1988	159	5	- 8.72
Sep. 1988	170	5	- 9.64
Oct. 1988	149	6	-10.43
Nov. 1988	155	5	-11.48
Dec. 1988	155	5	-10.73
Jan. 1989	143	6	-10.82
Feb. 1989	133	5	-11
Mar. 1989	146	5	-10.49
Apr. 1989	150	5	- 9.82
May. 1989	146	5	-10.46
Jun. 1989	166	5	-10.16
Jul. 1989	154	7	-10.46
Aug. 1989	143	5	-10.83
Sep. 1989	146	6	-11.14
Oct. 1989	138	5	-10.71
Nov. 1989	120	3	-11.32
Dec. 1989	116	4	-11.19
Jan. 1990	126	5	-11.58
Feb. 1990	133	5	-11.24
Mar. 1990	147	5	-11.35
Apr. 1990	140	5	-10.34
May. 1990	149	5	-10.54
Jun. 1990	151	5	-11.37
Jul. 1990	155	5	-10.63
Aug. 1990	158	5	-10.25
Sep. 1990	143	5	-10.62
Oct. 1990	133	5	-11.11
Nov. 1990	115	5	-11.04
Dec. 1990 (1)	109	6	-12.1
Dec. 1990 (2)	113	6	-10.16
Jan. 1991	101	5	-11.49
Feb. 1991	115	6	-12.4
Mar. 1991	118	6	-11.08
Apr. 1991	126	5	-10.66
May. 1991	127	5	-11.42
Jun. 1991	125	5	-10.28
Jul. 1991	132	6	-10.65
Aug. 1991	132	6	- 9.8
Sep. 1991	137	5	-10.5
Oct. 1991	120	4	-11
Nov/Dec. 1991	103	4	-11.33
Jan. 1992	100	4	-10.91
Feb. 1992	107	6	-11.14
Mar. 1992	113	6	-11.11
Apr. 1992	98	6	-10.8
May. 1992	96	5	-10.85
Jun. 1992	119	4	-10.71
Jul. 1992	122	6	-10.25

Month	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ unc. (‰)	$\delta^{13}\text{C}$ (‰ VPDB)
Aug. 1992	104	6	- 9.81
Sep. 1992	103	6	-10.4
Oct. 1992	113	6	-10.31
Nov. 1992	107	10	-10.41
Dec. 1992	98	6	-11.41
Jan. 1993	101	4	-10.51
Feb. 1993	95	4	- 9.34
Mar. 1993	114	4	- 8.77
Apr. 1993	123	4	- 8.58
May. 1993	125	4	- 8.56
Jun. 1993	120	4	- 8.61
Jul/Aug. 1993	116	4	- 8.36
Oct. 1993	105	4	- 8.71

Month	$\Delta^{14}\text{C}$ (‰)	$\Delta^{14}\text{C}$ unc. (‰)	$\delta^{13}\text{C}$ (‰ VPDB)
Nov. 1993	95	4	- 9.28
Dec. 1993	108	4	- 8.69
Jan. 1994	78	4	- 8.91
Feb. 1994	74	4	- 9.57
Mar. 1994	108	4	- 8.99
Apr. 1994	103	4	- 9.35
May 1994	105	4	- 8.78
June 1994	112	4	- 8.57
July 1994	106	4	- 8.42
Aug. 1994	111	4	- 8.44
Sep. 1994	105	4	- 8.27
Nov. 1994	101	4	- 8.87
Dec. 1994	100	4	- 8.92

PAPERS FROM THE WORKSHOP ON PAGES CHRONOLOGIES

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THE ^{14}C AGE OF THE ICELANDIC VEDDE ASH: IMPLICATIONS FOR YOUNGER DRYAS MARINE RESERVOIR AGE CORRECTIONS

W. E. N. AUSTIN,¹ EDOUARD BARD,² J. B. HUNT,³ DICK KROON¹ AND J. D. PEACOCK⁴

ABSTRACT. Increased marine ^{14}C reservoir ages from the surface water of the North Atlantic are documented for the Younger Dryas period. We use terrestrial and marine AMS ^{14}C dates from the time of deposition of the Icelandic Vedde Ash to examine the marine ^{14}C reservoir age. This changed from its modern North Atlantic value of *ca.* 400 yr to *ca.* 700 yr during the Younger Dryas climatic event. The increased marine reservoir age has implications for both comparing climatic time series dated by ^{14}C and understanding palaeoceanographic changes that generated the increase.

INTRODUCTION

We discuss here a correction applied to marine radiocarbon ages to compensate for the “marine reservoir age effect”. For the present-day ocean, the sea-surface ^{14}C reservoir age is on the order of 300–400 yr at low latitudes, rising to 1200 yr at higher latitudes in the southern ocean and the North Pacific. High-latitude surface waters are old because of upwelling of subsurface water, whose ^{14}C is not reset to atmospheric values. By contrast, no surface ^{14}C gradient is present between 40° and 70°N in the North Atlantic Ocean. The apparent natural ^{14}C age of these surface waters is almost constant at *ca.* 400 yr. This is linked to the northward advection of surface and thermocline waters from lower latitudes that travel through the Gulf Stream and North Atlantic current systems. Modern marine samples from coastal British waters suggest that the conventional ^{14}C ages obtained are typically too old and that a correction factor of 405 ± 40 yr should be applied (Harkness 1983). This correction factor is generally applied to all conventional marine ^{14}C ages (*eg.*, Stuiver, Pearson and Brazunias 1986; Peacock and Harkness 1990) to facilitate comparison with terrestrial ^{14}C ages.

The study of paired terrestrial and marine samples has demonstrated that the reservoir ages have varied during the Holocene, although it is characterized by a rather stable climate (for the Pacific Ocean, see Southon, Nelson and Vogel (1990); for the Atlantic, see Talma (1990) and Moore, McCormac and McCormick (1994)).

By contrast, the last deglaciation was a period of intense climatic and oceanographic changes that have profoundly affected the global carbon cycle. As evidenced in ice cores and deep-sea and continental sediments, the sizes of the major carbon reservoirs (atmosphere, biosphere and oceans) during the last deglaciation were quite different from their steady-state sizes today. In addition, the rates of exchange between these reservoirs were also probably different because of changes in deep-sea ventilation, wind speed and sea-ice distribution. All these changes must have left some imprint on the distribution of ^{14}C , which is today the most suitable tracer for studying the dynamics of the global carbon cycle.

New evidence suggests that the marine reservoir age may have differed significantly during the last glacial/interglacial transition. Specifically, we have compared the ^{14}C ages of shallow marine mollusks from the Hebridean Shelf of northwest Scotland and the apparent ages of the Icelandic Vedde ash from this region and elsewhere in the North Atlantic.

¹Department of Geology and Geophysics, University of Edinburgh, West Mains Road, Edinburgh, EH9 3JW, Scotland

²CEREGE, JE 192 and FU CNRS 17, Université d'Aix-Marseille III, Europole de l'Arbois, BP 80, 13545 Aix-en-Provence, CEDEX 4, France

³Department of Geography and Geology, Cheltenham and Gloucester College, St. Georges Place, Cheltenham, GL5 0PP, England

⁴18 McLaren Road, Edinburgh, EH9 2BN, Scotland

The extremely high sedimentation rates, with nearly 500 cm of sediment representing the Younger Dryas (YD) interval, allow us to resolve the Vedde ash into a single, stratigraphically well-constrained horizon within British Geological Survey vibrocore 57/-09/46 (Fig. 1) (*cf.* Selby 1989; Austin 1991; Peacock *et al.* 1992; Hunt *et al.*, in press; Kroon and Austin, in press; Austin and Kroon (ms.).)

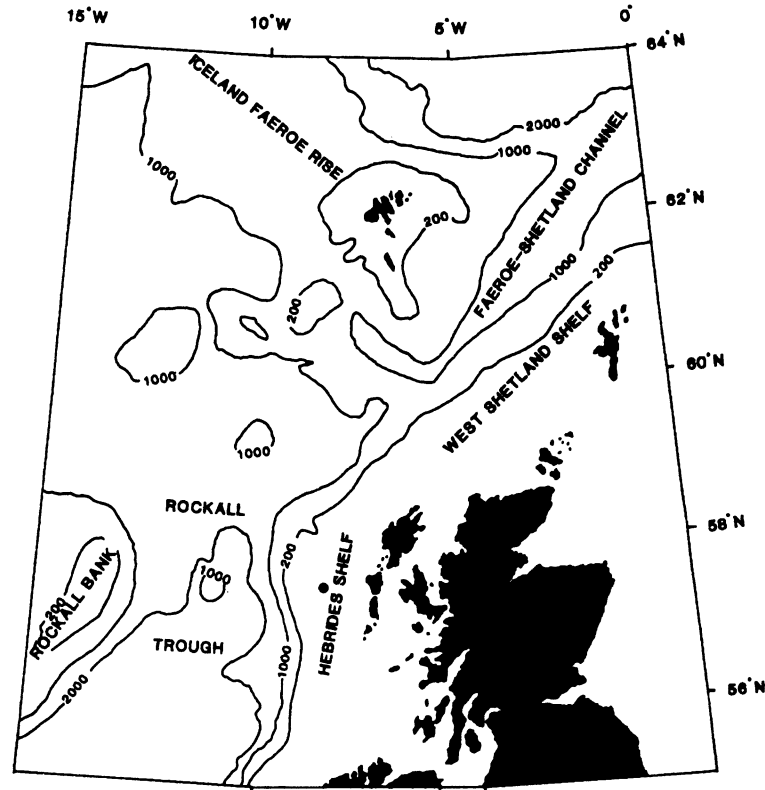


Fig. 1. Location map and bathymetry of core VE 57-09-46 (water depth 156 m)

METHODS AND RESULTS

We extracted molluscan samples from washed and dried bulk sediment samples and identified them by species before submitting them for accelerator mass spectrometry (AMS) ^{14}C analyses. The samples were processed by the NERC ^{14}C laboratory at East Kilbride, according to the procedures outlined by Gillespie, Hedges and Humm (1986) and prepared targets analyzed at Oxford (OxA) and Toronto (TO). Table 1 lists the results of the ^{14}C measurements. These data show a largely conformable sequence, with ^{14}C ages decreasing upwards through the core from $11,400 \pm 70$ yr BP (conventional ^{14}C yr) at a core depth of 565–568 cm to $10,380 \pm 100$ BP at a core depth of 47–51 cm.

We did not find evidence for the Late Glacial Interstadial–YD boundary at the base of this core. However, a nearby core (57/-09/89) from the margins of the St. Kilda Basin contains a longer record (Peacock *et al.* 1992) that yields a conventional ^{14}C age of $11,440 \pm 120$ measured on *Parvicardium ovale* underlying this regionally well-defined climatostratigraphic boundary. This suggests that the base of the core, at a depth of 579 cm, is almost coincident with the basal YD boundary.

TABLE 1. ^{14}C Ages Obtained by AMS on Mollusks from Core VE 57-09-46

Lab no.	Species	Core depth (cm)	Conventional ^{14}C age (yr BP)*
<i>VE57-09-46</i>			
OxA-2786	<i>Acanthocardia echinata</i>	47-51	10,380 ± 100
OxA-2787	<i>Nuculoma belotti</i>	105-130	10,580 ± 100
TO-3127	<i>Nuculoma belotti</i>	206-209	10,610 ± 70
TO-3128	<i>Nuculoma tenuis</i>	230-233	10,970 ± 70
OxA-2788	<i>Nuculoma belotti</i>	480-500	11,420 ± 120

*Ages are conventional ^{14}C ages (i.e., normalized for $\delta^{13}\text{C}$ but not corrected for reservoir age).

The YD-Holocene boundary is clearly defined by lithological and faunal evidence at a core depth of 85 cm (Austin 1991). It would therefore appear that almost the entire YD is present in this core and that it spans a period of >1000 ^{14}C yr (Johnsen *et al.* 1992; Alley *et al.* 1993). This interval is represented by nearly 500 cm of sediment, suggesting an average YD accumulation rate of 0.5 cm yr⁻¹ within the central St. Kilda Basin. Our evidence suggests that sedimentation rates did not remain constant during the YD. The best fit for the age-depth data is a second-order polynomial curve, with an r^2 value of 0.94 (Fig. 2). This fitted curve suggests that sediment accumulation rates were considerably higher during the early part of the YD, falling from 1.7 cm yr⁻¹ at 570 cm to >0.3 cm yr⁻¹ above 100 cm. From this fitted curve, we can derive an expression for the age-depth relation within core VE 57-09-46 during the YD

$$\text{conventional } ^{14}\text{C age} = 10212 + 3.146 (\text{depth cm}) - 1.695 \times 10^{-3} (\text{depth cm})^2 . \quad (1)$$

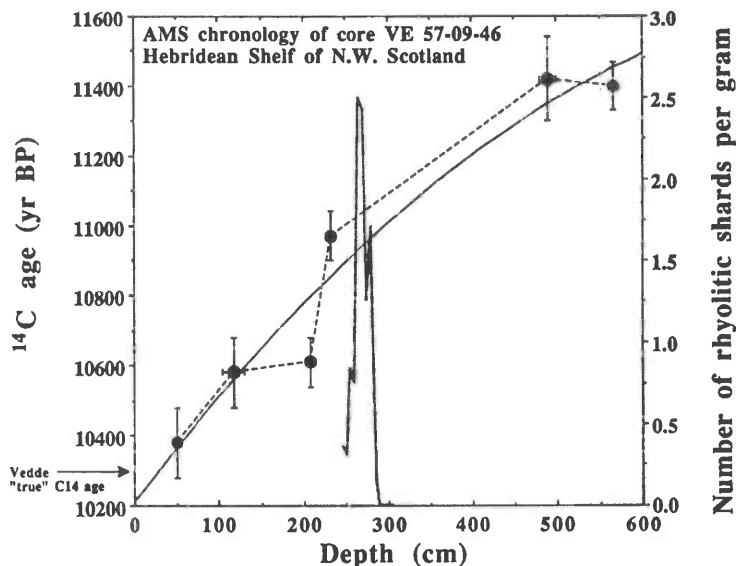


Fig. 2. ^{14}C age vs. depth model for core VE 57-09-46. The concentration of rhyolitic shards is indicated on the right axis. The ^{14}C age of the Vedde eruption is ca. 11,000 BP, based on the dates on mollusks. The true ^{14}C age of the eruption is 10,300 BP, based on data obtained in terrestrial sediments (Bjorck *et al.* 1992; Bard *et al.* 1994; Gulliksen *et al.* 1994; Wohlfarth, Bjorck and Possnert 1995).

The peak of clear acidic shards is centered around a core depth of 265 cm, with their first appearance at 285 cm and a gradual decline in shard numbers upward to 230 cm. These shards exhibit the typical "winged" morphologies associated with the rhyolitic Vedde ash (Fig. 3). These tephra can be correlated with the Rhy 1 shards studied by Kvamme *et al.* (1989) within North Atlantic Ash Zone 1 (NAAZ1) and with the Vedde ash described from lacustrine sediments in western Norway by Mangerud *et al.* (1984).



Fig. 3. SEM photomicrograph of a typical rhyolitic Vedde Ash shard

We determined the geochemistry of the tephra by electron microprobe analysis according to methods outlined by Hunt *et al.* (in press), who measured the basaltic tephra at the University of Edinburgh; H. Haflidason and W. Austin measured the acidic component of the Vedde ash at the University of Bergen, Norway. For our counts of clear, acidic, winged shards (Fig. 2), we used those with a sieved size of $>250 \mu\text{m}$. Figure 4 presents the results of the geochemical analyses, which agree with previously published analyses of the rhyolitic fraction of the Vedde ash (Mangerud *et al.* 1984; Kvamme *et al.* 1989).

We also recognized, in this core, three distinct basaltic populations, all geochemically indicative of an Icelandic origin (Kvamme *et al.* 1989) at depths between 340 cm and 100 cm (Hunt *et al.*, in press). Two of these basic populations, STK-1 and STK-2, are basaltic tholeiites and can be correlated with 1 Thol. 1 (similarity coefficient (s.c.) = 0.95) and 1 Thol. 2 (s.c. = 0.98), respectively. The third, STK-3, is a transitional alkali basalt and can be correlated with 1 Tab 1 (s.c. = 0.95). The latter is thought to represent the basic component of the Vedde ash. We are currently re-examining this core in an attempt to resolve the basaltic populations into a stratigraphic sequence of eruptive events. Earlier investigations by Selby (1989) and Peacock *et al.* (1992) suggest that this is possible.

Attributing a ^{14}C age to the Vedde ash eruption is not trivial because the ages are not directly measured on the shards. Potential problems include the depth of the dated mollusks while living, bioturbation, reworking of older sediment and ice rafting of ash shards. The living depth of the dated shells strongly affects the accuracy of the sediment chronology. However, the species chosen for AMS dating are small nuculacean bivalves that usually live at or a few cm below the sediment surface (*cf.*

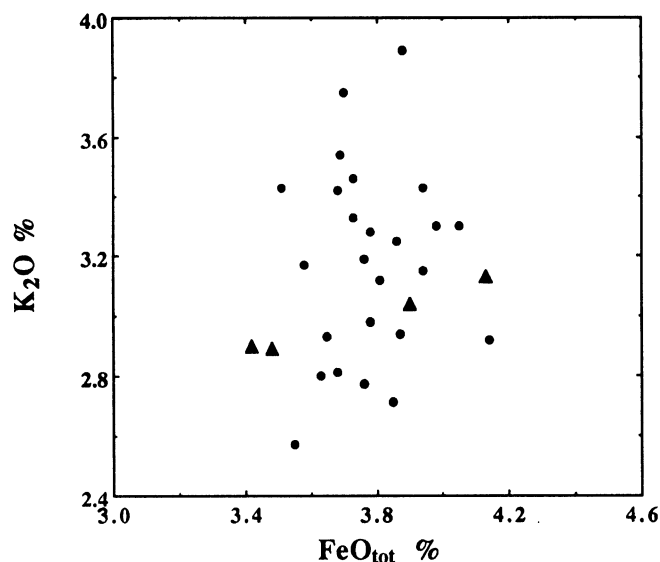


Fig. 4. K_2O vs. FeO_{tot} geochemical plot characterizing the Vedde Ash shards. ● = the Vedde Ash observed in Norway (Mangerud *et al.* 1984); ▲ = shards from core VE 57-09-46.

Yonge and Thompson 1976). Consequently, it is safe to assume that no systematic lag is created between the age of the shells and the true age of the sediment.

As bioturbation models clearly show (*e.g.*, Berger and Heath 1968), there is an initial downward transport from the sediment surface related to the mixed layer depth (usually <10 cm for the deep ocean). Fortunately, however, in a shelf area characterized by extremely high sedimentation rates on the order of 1 cm yr^{-1} , the bioturbation should be negligible, creating a maximum lag on the order of a decade for a bioturbation depth of 10 cm.

Nielsen, Heinemeier and Rud (1994) demonstrated convincingly that the sand-sized fraction of sediment can be continuously reworked through a process of lateral transportation to the site of deposition. They studied coastal cores from the Skagerrak region characterized by extremely high sedimentation rates (on the order of 1 cm yr^{-1}), observing significant and systematic discrepancies (ranging from several centuries to a few millennia) between the AMS ^{14}C ages obtained on mollusks and those measured on benthic foraminifera. Their interpretation is that the sand-sized foraminifera are continuously eroded from older marine deposits, transported and redeposited at the site, and that they are, on average, older than the true age of the sediment, which is given by the shells.

The shelf sediments from the St. Kilda region are also characterized by extremely high accumulation rates on the order of 1 cm yr^{-1} . Following the same line of reasoning as Nielsen, Heinemeier and Rud (1994), the shells of mollusks are immobile, and thus in place, whereas the sand-sized ash shards (>250 μm) have been reworked over a depth of *ca.* 30–40 cm. The true depth of the Vedde ash deposition should thus be its deepest occurrence in the sediment column, 285 cm, corresponding to a shell age of *ca.* 11,000 BP. An initial entrainment by bioturbation (<10 cm) would reduce the depth to *ca.* 275–285 cm.

On the continent, the volcanic ash was windblown (Mangerud *et al.* 1984), whereas in the deep-sea sediments, the ash shards were also rafted by drifting sea ice, which could have further delayed dep-

osition (Ruddiman and Glover 1972). The $>250\ \mu\text{m}$ size of the shards counted from core 57/-09/46 lie (at a distance of nearly 1000 km from Iceland) outside the size-distance relations for the subaerial fallout of tephra, according to the data from Fisher (1964) and Walker (1971). The high accumulation rate observed in St. Kilda sediments provides a unique opportunity to quantify this age delay between wind deposition and ice rafting. From Figure 2, it is clear that the shards are spread over a depth of 30–40 cm, a stratigraphic spread probably due to reworking. Assuming that the spread results from delayed deposition by icebergs and sea ice, it is possible to calculate an upper bound of *ca.* 30–40 yr for the duration of this ice-rafting effect. For the depth range 230–285 cm, this represents a period of 125 yr, whereas a 30-cm spread centered upon 265 cm represents a maximum of 70 yr. These simple calculations confirm the statement by Kvamme *et al.* (1989) that the delay in deposition was only a few years, and that the rhyolitic tephra can indeed be used as a geologically instantaneous time marker.

Based on the observed depths of the Vedde ash and the ^{14}C age-depth relation (Eq. 1), we can date the earliest occurrence at 285 cm to 10,970 BP, the peak in shard concentration at 265 cm to 10,930 BP, and the last occurrence at 230 cm to 10,850 BP. Taking into account the different phenomena described above, the best estimate of the age of the Vedde eruption is, thus, on the order of 11,000 BP (275–285 cm).

Comparison with Other Estimates of the Vedde Ash ^{14}C Age

Most of the age determinations of the Vedde ash layer were obtained from lacustrine sediments. The recent use of the AMS technique has enabled more accurate dating of selected terrestrial macrofossils, at the same time minimizing or eliminating the hard-water effect. Recent studies conducted on lacustrine sediments from Iceland (Björck *et al.* 1992), Norway (Bard *et al.* 1994, Gulliksen *et al.* 1994) and Sweden (Wohlfarth, Björck and Possnert 1995) indicate that the Vedde ash eruption occurred at *ca.* 10,300 BP. By comparing this age with the one determined in the St. Kilda area (close to 11,000 BP), we conclude that during the YD, the reservoir age at this location was on the order of 700 yr, whereas it is now on the order of 400 yr.

This new estimate of the reservoir age essentially agrees with those determined by using the same rationale for deep sea cores (Bard *et al.* 1994). In that case, the reservoir age is more difficult to quantify, since bioturbation also generates a significant age difference between ash layer and foraminifera. However, after correction for a bioturbation bias, it appears that the North Atlantic reservoir age was a few centuries greater than today (700–800 yr instead of 400–500 yr). Figure 5 gives three examples of deep-sea sediment cores in the Vedde ash layer section that have been AMS-dated; bioturbation displacement of ash shards is evident in each. The magnitude of the age bias between ash and foraminifera depends on the sedimentation rate and the bioturbation mixing depth (estimated from the exponential decrease of the ash concentration). For each core it is possible to derive a corrected ^{14}C age for the Vedde ash deposition on the order of 11,000–11,100 BP (Bard *et al.* 1994). Sediment reworking should not have a major effect on the study of these deep-sea sediments because additional proxies ($\delta^{18}\text{O}$, transfer function SSTs) were obtained on the samples that were used for AMS dating. These other measurements independently confirmed that the AMS-dated foraminifera were indeed living during the YD. Moreover, both ash shards and foraminifera would be reworked, because both are mobile when subject to reworking.

IMPLICATIONS FOR NORTH ATLANTIC PALEOCEANOGRAPHY

The finding that North Atlantic reservoir ages were larger during the YD has two important implications. First, it limits our ability to compare climatic time series dated by ^{14}C on foraminifera or

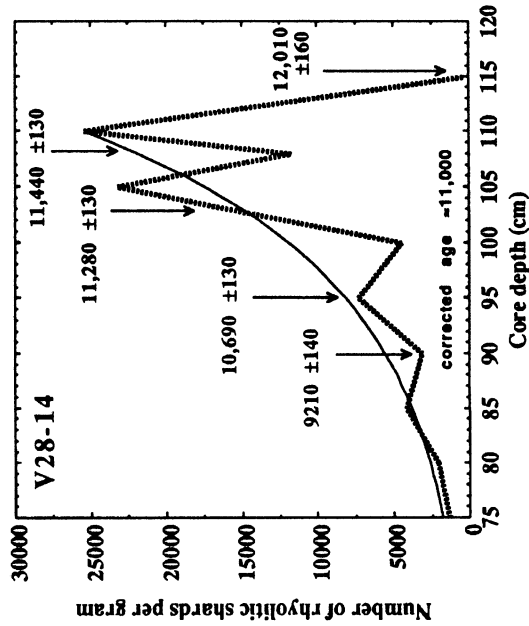
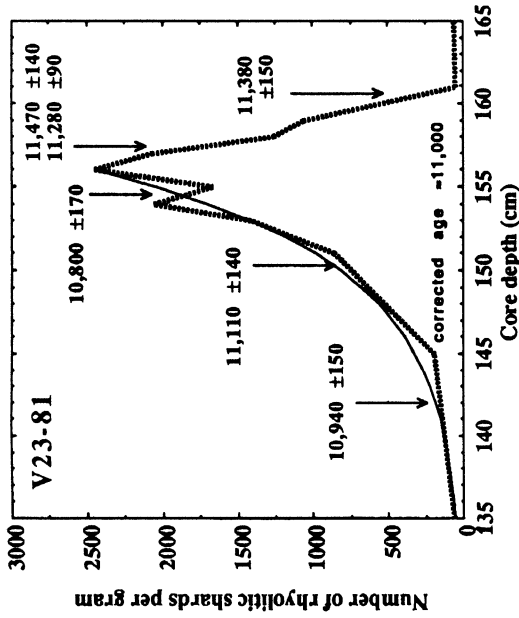
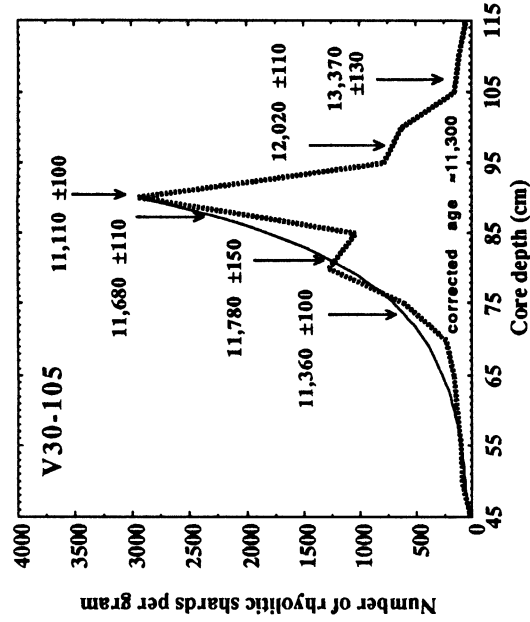


Fig. 5. Distribution of rhyolitic glass shards and conventional ^{14}C ages obtained on deep-sea cores V23-81, V28-14 and V30-105 (from Bard *et al.* 1994). Exponential fits of the distribution tails were used to calculate bioturbation depths for each core. Under each curve is given the ^{14}C age of the Vedde eruption, corrected for the effect of bioturbation (see Bard *et al.* 1994).

mollusks with other time series obtained in other oceanic areas or in continental deposits. The clues to the causes of the deglacial climatic changes will probably come from precise determinations of leads between the time series obtained in the different compartments of the ocean-atmosphere system. During the last deglaciation, the major climatic changes occurred as very abrupt steps (within a few centuries) and it is now crucial to provide ^{14}C ages at this level of accuracy.

As an example, consider the North-Atlantic record of core Troll 3.1 (Lehman *et al.* 1991; Lehman and Keigwin 1992), which has been extensively cited and compared with continental and ice-core chronologies. In Troll 3.1 sediments, the YD boundaries are well marked by sharp changes in $\delta^{18}\text{O}$ and in the relative abundance of the polar foraminifera *Neogloboquadrina pachyderma* (sinistrally coiled). The ^{14}C chronology is based on a regular sequence of *ca.* 10 AMS ages for the last 15 ka. The authors have used a constant reservoir correction of 440 yr, resulting in remarkable YD start and end dates of 11,280 BP and 10,530 BP, respectively (Lehman and Keigwin 1992). These ages are much older than the consensus values for these well-recognized climatic boundaries, 11 and 10 ka BP (Mangerud *et al.* 1974; Hajdas *et al.* 1993; Goslar *et al.* 1994). Peacock and Harkness (1990) suggest that polar waters returned to the seas adjacent to Scotland by *ca.* 10,850 BP and were present until *ca.* 10,200–10,100 BP. The discrepancy in Troll 3.1 could be removed by invoking larger reservoir ages during the last deglaciation (Stuiver and Brazunias 1994).

Second, it affects our understanding of the mechanisms that generated the increase in reservoir age. Today the North Atlantic is characterized by a reservoir age on the order of 400–500 yr, whereas all other high-latitude oceans show larger reservoir ages of 500–1000 yr (Stuiver, Pearson and Brazunias 1986; Bard 1988; Southon, Nelson and Vogel 1990). This major difference is linked to the northward advection of surface and thermocline waters from lower latitudes that travel through the Gulf Stream and North Atlantic current systems. After recirculation and winter convection, this water flux ultimately feeds the North Atlantic deep water (NADW).

Consequently, any change in the North Atlantic reservoir age is probably a response to variations in the NADW formation process and/or changes in the rate of ocean-atmosphere gas exchange. During the cold/warm swings of the last deglaciation, both processes changed dramatically, and the observed effect is probably the combined result of a decrease of NADW flux (Boyle and Keigwin 1987; Keigwin *et al.* 1991) and an increase of North Atlantic sea ice (Koc-Karpuz, Jansen and Hafli-dason 1993).

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THE YOUNGER DRYAS COLD EVENT—WAS IT SYNCHRONOUS OVER THE NORTH ATLANTIC REGION?

TOMASZ GOŚLAR,¹ MAURICE ARNOLD² and MIECZYŚLAW F. PAZDUR^{1,3}

ABSTRACT. Determined independently from annually laminated ice cores and lake sediments, and German pines, the calendar ages of Younger Dryas (YD) boundaries significantly disagree with one another. ¹⁴C dates, plotted vs. calendar ages for samples from different sediments, also reveal distinct offsets. The adjustment of varve chronologies to synchronize the boundaries of the YD nearly cancels the discrepancies between ¹⁴C data, and supports the synchronism of the YD cold period over the North Atlantic region. However, the exact timing of the event cannot be estimated in this way.

INTRODUCTION

The Younger Dryas (YD), an abrupt, temporary cooling *ca.* 12 ka ago, was the last in a long series of brief climatic oscillations during the past 70 ka. These events were associated with an apparent shift of surface-air temperature in the North Atlantic region of 4–7°C within several decades, as recorded in Greenland ice cores (Johnsen *et al.* 1992; Taylor *et al.* 1993b; Grootes *et al.* 1993). Although the YD has been documented mainly in Greenland ice (Johnsen *et al.* 1992; Alley *et al.* 1993; Dansgaard *et al.* 1993; Dansgaard, White and Johnsen 1993; Mayewski *et al.* 1993; Taylor *et al.* 1993a,b; Grootes *et al.* 1993), in lacustrine sediments in Europe (Watts 1980; Pons *et al.* 1987; Lotter *et al.* 1992; Zolitschka, Haverkamp and Negendank 1992; Goslar *et al.* 1993), and in deep-sea cores from the North Atlantic Ocean (Bard *et al.* 1987; Lehman and Keigwin 1992), evidence of this event has also been found in northeastern America and eastern Canada (Peteet *et al.* 1990; Mott *et al.* 1986; Levesque *et al.* 1993). In Colombia (van Geel and van der Hammen 1973), as well as in deep-sea cores from the Northwest Pacific and the Sulu Sea (Kudrass *et al.* 1991), observations of similar oscillations suggest that the YD was at least a hemispheric event. YD-like events have also been recognized in Africa and Antarctica (Roberts *et al.* 1993; Jouzel *et al.* 1992). Nevertheless, only a few records document the YD cold event with an annual resolution and provide an independent time scale of calendar years. Until now, such records have been concentrated in the North Atlantic region – Greenland Summit (GRIP and GISP2) ice cores, European annually laminated sediments (Swedish varves, Lake Gościąg, Lake Holzmaar and Soppensee) and German pine wood. These records are crucial for better understanding the response of climate in different parts of Europe to major climate shifts in the North Atlantic region. We discuss here the question of synchronism of YD reconstructed in these archives.

DEFINITIONS OF YD BOUNDARIES AND THEIR CALENDAR AGES IN DIFFERENT ARCHIVES

Table 1 lists estimates of the calendar age of the YD/Holocene boundary. The definitions of this boundary differ among archives. The boundary is defined most sharply in the change of accumulation rate of Greenland snow (completed in 20–30 yr). The changes of oxygen isotope ratios in Greenland ice and Lake Gościąg carbonates (50–70 yr) were as rapid as changes of fluxes of calcium and magnesium. The boundary, by convention, is placed in the middle of the period of rise (decline) of appropriate data. The transitions in vegetation cover (Gościąg, Soppensee, Holzmaar) responding to climate warming took a longer time, but major changes were completed in 100–200 yr. Here, the YD/PB (Preboreal) boundary is defined as a boundary between pollen assemblage

¹Radiocarbon Laboratory, Institute of Physics, Silesian Technical University, Krzywoustego 2, PL-44-100 Gliwice, Poland

²Centre des Faibles Radioactivités, CNRS-CEA, F-91198 Gif sur Yvette, France

³Deceased 11 May 1995

TABLE 1. Comparison of Calendar-Age Estimates of the Younger Dryas/Holocene Transition

Archive	Definition of boundary	Age (yr BP)	Reference
GRIP ice core	Abrupt increase of ^{18}O	11,550 \pm 90	Johnsen <i>et al.</i> (1992)
GISP2 ice core	Abrupt increase of ^{18}O	11,640 \pm 250	Alley <i>et al.</i> (1993); Taylor <i>et al.</i> (1993a,b); Mayewski <i>et al.</i> (1993)
Lake Gošciaż	Abrupt increase of ^{18}O , changes in terrestrial and lacustrine vegetation	11,440 \pm 120	Goslar <i>et al.</i> (ms.)
Lake Soppensee	Changes in terrestrial vegetation	10,986 \pm 69	Hajdas <i>et al.</i> (1993)
Lake Holzmaar	Changes in terrestrial vegetation	10,630 \pm 180*	Zolitschka, Haverkamp & Negendank (1992); Hajdas (1993)
Swedish varves	Onset of rapid retreat of ice margin	11,510 \pm 180†	Strömberg (1994)
	Second drainage of Baltic ice lake	10,940	
German pines	Increase of ^{13}C and D in wood	10,980	
		(10,970)‡	Becker, Kromer and Trimborn (1991); Kromer and Becker (1993)
		11,045§	

*Varve chronology of Lake Holzmaar

†Varve chronology of Lake Holzmaar corrected with the match of AMS ^{14}C dates to the ^{14}C calibration data

‡Boundary set originally in the pine chronology

§Boundary set originally in the pine chronology, shifted with a tentative tree-ring match to the oak master chronology

zones, and is placed approximately in the middle of the period of rapid change in vegetation. The slowest transition was that observed in isotopic composition of carbon and hydrogen in German pines (*ca.* 500 yr), and here the YD/PB boundary was set at the beginning of the period of change. The duration of major change in the Swedish study is difficult to determine. We must stress that the durations of major climate change, when reconstructed by proxy data of the same type, are similar, but the calendar ages of major change are different, and the differences are well beyond the durations of individual transitions. For that reason, the delay between climate warming recorded in Lake Gošciaż, Lake Holzmaar and Greenland Summit, and those recorded in the Swiss lake, Swedish varves and German pines must be regarded as real unless an error is found in the calendar age estimates of appropriate archives. The same problem can be observed in the climate cooling recorded at the transition between the Allerød (AL) and the YD (Table 2).

TABLE 2. Comparison of Calendar-Age Estimates of the Allerød/Younger Dryas Transition

Archive	Age (yr BP)	Reference
GRIP ice core	12,700 \pm 100	Johnsen <i>et al.</i> (1992)
GISP2 ice core	12,820 \pm 260	Alley <i>et al.</i> (1993)*
Lake Gošciaż	12,580 \pm 130	Goslar <i>et al.</i> (ms.)
Lake Soppensee	12,125 \pm 86	Hajdas <i>et al.</i> (1993)
Lake Holzmaar	11,080 \pm 210†	Zolitschka <i>et al.</i> (1992); Hajdas (1993)
	11,960 \pm 210‡	
Swedish varves	11,800	Wohlfarth <i>et al.</i> (1993)

*Age reported by Alley *et al.* (1993) was based on the changes in accumulation rate; the quoted age is that of the midpoint of major drop of ^{18}O (Grootes, personal communication)

†Varve chronology of Lake Holzmaar

‡Varve chronology of Lake Holzmaar corrected according to the match of AMS ^{14}C dates to the ^{14}C calibration data

COMPARISON OF CALENDAR CHRONOLOGIES

It is always possible that the uncertainties of chronologies, constructed by counting thousands of annual increments, are underestimated. To verify the non-synchronism of AL/YD and YD/PB boundaries apparent in different archives, independent, undoubtedly synchronic markers are necessary. Here, either the layers of volcanic tephra or the global synchronous changes of ^{14}C age can be used.

In Figure 1, we compare the ^{14}C dates from all the archives discussed here (except Greenland ice, of course), with the calibration data based on Barbados and New Guinea corals. In the upper part of the

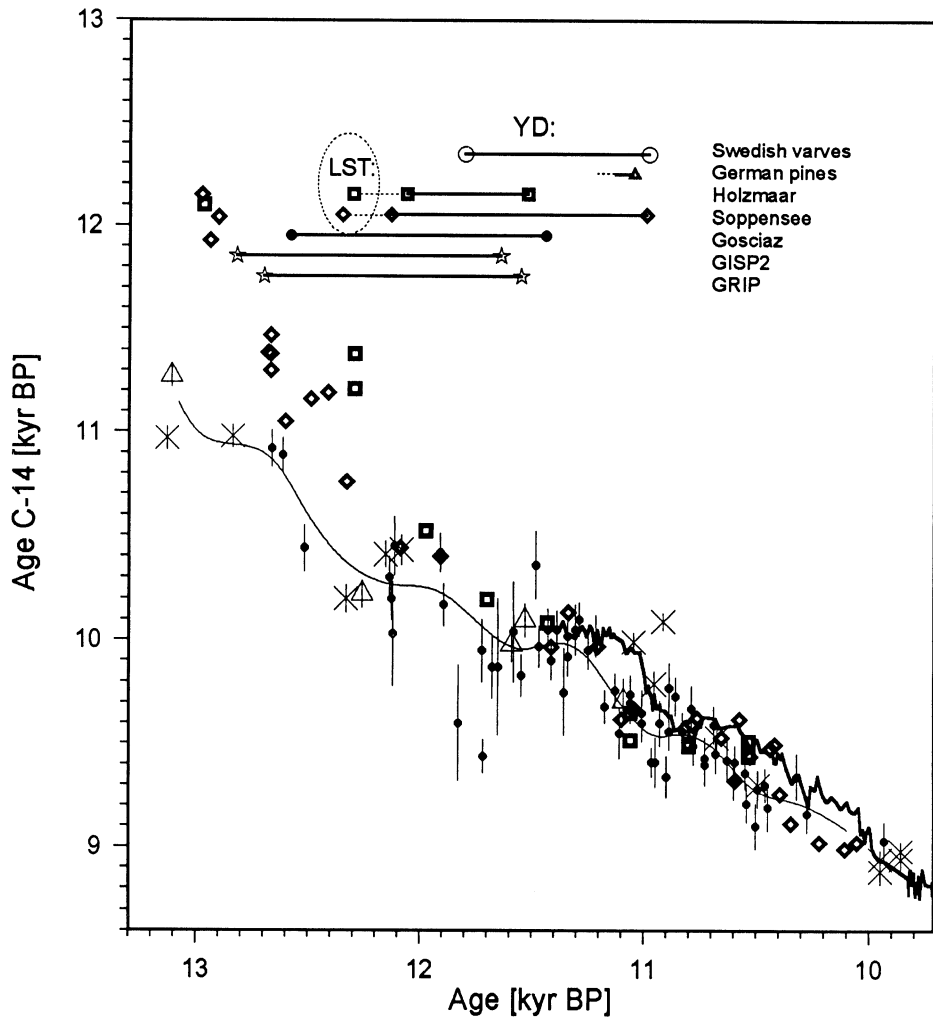


Fig. 1. Comparison of ^{14}C and calendar ages, derived from U/Th dates, dendrochronology and varve chronologies in the Late Glacial and Early Holocene, and of the age of boundaries of the YD cold period, reconstructed in different archives. In the upper part of the figure, the YD boundaries are shown by points connected with heavy lines (except the YD/PB boundary in German pines). The age of the Laacher See tephra is shown to the left of the Holzmaar and Soppensee bars. — = German pines (Kromer and Becker 1993); ■ = Lake Holzmaar macrofossils (Hajdas 1993); ◆ = Soppensee macrofossils (Hajdas *et al.* 1993); ● = Lake Gościadz macrofossils (Goslar *et al.* ms.); △ = Barbados corals (Bard *et al.* 1993); × = Huon Peninsula corals (Edwards *et al.* 1993). — = spline function fitted to Lake Gościadz data. ▲ shows the age of the YD/PB boundary reconstructed in German pines (Becker, Kromer and Trimborn 1991); ☆ = YD boundaries reconstructed in Greenland ice cores.

figure, we compare the calendar ages of boundaries of the YD. We also show the ages of Laacher See tephra (van den Bogaard and Schmincke 1985) found in Holzmaar and Soppensee. The age of the floating varve chronology of Lake Gošciaż was based on the match of ^{14}C dates to the calibration data from German oaks (Goslar *et al.*, ms.). The ages of the YD boundaries from these archives differ, but the discrepancies among ^{14}C data are also large. However, the differences among ^{14}C dates obtained using accelerator mass spectrometry (AMS) from adjacent samples in a single archive are not too large; thus, these dates seem reliable. Although the Lake Gošciaż data generally fit the coral data, the ^{14}C dates from Holzmaar and Soppensee are older. Apparently, not all of the calendar chronologies of Lakes Gošciaż, Holzmaar and Soppensee and corals are synchronous. The differences in age estimates of the AL/YD and YD/PB transitions may be due partly to errors in the calendar chronologies (Goslar *et al.*, ms.). An even higher offset is shown by recent Swedish data (Wohlfarth, Björck and Possnert 1995).

Thus, we tried to “correct” the calendar chronologies to obtain the ages of YD boundaries similar to those recorded in Greenland, and to synchronize exactly the level of Laacher See tephra. This required an addition of *ca.* 450 varves in the chronology of Soppensee below 10.4 ka BP, and *ca.* 600 varves to the sequence from Holzmaar below 11.8 ka BP. The age of the floating varve chronology of Lake Gošciaż was also adjusted to fit the YD boundaries in Greenland. The adjusted age (100 yr older) is still in the range allowed by wiggle-matching to German oaks. The German pine chronology was shifted to synchronize with the Gošciaż chronology. Goslar *et al.* (ms.) discuss in detail the synchronization of the Gošciaż and German pine chronologies. The separate fits of Lake Gošciaż dates to the ^{14}C calibration curve in the portion reconstructed on German oaks and pines suggest a revision of the tentative match of oak and pine chronologies.

In Figure 2, we compare the ^{14}C dates of “corrected” chronologies. We observe that the data from different archives are more consistent than in Figure 1. The plot in Figure 2 clearly demonstrates that the differences in the ages of YD boundaries in laminated sediments are produced mostly by the inadequate calendar chronologies. Some doubts may be connected with the two samples from the YD/PB boundary in Soppensee, distinctly younger than the plateau of 10 ka BP. However, the AMS data from non-laminated sediment of adjacent lake, Rotsee (Ammann and Lotter 1988), with a pollen diagram very similar to that for Soppensee, show the YD/PB boundary in the center of a distinct plateau at 10 ka BP, traced by as many as 11 dates (Fig. 3). Therefore, the two critical samples from Soppensee can be regarded as contaminated. As shown by Wohlfarth *et al.* (1993), the contamination of small macrofossils by modern carbon may sometimes alter a ^{14}C age by many hundred years. Obviously contaminated is one sample from Lake Gošciaż sediment (indicated in Fig. 2 by a question mark).

The only non-synchronous YD/PB boundary is that in German pines which, without any doubt, is delayed by *ca.* 200 yr with respect to that in Lake Gošciaż. Goslar *et al.* (ms.) discuss this delay elsewhere. Here, we note that the beginning of slow increases of $\delta^{13}\text{C}$ and δD in German pines, attributed to the YD/PB boundary (Becker, Kromer and Trimborn 1991) occurred *ca.* 200 yr after the main $\delta^{18}\text{O}$ increase in Lake Gošciaż, during a period of distinct development of elm trees, *i.e.*, after the YD cold period in Poland. As both regions are only 1000 km apart, at the common direction of westerly winds, the main air circulation heating Central Europe from the North Atlantic, it is difficult to imagine that warming on such a scale occurred in the east earlier than in the west. Thus, we conclude that the increased $\delta^{13}\text{C}$ and δD in German pines are, for unknown reasons, delayed with respect to the warming at the termination of the YD. This conclusion does not depend on which chronology (German pines or Gošciaż varves) needs to be revised.

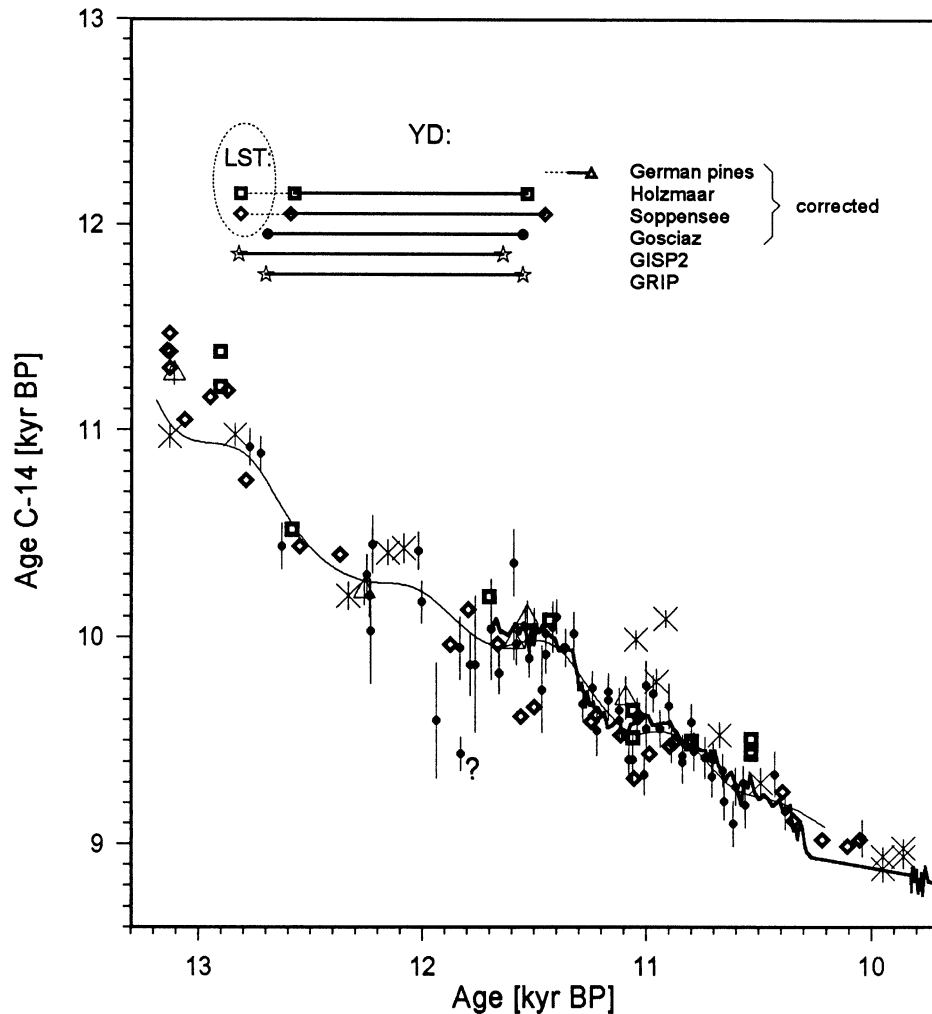


Fig. 2. Revised comparison of ^{14}C dates and boundaries of the YD cold period, reconstructed in archives considered in Fig. 1. The dates are modified after correction of varve chronologies of Lake Holzmaar and Soppensee to synchronize the boundaries of the YD in the North Atlantic region, the adjustment of the Lake Gościadz chronology, and the shift of the German pine chronology to synchronize with that of Lake Gościadz. Symbols are the same as in Fig. 1.

“REAL” AGE OF YOUNGER DRYAS BOUNDARIES

Although demonstrating the synchronism of YD boundaries, the plot in Figure 2 cannot identify their real calendar ages, because one could argue that, along with the Soppensee and Holzmaar chronologies, the uranium/thorium (U/Th) chronology of corals and the varve chronology of Lake Gościadz are inadequate. The correction of Holzmaar and Soppensee chronologies would require some hundred varves missing from the sequences, whereas the error of Lake Gościadz would require the fragment of some hundred varves to be doubled. It must be stressed that, based on AMS ^{14}C dates, Hajdas (1993) demonstrated the lack of *ca.* 880 varves in the Lake Holzmaar sequence from the 4th millennium BP. This gap was not detected previously when analyzing the varve structures. On the other hand, it is difficult to imagine doubling the laminated sequences (by a slump?) with no

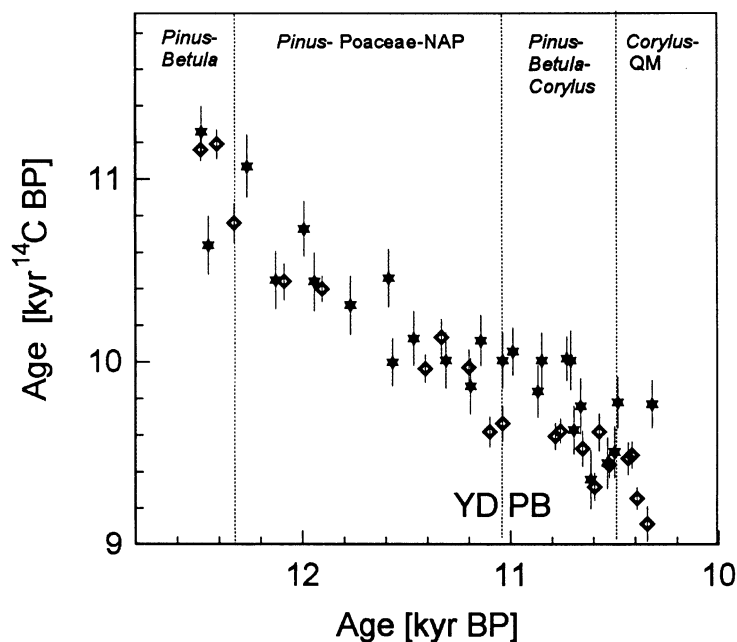


Fig. 3. Comparison of ^{14}C dates of macrofossils from Soppensee (\blacklozenge) and non-laminated sediments of Rotsee (\blackstar) (Ammann and Lotter 1988). The calendar ages of Soppensee samples are as published by Hajdas *et al.* (1993). The time scale for Rotsee was obtained by synchronization of boundaries between corresponding biozones in both lakes (Lotter *et al.* 1992) and linear interpolation between boundaries according to sample depth.

serious disturbance to the laminated structure, and thus seems impossible without visible evidence in varve quality. Further, the close varve-to-varve correlation of laminated sequences from two separate basins of Lake Gošciaż (Gošlar *et al.* 1993) seems to preclude the occurrence of a slump. The serious revision of Lake Gošciaż chronology would also require the revision of the U/Th chronology of corals, which seems unjustifiable. Supporting the validity of Lake Gošciaż and U/Th chronologies is the agreement of the ages of YD boundaries with those recorded in Greenland. That the above-mentioned arguments seem to indicate that varves are missing from the Soppensee and Holzmaar chronologies rather than the Lake Gošciaż chronology is erroneous. If not, we must agree that climate changes at the onset and termination of the YD in Europe were delayed by a few hundred years with respect to the case in Greenland. Further study is necessary to resolve this problem.

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RADIOCARBON DATING THE HOLOCENE IN THE GOŚCIAŻ LAKE FLOATING VARVE CHRONOLOGY

IRENA HAJDAS,^{1,2} GEORGES BONANI,¹ and TOMASZ GOŚLAR³

ABSTRACT. Terrestrial macrofossils selected from laminated sediment of Lake Gościąg were dated by AMS. Thus, part of the floating varve chronology (FVC) (Goslar *et al.* 1993) between radiocarbon ages of 4225 ± 45 and 7740 ± 85 BP can be compared and placed on the ^{14}C calibration curve. As a result of our dating, the top of the FVC is now dated between 3120 and 3300 cal BP, *i.e.*, 3210 ± 90 cal BP.

INTRODUCTION

Terrestrial macrofossils are most suitable for radiocarbon dating lake sediments because they are free of the hard-water effect (Olsson 1986). Short-lived parts of land plants provide the best ^{14}C estimate of the time when the sediment was deposited. A danger of dating reworked macrofossils still exists and cannot be avoided entirely. However, in the case of laminated sediments, this is usually limited to such events as the development of turbidites and slumps.

In many cases, laminations are of poor quality or have disturbed sequences in the upper parts of the sediment record (Hajdas 1993). Therefore, most long varve chronologies, sometimes extending to the Late Glacial period, are either floating or show large counting uncertainties. One possibility for solving this problem is to ^{14}C -date the floating varve chronology (FVC) and place it on the ^{14}C calibration curve (Hajdas *et al.* 1993, 1995.)

Lake Gościąg contains a sedimentary record of the last deglaciation (Goslar *et al.* 1993a; Ralska-Jasiewiczowa, Wicik and Wieckowski 1987), and most of it is well laminated. However, because of difficulty in varve counting, the uppermost 2900 yr were counted with an uncertainty of $-200/+500$ yr. Below this point, well-developed varves are observed, and a long varve chronology has been established (Goślar 1993; Goślar *et al.* 1993). To reduce the uncertainty after 2900 BP, we dated the younger part of the FVC to match it to the dendrochronologically based calibration curve. Sediment samples were selected to allow for coverage of the longest sequence of varves in this lake. Macrofossils from these samples were dated using accelerator mass spectrometry (AMS).

METHODS

Sediment samples containing *ca.* 100 varves (exact number given in Table 1) were treated with acid (10% HCl) to remove carbonates and facilitate washing macrofossils. Gościąg sediment contains *ca.* 40–60% carbonate, so that most of the sediment was removed in this step, followed by washing through a sieve (250-mm mesh). From the remaining residue, we selected only fragments of terrestrial plants (Table 1), so that the hard-water effect, a possible uncertainty of dating, could be ruled out (Hajdas 1993).

We applied a standard cleaning procedure of a consecutive acid/base/acid soaking of organic matter at 60°C for 1 h (Hajdas 1993). Each step was followed by rinsing to neutral pH. Combustion graphitization and sputter target preparation followed the usual procedure. The targets were measured in

¹Institut für Teilchenphysik, ETH-Hönggerberg, CH-8093 Zürich, Switzerland

²EAWAG, Überlandstrasse 133, CH-8600 Dübendorf, Switzerland

³Gliwice Radiocarbon Laboratory, Institute of Physics, Silesian Technical University, Krzywoustego 2
PL-44-100 Gliwice, Poland

TABLE 1. AMS-Dated ^{14}C Ages and $\delta^{13}\text{C}$ Values for Macrofossils Selected from Sediment of Lake Gošćiaź

ETH-no.	Sample	Relative varve age*	^{14}C age (yr BP)	$\delta^{13}\text{C}$ (‰)	Material	C content (mg)
12417, 12418†	G(1+2)MZ	5717 ± 88	7740 ± 85	-31.5 ± 1.2	Bark, catkin scales	1.5
12423	G7MZ	5227 ± 43	7740 ± 70	-33.0 ± 1.1	Bud, twig	2.2
12425	G9MZ	4645 ± 60	7075 ± 80	-27.0 ± 1.2	Twig(s), bark	1.75
12427, 12428†	G(11+12)MZ	2860 ± 80	5505 ± 90	-18.9 ± 1.2	Catkin scales, seeds	0.6
12430	G14MZ	1790 ± 45	4225 ± 70	-24.0 ± 1.3	Needle, leaf fragments	2.4

*In years from top of FVC; macrofossils of two samples were combined into one.

†Uncertainty corresponds to the thickness of sediment sampled for dating.

a cassette with standards (Oxalic Acid I and ANU sucrose) and blanks at the ETH/PSI AMS facility. $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ ratios were measured quasi-simultaneously (Bonani *et al.* 1987).

RESULTS

Conventional ^{14}C ages listed in Table 1 were calculated according to the procedure suggested by Stuiver and Polach (1977). Age correction for the fractionation is based on $\delta^{13}\text{C}$ values (Table 1), which were measured for each graphite sample (see above). For each sample, the type of macrofossil and the amount of carbon is given. A comparison between the fitted chronology and tree-ring curve indicates that two ages (ETH-12417 and ETH-12430) are too young and fall below the calibration curve (Fig. 2). Both samples contained >1 mg of carbon (Table 1), and contamination is unlikely here. We tried to obtain better dating resolution, but the amount of organic material selected from sediment from other depths was not sufficient for dating.

Gošćiaź FVC consists of 7000 varves counted between the uppermost, poorly laminated sediment and the massive layer deposited during the YD (Gošlar *et al.* 1993). Previously, the top of the well-laminated sediment was dated at 2900 +500/-200 cal BP. Figure 1 shows ^{14}C ages of macrofossils plotted vs. relative varve time. Intervals between macrofossil samples contain >500 varves, and the whole ^{14}C -dated varve chronology covers almost 4000 varve years.

We matched the Gošćiaź FVC with the calibration curve using the modified least-squares minimization method (Pearson 1986). We found an absolute offset of the varve age by minimizing the weighted sum of squared differences between ^{14}C ages of macrofossils and those derived from the calibration curve. The weight included uncertainties from sample sizes and AMS dating of macrofossils (Table 1) as well as uncertainties from the calibration curve. Figure 2 shows the FVC fitted to the calibration curve. The top of the FVC dates between 3120 and 3300 cal BP, *i.e.*, 3210 ± 90 cal BP.

CONCLUSION

Radiocarbon dating terrestrial macrofossils is a useful tool for the dating of lake sediments. Dating a sequence of five samples selected from the sediment deposited during a 4000-yr period allowed absolute dating of this segment. Such a strategy, called "wiggles matching" if performed with very high-resolution dating, allows more precise dating of Holocene records. In the case of the Gošćiaź FVC, higher-resolution ^{14}C dating could improve the fit with the ^{14}C calibration curve.

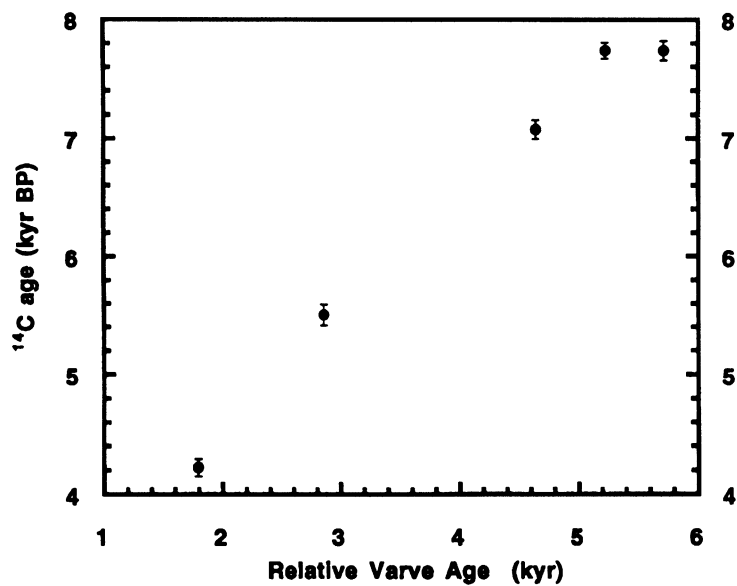


Fig. 1. ¹⁴C ages of terrestrial macrofossils plotted vs. their relative varve age derived from varve counting

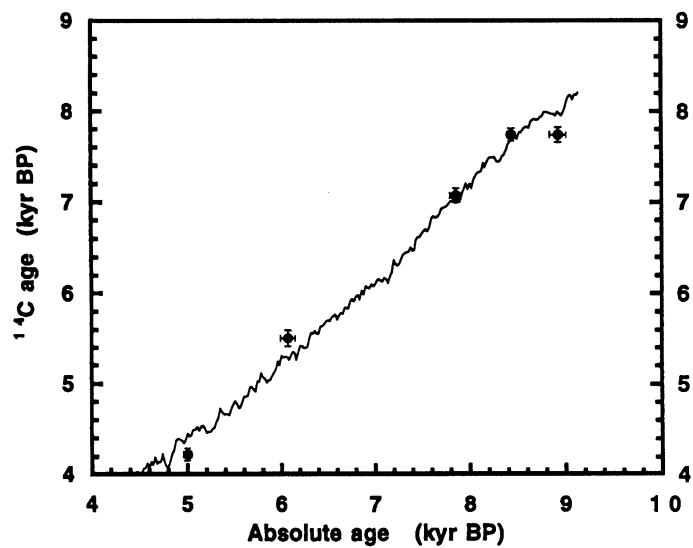


Fig. 2. The best fit of the FVC to the ¹⁴C calibration curve. The top of this chronology dates to 3210 ± 90 cal BP.

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PROBLEMS IN THE EXTENSION OF THE RADIOCARBON CALIBRATION CURVE (10–13 KYR BP)

IRENA HAJDAS,^{1,2} SUSAN D. IVY-OCHS^{1,3} and GEORGES BONANI¹

ABSTRACT. Radiocarbon dating of varved lake sediments shows that, during the Late Glacial (10–12 kyr BP), the offset between the ¹⁴C and the absolute time scales was *ca.* 1 kyr. Varve counting and accelerator mass spectrometry (AMS) dating were used to build absolute and ¹⁴C time scales of sediments from two lakes—Soppensee, Switzerland and Holzmaar, Germany. The resulting chronologies extend back to *ca.* 12.9 kyr cal BP (12.1 kyr BP) in the case of Soppensee and to *ca.* 13.8 kyr cal BP (12.6 kyr BP) in the Holzmaar record. They compare well with each other but differ significantly from the ¹⁴C-U/Th chronology of corals (Bard *et al.* 1993; Edwards *et al.* 1993).

EXTENSION OF THE RADIOCARBON CALIBRATION CURVE

The radiocarbon time scale is affected by variations in the ¹⁴C production rate and changes in the global carbon cycle. The differences between ¹⁴C age and absolute/calendar age can be traced by ¹⁴C dating of material of known absolute age such as trees (calibration curve). Although the continuous tree-ring calibration curve, which is the most precise ¹⁴C calibration method, is available only for the last 11.5 kyr (calendar) (Kromer and Becker 1993), many studies have recently focused on archives alternative to tree rings. Using mass spectrometry (Edwards, Chen and Wasserburg 1987) for precise U/Th dating of small coral samples, Bard *et al.* (1993) and Edwards *et al.* (1993) constructed the ¹⁴C-U/Th curve for the last 20 kyr BP. Another option for studies of variations in the atmospheric ¹⁴C/¹²C ratio is provided by dating of laminated sediments. The accelerator mass spectrometry (AMS) ¹⁴C dating of terrestrial macrofossils deposited in annually laminated (varved) sediments from lakes can be compared with absolute time derived from varve counting. However, as the method is quite often limited by the quality of laminations or the lack of a continuous record, studies of many independent records are required. Two sites, Soppensee (Hajdas *et al.* 1993) and Holzmaar (Hajdas *et al.* 1995b), have been studied for the extension of the calibration curve.

VARVE CHRONOLOGIES – RESULTS FROM SOPPENSEE AND HOLZMAAR

Coupled ¹⁴C/varve time scales were built for Soppensee and Holzmaar. In each case, the ¹⁴C scale is based on AMS measurements on terrestrial macrofossils and the absolute time scale consisted of varve counting. The annual varves (Lotter 1989) in Soppensee are alternating calcite (pale) and organic material (dark) layers, whereas the varves in Holzmaar are clastic (Zolitschka 1991). In both cases, complications in the varve chronologies required corrections. The corrections to each varve chronology were made independently, by statistically matching the younger part to the established tree-ring calibration curve (for details, see Hajdas *et al.* 1993; Hajdas *et al.* 1995b). The ¹⁴C/varve chronology of Soppensee sediments extends to 12.1 kyr BP, which corresponds to *ca.* 13 kyr cal BP (Table 1, Fig. 1). The ¹⁴C record of Lake Holzmaar extends to *ca.* 12.6 kyr BP. On the varve time scale, this corresponds to *ca.* 14 kyr cal BP (Table 1, Fig. 1).

The agreement we obtained between both varve chronologies is best illustrated by the absolute dating of the Laacher See tephra (LST). The Laacher See volcano (West Eifel, G) erupted *ca.* 11.2 kyr BP (Hajdas *et al.* 1995a), and the layer of ash can be found, as an excellent time marker, in sediments

¹Institut für Teilchenphysik, ETH-Hönggerberg, CH-8093 Zürich, Switzerland

²Swiss Federal Institute for Environmental Science and Technology (EAWAG) Überlandstrasse 133, CH-8600 Dübendorf, Switzerland

³Ingenieurgeologie, ETH-Hönggerberg, CH-8093 Zürich, Switzerland

of many European lakes and bogs. The Soppensee varve chronology dates the LST layer at $12,350 \pm 150$ yr cal BP, whereas the varve chronology of the Holzmaar record yields an age of $12,201 \pm 224$ yr. The agreement between the chronologies is exemplified by the absolute age of the LST from the two independent records (Fig. 2).

New results from the Swedish varves (Wohlfarth *et al.* 1995) and Japanese laminated Lake Suigetsu (Kitagawa *et al.* 1995) agree with the Soppensee and Holzmaar varve chronologies up to 12 kyr BP. However, in their reconstruction, ^{14}C ages become close to calendar ages beyond 12 kyr BP. In the Holzmaar chronology, an offset between the ^{14}C and the calendar age is *ca.* 1.4 kyr (see Fig. 1, Table 1) at 12.6 kyr BP. This shows that the atmospheric $^{14}\text{C}/^{12}\text{C}$ ratio was higher at the beginning of the Late Glacial than during the Younger Dryas (YD) and the early Holocene (~ 10 kyr). The ^{14}C -U/Th curve (Bard *et al.* 1993; Edwards *et al.* 1993) shows an even larger (up to 2 kyr) offset between ^{14}C

TABLE 1. AMS ^{14}C Ages of Terrestrial Macrofossils Selected from Sediments of Soppensee and Holzmaar and the Corresponding Varve Ages

Site	ETH-no.	Sample, depth (cm)	^{14}C age (yr BP)	Varve age (cal yr BP)
<i>Holzmaar*</i>				
	7246	HZM9	9515 ± 75	10,904
	7241-1	HZM10.1	$10,085 \pm 80$	11,245
	7248-3	HZM11.3	$10,195 \pm 85$	11,510
	7249	HZM12	$10,520 \pm 90$	11,786
	7250	HZM13-a	$11,210 \pm 95$	12,101
	7250	HZM13-b	$11,380 \pm 90$	12,101
	12471+2†	HZM30.1+2	$11,250 \pm 110$	12,236
	12475	HZM32.1	$11,600 \pm 140$	12,737
	12476	HZM32.2	$11,940 \pm 130$	12,750
	7254	HZM17	$12,100 \pm 110$	12,781
	12481+2	HZM35.1+2	$12,570 \pm 130$	13,571
	7255	HZM18	$12,430 \pm 110$	13,752
	7256	HZM19-a	$12,590 \pm 110$	13,757
	7256	HZM19-b	$12,520 \pm 110$	13,757
<i>Soppensee‡</i>				
	7701	540.5–544.5	9970 ± 100	$11,204 \pm 104$
	7710	544.5–549.5	$10,135 \pm 100$	$11,335 \pm 118$
	6803	549.5–551.5	9965 ± 75	$11,413 \pm 102$
	6828	568.5–569.5	$10,400 \pm 70$	$11,909 \pm 123$
	7703	573.5–580.5	$10,440 \pm 100$	$12,088 \pm 157$
	5290	593–595	$10,760 \pm 105$	$12,329 \pm 139$
	6930	596.5–598.5	$11,190 \pm 80$	$12,412 \pm 146$
	6932	599.5–601.5	$11,160 \pm 60$	$12,488 \pm 154$
	6804	603.5–605.5	$11,050 \pm 80$	$12,604 \pm 153$
	6933	605.5–606.5	$11,470 \pm 70$	$12,668 \pm 153$
	5305	606–808	$11,380 \pm 105$	$12,668 \pm 152$
	6805	609.5–610.5	$11,300 \pm 90$	$12,668 \pm 152$
	6806	610.5–611.5	$11,385 \pm 90$	$12,681 \pm 152$
	6807	628.5–630.5	$12,040 \pm 90$	$12,904 \pm 152$
	6808	631–632	$11,930 \pm 90$	$12,940 \pm 151$
	6809	633–634	$12,150 \pm 90$	$12,977 \pm 151$

*Hajdas *et al.* (1993)

†Hajdas *et al.* (1995a)

‡Hajdas *et al.* (1995b)

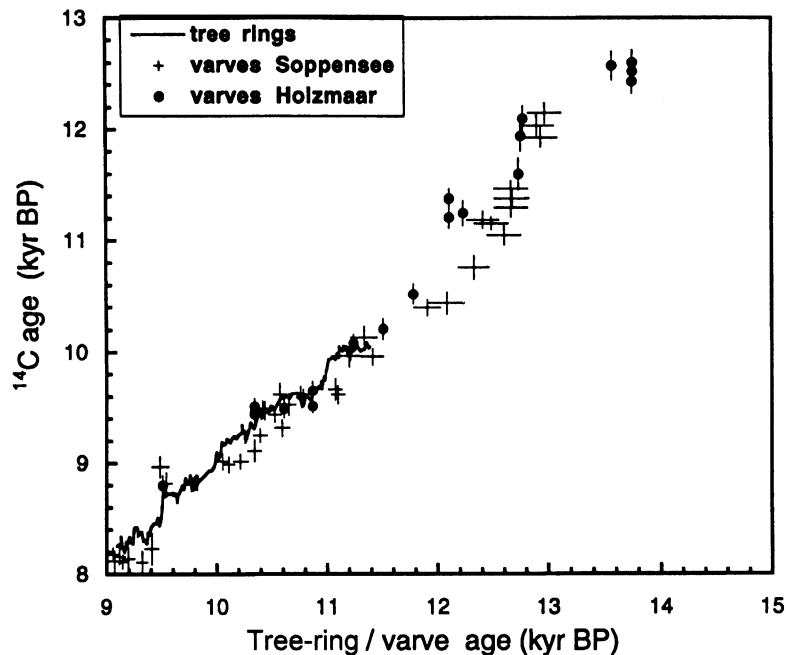


Fig. 1. Comparison between Soppensee and Holzmaar ^{14}C /varve chronologies. The AMS ^{14}C ages (plotted with $1-\sigma$ uncertainty) obtained on terrestrial macrofossils selected from sediment of Soppensee, Holzmaar are plotted vs. calendar years (tree rings and varve years). Both ^{14}C /varve chronologies show features of the tree-ring curve, e.g., plateaus in ^{14}C age at 9.6 and 10 kyr BP.

and calendar years during this period (Fig. 2). A lack of agreement exists between all the methods and an extension of the ^{14}C calibration curve beyond tree-ring data, i.e., 10 kyr BP is still problematic. Because the differences first appear beyond the tree-ring curve (Fig. 2), dating of the Late Glacial is very important for this discussion.

Recently, the duration of the YD, the calendar age of the onset and the end of the event have been determined by studies of various high-resolution records, e.g., ice cores, laminated sediments and tree rings. A comparison between them shows certain similarities but also differences, which cannot yet be explained. There is an offset between the absolute age of the YD/PB (Preboreal) transition in ice cores and the Soppensee record. Although ice cores place it at 11,580 BP (GRIP (Johnsen *et al.* 1992)) and 11,640 BP (GISP 2 (Taylor *et al.* 1993)), i.e., just at the beginning of the 10-kyr ^{14}C age plateau, the Soppensee varve chronology dates the transition at ca. 11 kyr cal BP (Fig. 3), which is at the end of the age plateau. A similar age of ca. 11 kyr cal BP is also indicated by the tree-ring data (Kromer and Becker 1993), although the transition in this record is based on variations of δD and $\delta^{13}\text{C}$ in wood (Becker, Kromer and Trimborn 1991). In sediments of Lake Gościąg, the end of the YD was dated at 11,200 (+500/-200) yr cal BP (Goslar *et al.* 1993). It must be noted that a discrepancy exists between the length of the YD in Soppensee (1140 yr) and Holzmaar (450 yr) (Hajdas *et al.* 1995b). The length and the boundaries of the YD in Holzmaar are currently being more closely studied on new cores (B. Zolitschka, personal communication).

Recently reported ^{14}C dating results from the Norwegian Lake Kråkenes (Gulliksen *et al.* 1994) show that most of the 10-kyr plateau belongs within the YD. Also new data from Gościąg (Goslar *et*

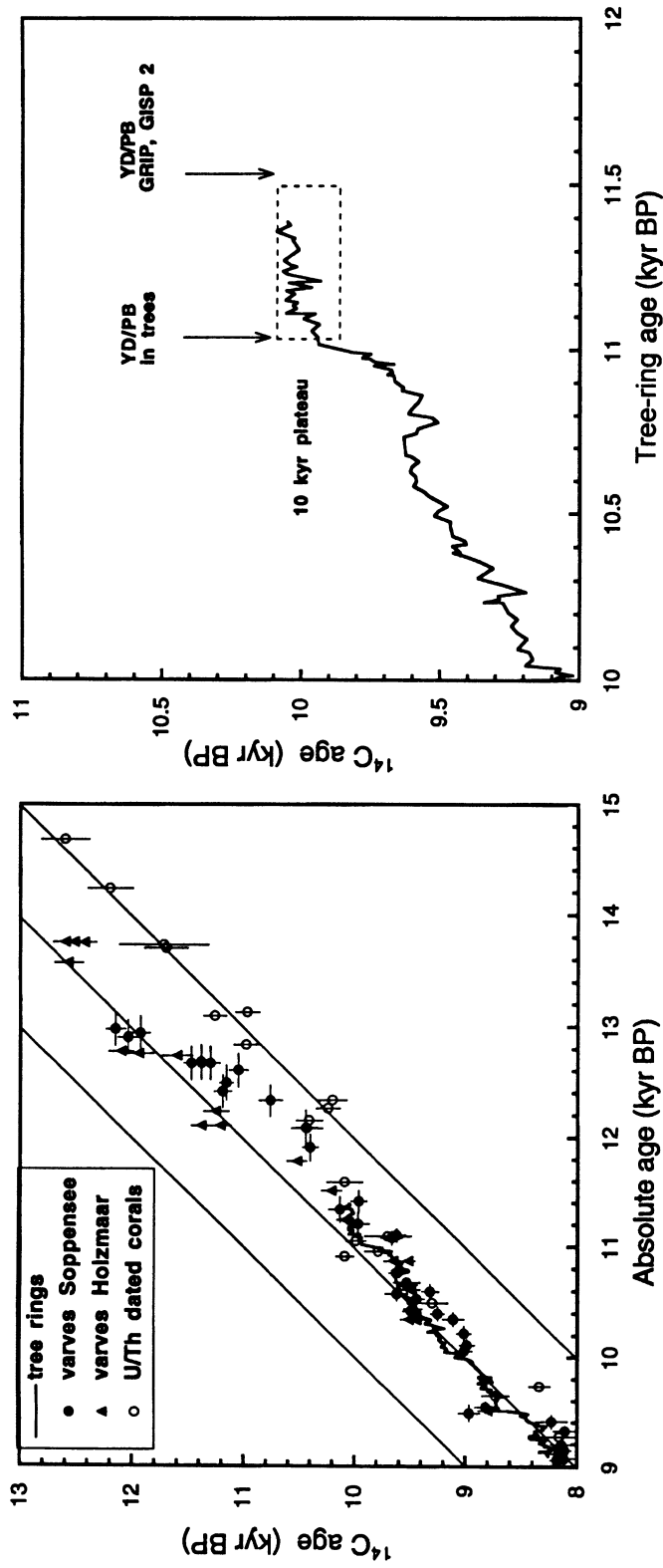


Fig. 2. Extension of the ^{14}C calibration curve beyond 10 kyr BP. Good agreement between tree ring data, varve chronologies (Fig. 1) and the ^{14}C -U/Th chronologies from corals can be seen for the last 10 kyr BP (ca. 11 kyr cal BP). An offset of ca. 1 kyr between the ^{14}C time scale and the calendar time scale of the tree-ring calibration curve is observed at the beginning of the Holocene (10 kyr BP). In the Late Glacial period (10–12.5 kyr BP), varve data indicate the same trend of the ^{14}C calibration curve, *i.e.*, varve ages are ca. 1 kyr older than ^{14}C ages of macrofossils. In contrast, the U/Th ages of corals are ca. 2 kyr older than their corresponding ^{14}C ages.

Fig. 3. Dating of the YD/PB boundary. The arrows indicate the location of the beginning of the YD as determined from the absolute time scale of ice cores and tree rings with respect to the 10-kyr plateau.

al. 1994) do not place the transition at the beginning of the plateau (Fig. 3) as the ice core records do. Assuming that the event was felt simultaneously over the North Atlantic region or even the whole world (Alley *et al.* 1993), differences of up to 500 yr in dating of such dramatic changes seem to be unlikely. Resolving this problem is critical for dating the Late Glacial as well as for the extension of the ^{14}C calibration curve.

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DATING OF LAKE AND LOESS SEDIMENTS

JUERG BEER and MICHAEL STURM

Swiss Federal Institute for Environmental Science and Technology (EAWAG)
CH-8600 Dübendorf, Switzerland

ABSTRACT. Lake and loess sediments represent archives that record important information about the local, regional and global climate conditions in the past. Lake sediments consist of autochthonous particles formed by biogeochemical processes within the lake and allochthonous particles brought into the lake from the catchment area. After deposition, the stratigraphy of the sediment can be altered by chemical, physical and biological processes. Under favored conditions, the sediment shows individual annual layers (varves), which can be used to date the sediment. Other dating methods are based on radioactive decay (^{14}C , ^{210}Pb) or on time markers such as tephra layers, deposits of natural catastrophes, *e.g.*, floods, or radioactivity, *e.g.*, emissions from a nuclear power plant.

Loess is a windblown deposit of fine dust originating from deserts and mountain areas. During the last 2.5 Ma, dust transported over several thousand kilometers formed plateaus up to 200 m thick. During warm and humid conditions, loess deposits are transformed by biochemical processes into soil. Until now, the main dating techniques applied to loess have been magnetostratigraphy (magnetic reversals), matching the ^{10}Be concentration with the $\delta^{18}\text{O}$ deep-sea record, radiocarbon (^{14}C) dating (for the last 40 ka) and thermoluminescence (TL) dating (for the last 100–200 ka).

INTRODUCTION

Terrestrial archives such as sediments and loess contain important information on local, regional and global environmental conditions of the past. They offer a unique opportunity to study the dynamics of the environmental system spanning a period from 10 ka (lakes) up to 1 Ma (loess). Understanding the properties and natural variability of the environmental system is fundamental to estimating the importance of global changes caused by humans. Two main difficulties in using natural archives to reconstruct the history of the environment are to read the information and to establish a precise and reliable time scale. We discuss here two types of archives, lake sediments and loess, in some detail; our intent is not to provide a complete overview of the field, but rather to show the potential contribution of lake and loess sediments to global change study.

LAKE SEDIMENTS

Lake sediments are formed by a variety of biogeochemical and physical processes (Kelts and Hsu 1978). The amounts of autochthonous or allochthonous particles formed by these processes reflect different environmental conditions within a lake, *e.g.*, algal blooms, chemical precipitation, or its catchment area, *e.g.*, erosion, floods, formation of vegetation, respectively. These sediments often show seasonal variations, leading to the deposition of regularly laminated couplets (varves), which represent the duration of a year (Sturm 1979).

Autochthonous biogeochemical processes during the course of a year in lakes without prominent river input are responsible for the formation of suspended particles, which are deposited and subsequently recorded in the composition of an individual varve couplet (Lotter *et al.* 1992). Such biogeochemical varves are distinctly different from physical varves, which are formed in tributary-controlled lakes with high allochthonous particle-loading, reflecting annual meltwater cycles.

After deposition, the main processes that may alter the sedimentary record are:

- Turbidity currents induced by earthquakes or internal waves, which transport large amounts of sediment from unstable slopes to flat parts of a deep lake

- Physical resuspension by currents along the water/sediment interface, which pick up fine particles to redeposit them in different parts of the lake basin
- Chemical remobilization of ions during changing redox conditions
- Bioturbation by activity of benthic fauna may mix the upper centimeters of the sediment.

Figure 1 summarizes the different processes involved in the formation and alteration of sediments.

Environmental Information

From a conceptual point of view, every process governing the input of particles into a lake or the formation of particles within a lake is dependent in complex ways on the climate, the local vegetation and the physical, chemical and biological conditions of the lake. A complete analysis of the chemistry, mineralogy, biology and paleomagnetism of the sediments is usually not feasible, but information that can be extracted includes:

- Regional and global climate—information is provided mainly by stable isotopes ($\delta^{18}\text{O}$) and pollen analysis
- Natural catastrophes—frequency and magnitude of extreme events, such as floods, earthquakes and volcanic eruptions
- Atmospheric fallout—natural and artificial radionuclides and other species removed from the atmosphere are built into the sediments
- Human influence on the catchment area—activities such as deforestation, eutrophication, agriculture, and pollution by xenobiotic organic substances and heavy metals.

Lake Sediments As an Archive

An advantage of studying lake sediments is that they are widespread and relatively accessible. They record a variety of natural processes of the past, reflecting changes of environmental conditions, such as climate, hydrology, biology and human activity. Although they contain global information, the dominant signal is determined by local and regional properties of the catchment area. Accumulation rates of lake sediments are typically on the order of $0.1\text{--}1.0\text{ cm yr}^{-1}$. Easily accessible sediment cores cover a time range up to the Late Glacial (15 ka BP). A core with a length of 50–100 m may cover an entire glacial and interglacial period (>100 ka BP).

Dating

The value of an archive is strongly dependent on how datable it is. The establishment of a continuous time scale can be seriously hampered by disturbances in the sedimentary record, caused by slumps, bioturbation and chemical remobilization. Several methods can be used to establish a time scale. The most precise dating is based on counting of annual laminae (varves). Other dating methods are based on the radioactive decay of ^{210}Pb ($t_{1/2} = 22.3\text{ yr}$) (von Gunten and Moser 1993) and ^{14}C ($t_{1/2} = 5730\text{ yr}$) (Hajdas *et al.* 1993). Absolute time markers, such as ^{137}Cs horizons at 1963 (caused by nuclear weapons testing) and 1986 (caused by the Chernobyl accident), volcanic ash layers (Laacher tephra) or historically recorded events, such as rock falls or earthquakes, can also be used for absolute dating. Records can be synchronized by matching these time markers and by correlating records of magnetic susceptibility, pollen/diatom stratigraphy and geochemical parameters, such as $\delta^{18}\text{O}$, $\delta^{13}\text{C}$, nutrients and metals.

LOESS

Loess is a terrestrial windblown silt deposit consisting chiefly of quartz, feldspar, mica, clay minerals and carbonate grains in varying proportions. Loess originates in desert or mountain areas, where

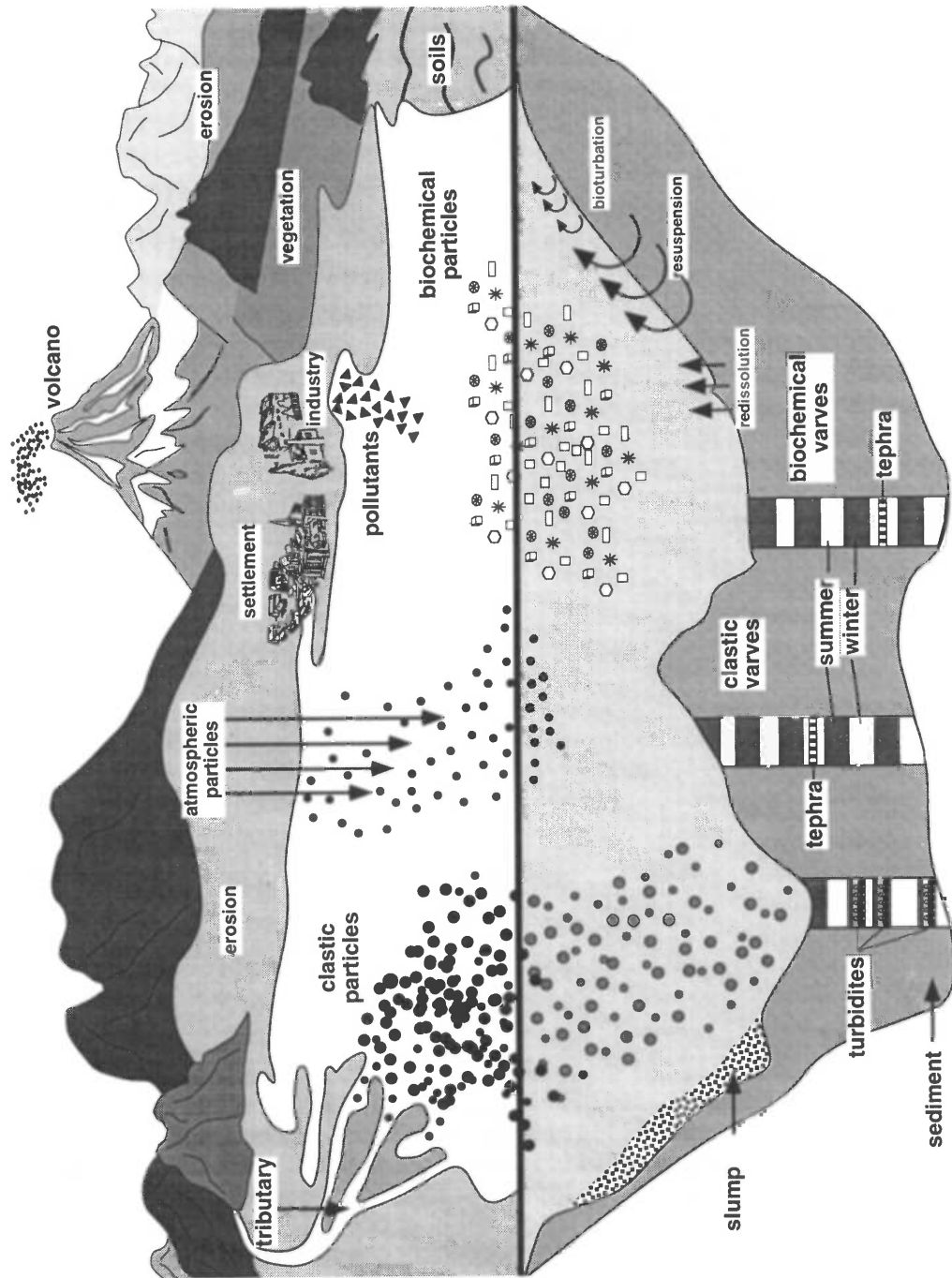


Fig. 1. Overview of the processes forming and altering sediments in lakes (after Sturm and Lotter 1995)

it is formed by such processes as weathering and glacial grinding. The dust is picked up by strong winds, and can reach the stratosphere, where it is transported over several thousand kilometers. *Ca.* 10% of the continents are covered with loess of different thicknesses. The largest loess areas are found in China, with thicknesses up to 200 m, covering a period of 2.5 Ma. The grain-size distribution peaks between 20 and 40 μ . Larger, heavy particles are removed quickly from the atmosphere.

After deposition, the loess is subject to internal changes depending on the climate conditions. During cold and dry periods, the accumulation rate is high, probably due to strong winds and relatively large source regions. Some reworking occurs through raindrop impact, surface wash and soil creep. These are only weak processes. During warm and wet periods, however, the primary loess is seriously modified by weathering, soil formation and diagenesis. Growing vegetation slowly transforms loess into soil, as indicated by change in color, smaller grain sizes and dissolution of CaCO_3 . Typical accumulation rates for loess deposits are centimeters per thousand years.

Loess represents an archive that is well-suited for studying glacial and interglacial periods, and for deriving information on wind speed and aridity/humidity. For resolution and time span, it is a comparable archive to deep-sea sediments, but far simpler and less expensive to collect (Liu 1985).

Loess Dating

Stratigraphic Methods

Magnetostratigraphy was one of the first methods to show that loess profiles cover several million years (Heller and Liu 1982). Magnetic reversals are very clearly recorded in loess profiles, their ages determined by the K-Ar or Ar-Ar methods on magma of volcanic eruptions. The disadvantage of magnetostratigraphic dating is the limited number of reversals, the youngest of which (B-M) is already 770 ka old. As reversals cannot be distinguished from each other, a complete analysis of the whole record is needed to identify them reliably.

The sequence of loess and paleosol layers can be correlated with glacial and interglacial periods, represented by dated $\delta^{18}\text{O}$ variations in deep-sea sediments. Dating by matching two records is a relatively imprecise technique and can easily lead to mismatches between glacial periods and loess layers. However, in combination with magnetostratigraphy, it may be useful, especially for the period 0–770 ka.

Two methods have been used to improve dating between glacial-interglacial transitions, and which can also relate to $\delta^{18}\text{O}$ deep-sea records. The first is based on the assumption of a constant atmospheric fallout of magnetic susceptibility. Changes of susceptibility in loess are explained by a varying dilution effect, which is proportional to the accumulation rate (Kukla *et al.* 1988). ^{10}Be measurements on the same samples, however, have shown that, during warm and humid periods, a significant part of the susceptibility is due to *in-situ* diagenetic processes. The concept of a constant fallout is therefore not valid (Beer *et al.* 1993).

A similar concept can be used for ^{10}Be ; its flux onto a loess plateau can be attributed to two main sources. The first is the local ^{10}Be fallout caused by cosmic-ray-induced spallation reactions in the atmosphere. The second source is the dust forming the plateau. Because the dust originates from the desert, where it has been exposed to atmospheric fallout for a long time, it contains high concentrations of ^{10}Be . Therefore, the total ^{10}Be flux onto the loess plateau is different for different climate regimes, and leads to increased concentrations during calm climate periods with low accumulation rates. A comparison of ^{10}Be concentration with the $\delta^{18}\text{O}$ record shows good agreement, allowing synchronization of the two records (Shen *et al.* 1992). Figure 2 provides a comparison of

the ^{10}Be concentration record from Xifeng, China (Beer *et al.* 1993) with the SPECMAP $\delta^{18}\text{O}$ deep-sea record.

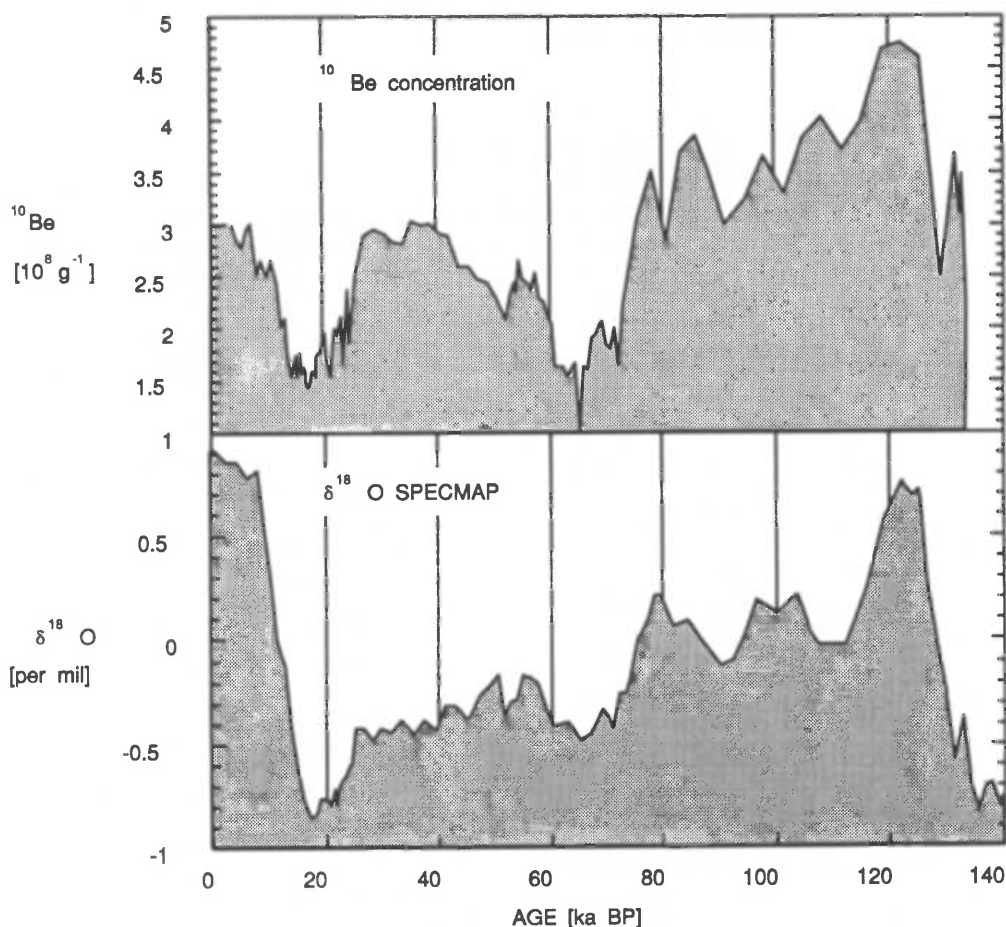


Fig. 2. Dating of the ^{10}Be concentration record of a loess profile from Xifeng, China by matching it to the SPECMAP $\delta^{18}\text{O}$ deep-sea record

Radioactive Decay

The most important dating method based on radioactive decay is ^{14}C dating. However, due to the relatively short half-life of 5730 yr, it is applicable only to the last *ca.* 40 ka. ^{14}C dating also requires well-defined carbon material of organic origin to guarantee that it reflects the atmospheric $^{14}\text{C}/^{12}\text{C}$ ratio. Only material that has never exchanged with other carbon (*e.g.*, CaCO_3) present in the loess should be used. Finally, one must be aware that ^{14}C ages have to be calibrated, and at present, the necessary calibration curve covers only the last 13 ka. Older ages may show a 1–2 ka uncertainty, due to changes of the atmospheric $^{14}\text{C}/^{12}\text{C}$ ratio.

Thermoluminescence (TL) dating has also been applied to loess from the last 100–200 ka. This method is based on several assumptions. During transport, dust is exposed to sunlight, which

removes the signal and, therefore, resets the clock. After deposition, the loess is radiated by cosmic rays and the primordial radioactive isotopes of U, Th and K and their daughters. The accumulated dose is assumed to be proportional to the exposure time, and the TL signal released by heating the sample proportionally to the dose. Because these assumptions may not always be fulfilled, not all of the published data are consistent (Wintle 1990).

CONCLUSION

Analyses of lake and loess sediments combined with information revealed from ice and deep-sea sediments provide a rather complete picture of the many complex, interactive, non-linear processes occurring in the past. Studies such as these are important for improving our models and for making realistic predictions about the future development of the environmental system.

ACKNOWLEDGMENT

We thank K. Farrenkothen for designing Figure 1 and R. C. Finkel for useful comments.

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RADIOCARBON UPDATES

16th International Radiocarbon Conference

Marian Scott announces the results of the ballot for the venue of the next radiocarbon conference:

Groningen, The Netherlands

A clear majority of votes were cast for Groningen. Congratulations and good luck to Wim Mook and Hans van der Plicht! We look forward to your gracious hospitality in 1997.

Advances in Liquid Scintillation Spectrometry 1997

The next LSC conference will be held in the Smokey Mountains, Tennessee, summer 1997. The co-organizers will be Gordon T. Cook, SURRC, Glasgow, Scotland, K. J. Hofstetter, Westinghouse Savannah River Laboratory, Aiken, South Carolina, USA, John E. Noakes, CAIS, Athens, Georgia, USA and Franz Schönhofer, The Austrian Society for Liquid Scintillation Spectrometry, Vienna.

New Radiocarbon Laboratories

A new laboratory has been installed with technical assistance from the IAEA:

Drs. M. A. Cristina Ures and Roberto Bracco

¹⁴C Laboratory

Faculty of Chemistry

Gral Flores 2124

Montevideo, Uruguay

Tel: 598 2 35 14 81

Fax: 598 2 94 19 06

Another new laboratory in Buenos Aires, Argentina:

Dr. J. Fernández-Niello

Laboratorio TANDAR

Comisión Nacional del Energía Atómica

Avenida del Libertador 8250

7429 Buenos Aires, Argentina

Tel: 54 1 754 7058

Although not brand new (we apologize for our delay in announcing this lab), an AMS laboratory was installed at Nagoya University:

Toshio Nakamura

Dating and Materials Research Center

Nagoya University

Chikusa

Nagoya 464-01 Japan

Tel: 81 52 789 2578

Fax: 81 52 789 3095

E-mail: g44466a@nucc.cc.nagoya-u.ac.jp

Passing of a Friend

We regret the sudden passing of our dear friend and colleague, Mieczysław F. Pazdur, Dean of the Faculty of Science at the Silesian Technical University, Gliwice, Poland and former head of the Radiocarbon Laboratory at the same institute. His wife, Anna, now heads the laboratory. She will prepare a memorial tribute for one of the next issues of *RADIOCARBON*.

New Manager

Dr. Peter Englert is the new Group Manager of the Nuclear Sciences Group of the Institute of Geological and Nuclear Sciences (IGNS), Lower Hutt, New Zealand. We wish all well in this promising arrangement, especially Rodger Sparks, who continues as Director of the Rafter Radiocarbon Laboratory.

From RADIOCARBON

*In Association with the American School of Prehistoric
Research, Peabody Museum, Harvard University*

**Late Quaternary Chronology and
Paleoclimates of the Eastern
Mediterranean**

Edited by OFER BAR-YOSEF and RENEE S. KRA

This sourcebook results from a workshop convened by the editors at the 14th International Radiocarbon Conference, 24 May 1991, in Tucson, Arizona. *Late Quaternary Chronology and Paleoclimates of the Eastern Mediterranean* brings together the results of varied radiometric dating techniques into one convenient reference. The volume includes: 1) discussions of TL, ESR and U/Th dating relevant to the hotly debated issues of the origins of modern humans and the fate of the eastern Mediterranean Neanderthals; 2) comprehensive compilations of radiocarbon dates encompassing the past 40,000 years, with special reference to the shift from foraging to agriculture and animal domestication, as well as critical re-evaluations of the available dates; 3) summaries of the paleoclimates of the area during the last 20,000 years as viewed through marine, continental, palynological and paleohydrological sequences. This 377-page book contains 23 articles by international scholars well-known in their respective fields.

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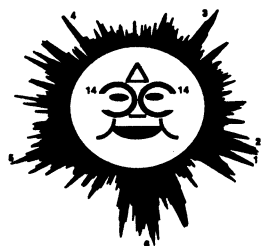
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From *RADIOCARBON*
An International Journal of Cosmogenic Isotope Research

CALIBRATION 1993

A Special Hardcover Edition

Edited by Minze Stuiver, Austin Long and Renee S. Kra

CALIBRATION 1993 amends and extends the time series published in the 1986 Calibration Issue. This special hardcover edition contains tree-ring-derived calibration curves for the radiocarbon time scale, a marine calibration curve employing U/Th coral data, and discussions of atmospheric and marine reservoir influences on measured ^{14}C ages. Calibration procedures are also reviewed.

A 5¼" diskette of the new IBM-PC based program, CALIB 3.0.3C (M. Stuiver and P. Reimer, University of Washington), is included. The program integrates the new atmospheric and marine data presented in this issue, and allows for calibrations from "conventional radiocarbon years" to calendar dates for the past 18,360 ^{14}C years.

CALIBRATION 1993 represents the state-of-the art calibration, and provides an essential tool for ^{14}C research and dating.

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LSC 92



VIENNA, AUSTRIA
14-18 September 1992

From **RADIOCARBON**
An International Journal of Cosmogenic Isotope Research

LIQUID SCINTILLATION SPECTROMETRY 1992

Edited by JOHN E. NOAKES,
FRANZ SCHÖNHOFER and HENRY A. POLACH

Liquid Scintillation Spectrometry 1992 contains papers presented at an international conference, "Advances in Liquid Scintillation Spectrometry", held in Vienna, Austria, 14-18 September 1992. The volume reports state-of-the-art research and technology in the field of liquid scintillation counting. The Methods section contains sample preparation and measurement techniques, scintillators and solvents, alpha measurements and standardization. The Bioscience Applications section is an overview of liquid scintillation spectrometry in molecular biology and implications of epidemiological studies. Environmental Applications include the use of tritium, radon, radium, uranium and other radionuclides in studies of radiation protection, tracer techniques and waste management. The editors are leading scientists from the USA, Austria and Australia, and the authors are international academic scholars and industrial researchers. The 512-page hardcover volume contains extensive bibliographical references and a comprehensive index. It was published in October 1993 by *RADIOCARBON*.

ISBN 0-9638314-0-2

LSC 94 – PROCEEDINGS OF THE INTERNATIONAL CONFERENCE GLASGOW, SCOTLAND 8-12 AUGUST 1994

Liquid Scintillation Spectrometry 1994 continues the series of conference proceedings, most recently from Glasgow. Themes include: New Instrumentation, Advances in Liquid and Solid Scintillators, Bioscience Applications, Environmental Applications, Alpha Counting, Cerenkov Counting, Data Handling Algorithms/Computer Applications and Software, and Sample Handling and Disposal, among others. This volume contains peer-reviewed articles covering a wide range of liquid scintillation topics. It will be available in Fall 1995.

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7th International Conference on Accelerator Mass Spectrometry

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Tel.: (520) 621-6810
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SECOND CIRCULAR – MAY 1995

THE SEVENTH INTERNATIONAL CONFERENCE ON ACCELERATOR MASS SPECTROMETRY will take place May 20 through May 24, 1996 in Tucson, Arizona. We expect about 150 participants, based on returns of the first circular. There will now be four associated workshops. Two specialized pre-conference workshops will be organized by Lawrence Livermore National Laboratory, and will focus on **^{129}I studies of AMS (May 16, 1996)** and **Biomedical ^{14}C Applications (May 17, 1996)**. A larger pre-conference workshop will take place on **May 18, 1996** at Scripps Institution of Oceanography in La Jolla, California and will focus on **Applications of AMS to Global Change**. We expect up to 75 people at the La Jolla workshop. The times of the workshops will not overlap, so it will be possible to travel from Livermore to San Diego to participate in the Global Change workshop. A post-conference workshop will be held in Tucson (May 25, 1996) on **Geological Applications of AMS**. Descriptions of the workshops are enclosed. There will also be a post-conference field trip. The **THIRD CIRCULAR** containing detailed conference and workshop information will be mailed in October 1995.

The AMS-7 conference will be at the **Westward Look Resort**, 245 East Ina Road, Tucson, AZ 85704, located in the foothills of the Santa Catalina Mountains. The conference rate for the rooms is expected to be \$106.50 including tax per room. Rooms are equipped with refrigerators and other amenities. The hotel has several pools, tennis courts, two restaurants, a lounge bar and other facilities. Details about hotel reservations and transportation will be sent with the **THIRD CIRCULAR**.

Evening and social activities start with a traditional **Mexican reception** on Monday evening. Wednesday is the day to relax, with an optional half-day Conference Outing to the Arizona-Sonora Desert Museum and San Xavier Mission on Wednesday morning. Wednesday afternoon is free for you to relax by the pool, go shopping, explore the surrounding area, or meet with colleagues. The banquet on Wednesday evening will feature regional fare. On **Thursday**, afternoon sessions will be held on the University of Arizona campus. We invite conference participants to **tour our laborato-**

ries on the University campus after sessions end on Thursday evening, after which we hope to have a small reception.

Registration information will be sent with the THIRD CIRCULAR. The registration fee will be \$285.00 before February 1, 1996, and \$325.00 after this date. **Conference Proceedings** will be published by Nuclear Instruments and Methods, Section B. Authors should bring their manuscripts to the meeting. Conference abstracts will be published by Radiocarbon. The conference abstracts and proceedings will be provided free to all registered attendees. The registration fee includes the conference volume, the opening reception on Monday evening, continental breakfasts and coffee-breaks, the banquet and to the UA campus on Thursday, laboratory tour and reception.

Travel information for participants in the pre-conference workshops. Sufficient travel time is allowed between the various pre-conference workshops and the main conference to all participants to travel from one location to the other. We will provide complete detailed travel information in the THIRD CIRCULAR, to be sent in October 1995. A FINAL CIRCULAR including Conference Program will be sent to registered participants in April 1996.

ABSTRACTS

Abstracts should be typed using 10 or 12 point font, to fit onto one page of letter-sized or A4 paper. Allow 2.5 cm for margins on either side. Please also provide an electronic version of your abstract either as an ASCII text file, WordPerfect or Microsoft Word file. We can handle either PC or Macintosh diskettes, but please identify this on your diskette. E-mail copies of abstracts are encouraged and should be sent to wbeck@physics.arizona.edu. Advance copies of abstracts are also encouraged, as this will allow us to devise the program. Please send final abstracts for all sessions and pre-conference workshops by Feb. 1, 1996 to: **AMS-7 Abstracts, NSF-Arizona AMS Facility, Physics Building, The University of Arizona, Tucson, AZ 85721, USA. Please make sure you indicate whether the paper is for a workshop or the main conference.**

DEADLINE FOR ABSTRACTS IS FEBRUARY 1, 1996

TRANSPORTATION

The workshop and conference locations and hotels can be conveniently reached by several means. Travel between the San Francisco Bay Area (for Livermore), San Diego and Tucson by air is frequent and inexpensive. Complete details of these and other travel possibilities will be included in the THIRD CIRCULAR. **Southwest, America West** and other airlines provide direct flights to Phoenix (*with very reasonable fares*) from Oakland, San Francisco and San Diego, with connections to Tucson International Airport via Phoenix, Las Vegas or Los Angeles. There are two non-stop flights per day on Southwest Airlines from San Diego to Tucson. Frequent bus and van shuttle service from Phoenix Airport to Tucson is also available. If there is sufficient interest, we may run a direct bus from the Scripps Institution to the Westward Look Resort in Tucson. The THIRD CIRCULAR will provide detailed information on all travel arrangements.

POST-CONFERENCE FIELD TRIP

Arrangements are underway for a **POST-CONFERENCE TOUR**. We have received over 30 responses showing interest in the post-conference tour. This tour will be limited to 45 participants. The trip will include four days/three nights, with overnight stops in Sedona, Flagstaff and northeastern Arizona. Other highlights of the trip will include a visit to prehistoric ruins at **Montezuma Castle, Flagstaff**, the impressive **Grand Canyon**, archaeological ruins and the volcanic cone of **Sunset Crater**, the beautiful **Canyon de Chelly** located on the Navajo Reservation on northeastern Arizona, **Hubbell's Trading Post, Walnut Canyon** with more evidence of the extensive Anasazi pueblo culture, **Oak Creek Canyon**, and the beautiful town of Sedona (for restaurants and shopping). We expect this tour to cost \$400 per person, double occupancy. We can arrange a roommate for you, but if you require a single room, the cost will be \$600.

For further information, contact us at the address below, or return the questionnaire sent with the first circular:

AMS-7 Conference Field Trip
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CONFERENCE WORKSHOPS

Please contact the organizers for further information

GLOBAL CHANGE

Applications of AMS to Global Change Research

Scripps Institution of Oceanography

La Jolla, California

May 18, 1996

There are many applications of AMS to **Global Change Research**. This workshop will be held at the Scripps Institution of Oceanography, on cliffs overlooking the **Pacific Ocean** in La Jolla, California. The workshop will allow new results and discussion of the importance of ^{14}C AMS dating, other radionuclides and related studies to the important problems of research in oceanography, atmospheric science, geology and other fields to the understanding of how our climate has changed in the past, and may change in the future. The conference hotels will be within easy reach of the Scripps Institution, restaurants and the beach. A highlight of the workshop will be a reception on Saturday evening, May 18, in the beautiful Scripps Aquarium, which is on a hill overlooking La Jolla and the Pacific Ocean. Lodging information will follow in the **THIRD CIRCULAR**.

Organizers:

A. J. T. Jull and J. W. Beck
The University of Arizona
NSF Arizona AMS Facility
Physics Building
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Tel.: (520) 621-6810
Fax: (520) 621-9619

POST-CONFERENCE WORKSHOP

Applications of AMS to Geological Applications

Westward Look Resort

Tucson, Arizona

May 25, 1996

There are many new applications of AMS to **Earth Sciences**. This workshop will focus on the uses of AMS measurements of different radionuclides for two particular areas: 1) seismic hazards studies and 2) studies of *in-situ*-produced radionuclides, which have much use in geomorphology, studies of uplift, etc. Other specialized applications to the earth sciences are also welcome. This workshop will be held at the Westward Look after the main conference. This will allow a relaxing day of less-formal discussions on these important topics. The workshop will allow new results and discussion of the importance of ^{14}C AMS dating especially to earthquakes and seismic hazards, other radionuclides and studies to the important problems in the development and use of *in-situ* cosmogenic radionuclides in geologic surfaces. Separate one-half-day sessions will be devoted to each of these two important topics.

The workshop will follow the main Conference on Saturday, May 25th. We look forward to an informative and informal discussion of advances and problems in geological applications.

Organizers:

Dr. George S. Burr
The University of Arizona
NSF-Arizona AMS Facility
Physics Building
Tucson, AZ 85721
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Dr. Susan D. Ivy-Ochs
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Institut für Mittelenergiephysik, HPK G13
CH-8093 Zürich, Switzerland
Tel.: 41-1-633-2042
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PRE-CONFERENCE WORKSHOPS AT LAWRENCE LIVERMORE NATIONAL LABORATORY

Please contact the organizers for further information

Lawrence Livermore National Laboratory (LLNL) is planning to host two separate pre-conference workshops. LLNL will make all arrangements for these two workshops.

WORKSHOP ON ^{129}I AMS

LLNL will host a topical workshop on ^{129}I studies by AMS on **Thursday, May 16, 1996**. This workshop will include talks about applications of the ^{129}I AMS technique. For more information about the ^{129}I AMS workshop please contact:

Mark Roberts
L-397, LLNL
P.O. Box 808
Livermore, CA 94551 USA

Tel.: (510) 422-3431
Fax: (510) 423-7884
E-mail: roberts5@llnl.gov

WORKSHOP ON BIOMEDICAL ¹⁴C AMS

The second LLNL workshop will be on biomedical ¹⁴C AMS. This workshop will include talks on sample preparation and handling problems unique to biomedical ¹⁴C AMS. This workshop is scheduled for **Friday, May 17, 1996**. For more information about the biomedical ¹⁴C AMS workshop please contact:

John Vogel
L-397, LLNL
P.O. Box 808
Livermore, CA 94551 USA

Tel.: (510) 423-4232
Fax: (510) 423-7884
E-mail: vogel2@llnl.gov

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AMS-7 CONFERENCE, TUCSON, ARIZONA, MAY 20-24, 1996

Request for further information

Please return this form only if you did not return the questionnaire in the first circular. If you need further information, or did not return the first circular questionnaire, please return this form by October 1, 1995 in order to receive the third mailing.

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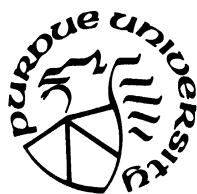
If you are interested in the one-day workshops, please contact the organizers indicated for further information.

I will attend AMS-7 _____ I am interested in the post-conference field trip: _____

Faculty Position – *GEOSCIENTIST (AMS)*

Purdue University

*Department of Earth and
Environmental Sciences*



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NOTICE TO READERS AND CONTRIBUTORS

The purpose of *RADIOCARBON* is to publish technical and interpretive articles on all aspects of ^{14}C and other cosmogenic isotopes. In addition, we present regional compilations of published and unpublished dates along with interpretive text. Besides the triennial Proceedings of Radiocarbon Conferences, we publish Proceedings of conferences in related fields and Special Issues that focus on particular themes. Organizers interested in such arrangements should contact the Managing Editor for information.

Our regular issues include NOTES AND COMMENTS, LETTERS TO THE EDITOR, RADIOCARBON UPDATES and BOOK REVIEWS. Authors are invited to extend discussions or raise pertinent questions regarding the results of investigations that have appeared on our pages. These sections also include short technical notes to disseminate information concerning innovative sample preparation procedures. Laboratories may also seek assistance in technical aspects of radiocarbon dating. We include a list of laboratories and a general index for each volume.

Manuscripts. When submitting a manuscript, include three printed copies, double-spaced, and a floppy diskette, single-spaced. We will accept, in order of preference, FrameMaker, WordPerfect 6.0 or 5.1, Microsoft Word, Wordstar or any standard IBM word-processing software program on 3½" or 5¼" IBM disks, or high-density Macintosh diskettes. ASCII files are also acceptable. We also accept E-mail and ftp transmissions of manuscripts. Papers should follow the recommendations in INSTRUCTIONS TO AUTHORS (1994, vol. 36, no. 1). Offprints of these guidelines are available upon request. Our deadlines for submitting manuscripts are:

<i>For</i>	<i>Date</i>
Vol. 38, No. 1, 1996	September 1, 1995
Vol. 38, No. 2, 1996	January 1, 1996
Vol. 38, No. 3, 1996	May 1, 1996

Half-life of ^{14}C . In accordance with the decision of the Fifth Radiocarbon Dating Conference, Cambridge, England, 1962, all dates published in this volume (as in previous volumes) are based on the Libby value, 5568 yr, for the half-life. This decision was reaffirmed at the 11th International Radiocarbon Conference in Seattle, Washington, 1982. Because of various uncertainties, when ^{14}C measurements are expressed as dates in years BP, the accuracy of the dates is limited, and refinements that take some but not all uncertainties into account may be misleading. The mean of three recent determinations of the half-life, 5730 ± 40 yr, (*Nature*, 1962, vol. 195, no. 4845, p. 984), is regarded as the best value presently available. Published dates in years BP can be converted to this basis by multiplying them by 1.03.

AD/BC Dates. In accordance with the decision of the Ninth International Radiocarbon Conference, Los Angeles and San Diego, California, 1976, the designation of AD/BC, obtained by subtracting AD 1950 from conventional BP determinations is discontinued in *RADIOCARBON*. Authors or submitters may include calendar estimates as a comment, and report these estimates as cal AD/BC, citing the specific calibration curve used to obtain the estimate. Calibrated dates should be reported as "cal BP" or "cal AD/BC" according to the consensus of the Twelfth International Radiocarbon Conference, Trondheim, Norway, 1985.

Measuring ^{14}C . In Volume 3, 1961, we endorsed the notation Δ , (Lamont VIII, 1961), for geochemical measurements of ^{14}C activity, corrected for isotopic fractionation in samples and in the NBS oxalic-acid standard. The value of $\delta^{14}\text{C}$ that entered the calculation of Δ was defined by reference to Lamont VI, 1959, and was corrected for age. This fact has been lost sight of, by editors as well as by authors, and recent papers have used $\delta^{14}\text{C}$ as the observed deviation from the standard. At the New Zealand Radiocarbon Dating Conference it was recommended to use $\delta^{14}\text{C}$ only for age-corrected samples. Without an age correction, the value should then be reported as percent of modern relative to 0.95 NBS oxalic acid (Proceedings of the 8th Conference on Radiocarbon Dating, Wellington, New Zealand, 1972). The Ninth International Radiocarbon Conference, Los Angeles and San Diego, California, 1976, recommended that the reference standard, 0.95 NBS oxalic acid activity, be normalized to $\delta^{13}\text{C} = -19\text{‰}$.

In several fields, however, age corrections are not possible. $\delta^{14}\text{C}$ and Δ , uncorrected for age, have been used extensively in oceanography, and are an integral part of models and theories. Thus, for the present, we continue the editorial policy of using Δ notations for samples not corrected for age.

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