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# Radiocarbon

An International Journal of Cosmogenic Isotope Research



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# FROM THE EDITOR

The reader will notice that despite our previous announcement (Long 1995) of *RADIOCARBON*'s discontinuance of date-list publication, this issue contains four date lists, plus one article that is a compilation of dates with text related to a specific research topic. What is going on here? Do our actions belie our words? I prefer to call it institutional flexibility.

In view of the exploding volume of radiocarbon dates in recent years, mostly AMS-based, neither this nor any journal can be a repository of all dates. Individual laboratory databases and research publications are now the primary <sup>14</sup>C date archives. My back issues of *RADIOCARBON* have often been a valuable resource for dates that had been published only here. These date lists also revealed important details of laboratory techniques and pretreatment procedures that were often omitted in application-focused publications. In some cases, date lists served as formal announcements of the initiation of new <sup>14</sup>C dating facilities and personnel. When we received the date list submissions in this issue, we realized that nearly 50 years of tradition is hard to break. Moreover, we recognize the importance of providing a showcase for relatively new laboratories to demonstrate their capabilities and application interests.

Thus, we now find ourselves in a transition state. We cannot predict how the date-list situation will sort out. We do know that fewer date lists arrive in our mailbox. We also know that establishing a comprehensive, single-site <sup>14</sup>C database is not feasible. One possibility is that we could publish the web site address for each laboratory whose directors wish to make available details of their techniques and personnel. They could even advertise here the availability of online, searchable <sup>14</sup>C databases for dates they have produced. Meanwhile, recognizing that these are changing times, we will publish date lists at the discretion of the editors.<sup>1</sup>

Please contact us here with feedback about how we (the  ${}^{14}C$  community) should deal with the future of  ${}^{14}C$  data information.

Austin Long

# REFERENCE

Long, A. 1995 From the Editor. Radiocarbon 37(3): iii.

<sup>1</sup>This reintegration of date lists also means that we are discontinuing our separate publication of *Radiocarbon Date Lists*, whose 1995 and 1996 issues were available as computer files only.

iv From the Editor

# ERRATUM

Our annual laboratory list entry for the Gif beta-counting <sup>14</sup>C laboratory, published in *RADIOCARBON* Volume 38, Number 3 (1996), p. 620, had incorrect contact information for the lab director. The correct entry is:

Gif Dr. Michel Fontugne Centre des Faibles Radioactivités Laboratoire mixte CNRS-CEA F-91198 Gif sur Yvette Cedex, France Tel: +33 1 69 82 35 25; Fax: +33 1 69 82 35 68 E-mail: Michel.Fontugne@cfr.cnrs-gif.fr and Laboratoire Souterrain de Modane Laboratoire mixte IN2I 3-CNRS/DSM-CEA 90, Rue Polset F-73500 Modane, France

Our apologies to Dr. Fontugne for inadvertently "retiring" him!

# Radiocarbon

1997

# RADIOCARBON AGES OF MAMMOTHS IN NORTHERN EURASIA: IMPLICATIONS FOR POPULATION DEVELOPMENT AND LATE QUATERNARY ENVIRONMENT

# YURIJ VASIL'CHUK,<sup>1</sup> JAAN-MATI PUNNING<sup>2</sup> and ALLA VASIL'CHUK<sup>3</sup>

ABSTRACT. Many mammoth remains have been radiocarbon-dated. We present here more than 360 <sup>14</sup>C dates on bones, tusks, molars and soft tissues of mammoths and discuss some issues connected with the evolution of mammoths and their environment: the problem of the last mammoth; mammoth taphonomy; the plant remains and stable isotope records accompanying mammoth fossils; paleoclimate during the time of the mammoths and dating of host sediments. The temporal distribution of the <sup>14</sup>C dates of fossils from the northern Eurasian territory is even for the entire period from 40 to 10 ka BP.

#### INTRODUCTION

Mammoth remains are very valuable objects for the study of Late Quaternary geochronology and paleoecology. The first finds of mammoth remains on northern islands in the Arctic and valleys of the great Siberian rivers drew the attention of scientists to the northern territories more than two centuries ago. The development of the mammoth population is one of the most interesting problems in reconstructing the dynamics of the environment during the Quaternary. The wide use of radiocarbon and paleoecological methods has provided valuable information on these dynamics. Today it is widely accepted that various fossil species of the genus *Mammuthus* characterize Late Quaternary periglacial environments.

Since the <sup>14</sup>C method was first applied to the age determination of mammoth remains, the main problem has been reliability of the data. Geochemically, the most desirable materials for <sup>14</sup>C analysis are well-preserved organic residues, *i.e.*, bones rich in collagen, frozen carcasses, cud, dung or stomach contents from frozen ground or dry caves. In Russia, there are numerous sites from which whole carcasses of fossil mammoths have been dated by <sup>14</sup>C (Fig. 1, Appendix): Yuribey River (Gydan Peninsula), Gydan (Gyda River), Pyasina River (Taimyr Peninsula), Mochovaya River (Taimyr Peninsula), Mammoth Shrenk (Taimyr Peninsula), Chekurovka Settlement (Lower Lena River), Bukovskiy (Lena River), Beryosovka River, Shandrin River, Kirgilyakh River (baby mammoth "Dima"), Lyakhovskiy Bol'shoi Island, Tirekhtyakh River, Enmynveem (Chukotka Peninsula).

It has been established that woolly mammoths spread over a vast area in northern Eurasia in the Late Pleistocene and even into the Holocene. The first <sup>14</sup>C measurement of the fossil remains of mammoths was carried out by Heintz and Garutt (1965). Sulerzhitskij (1995) published more than 180 dates of Late Quaternary mammoths, most of which he had collected and dated himself. Stuart (1991) summarized the majority of <sup>14</sup>C dates of mammoths of various species from northern Eurasia and North America.

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**METHODS** 

We sampled (Vasil'chuk 1992) mammoth bones from many reference sites in northern Yakutia: in the depression near Kular settlement (37.7 ka BP), near Zelyonyj Mys settlement (43.7 and >50 ka BP), Duvannyi Yar natural exposure (28.6, 33.8, 34.7 and >50 and 53 ka BP), and in Chukotka in the Mayn River valley and Ledovyi Obryv natural exposure (15.1 ka BP). We found redeposited bones (drifted fossils) in the Holocene alas sediments, in the Kolyma River valley near the Omolon River mouth (15 ka BP).

Earlier we summarized a number of <sup>14</sup>C dates on mammoth remains from northern Asia in order to reconstruct permafrost evolution during the last 40 ka (Vasil'chuk 1992). In addition to that list, we here summarize (Appendix) all the known <sup>14</sup>C dates (>360) of mammoth remnants from northern Eurasia, mainly from permafrost areas (Appendix and Fig. 1).

One serious concern in using data obtained by dating of mammoth remains is its reliability. The systematic checking of bone data is underway at the Radiocarbon Laboratory of the Geological Institute in Moscow (L. Sulerzhitskij, personal communication). It is possible to draw some conclusions using the data obtained by multiple analysis of material from the same layer. There are many samples in the Appendix dated by bone material with different degrees of weathering (samples 12, 127, 139, 156, 246). The differences in the data are <1000 yr, in most cases within the statistical uncertainty of the dates. In some cases, different materials from the same layers were dated (Appendix, samples 85, 86, 87; 170, 171; 210, 211; 265, 266; 268,269; 271, 272). The differences in data are modest and also within the limit of statistical uncertainty. This demonstrates that, in principle, fossils of mammoth fauna can be considered reliable as material for <sup>14</sup>C dating.

The dates in the interval from 10 to 40 ka BP are distributed rather evenly; furthermore, there is no remarkable spatial grouping of mammoth finds in northern Eurasia. If <sup>14</sup>C dates are an unbiased sample of mammoth populations, this indicates that mammoths lived constantly and continuously over northern Eurasia during the Middle/Late Würm. Eastern Europe is characterized by a series of dates from 9.7 to >47.7 ka BP. The finds consist predominantly of molars, bones and tusks. In the northwestern part of European Russia, mammoth remains have been dated from 18.3 to >36 ka BP. In southern parts of western Siberia the dates range from 12.8 to 41.9 ka BP.

Whole carcasses have been found in the Yuribey and Gyda River valleys in northwestern Siberia; mammoth remains from this region have been dated from 9.6 to 35 ka BP. On the Taimyr Peninsula, Sulerzhitskij (1995) obtained several dates ranging from 9.6 to >53 ka BP without any significant time gaps. In central Siberia, fewer dates cover the interval from 20.7 to 49.7 ka BP (Appendix); this reduction may be explained by the topography and, consequently, biogeographical conditions of this mountainous region. The abundant <sup>14</sup>C dates on mammoth remains in northern Yakutia range from 10.3 to >53 ka BP. From the Magadan region, data are available on the carcass of the Kirgilyakh baby mammoth and a few other finds, with ages from 21.6 to 41 ka BP. The Chukotka Peninsula is characterized by dates from 14.3 to 32 ka BP. In the Kamchatka Peninsula, <sup>14</sup>C dates were obtained in the interval 12.6 to 40 ka BP (Sulerzhitskij 1995) (Appendix). Interesting dates are available from Arctic islands, especially Wrangel Island. Dates obtained from a dwarf form of mammoth are the youngest for mammoth finds in the world, falling into the interval 3.9 to 7.7 ka BP. The dates from 12.7 to 20 ka BP from these islands belong to mammoths of normal form (Vartanyan, Garutt and Sher 1993; Vartanyan *et al.* 1995). The oldest date on mammoth remains from the Arctic islands is 32 ka BP.

Mammoth bones also occur rather often in western Europe (Kurten 1968). Berglund *et al.* (1976) and Hakansson (1976) refer to some <sup>14</sup>C dates on mammoth remains from South Sweden: 13, 19, 22

# 4 Y. Vasil'chuk, J.-M. Punning and A. Vasil'chuk

and >30 ka BP. In Norway, some <sup>14</sup>C dates on mammoth remains in the time interval from 19 to 32 ka BP have been obtained (Follestad and Olsson 1979). In Finland, Jungner and Sonninen (1983) and Donner, Jungner and Kurten (1979) obtained three <sup>14</sup>C dates on samples collected by Donner: 15.5, 25.2 and >43 ka BP. Mammoth remains from Denmark have <sup>14</sup>C ages from 13 to >39 ka BP (Aaris-Sorensen *et al.* 1990). In Germany, the dates vary from 15.8 to 30.3 ka BP; in Switzerland, from 12.2 to 34.6 ka BP; in Poland, from 20.2 to 23.0 ka BP; and in France, from 12 to 25.8 ka BP. Mammoth remains from Great Britain and Ireland have ages from 11.6 to >39.5 ka BP (Coope and Lister 1987; Stuart 1991).

The temporal distribution of mammoth remains (Fig. 2) displays no essential differences in the number of dated mammoths in the time interval from 15 to 45 ka BP. The spatial distribution of <sup>14</sup>C-dated mammoth remains shows that mammoth fauna were connected mainly with Late Quaternary permafrost conditions (Fig. 1).

# DISCUSSION

Several issues connected with mammoth evolution are of great scientific interest: the problem of the last mammoth; mammoth taphonomy; paleoclimate during the time of the mammoths and use of mammoth remains for the dating of host sediments.

# The Problem of the Last Mammoth

The youngest <sup>14</sup>C data for quasi-mammoth remains from the Western Hemisphere are those from North America on the *Mammut americanum*: 4470  $\pm$  160 (M-2436) from Kuhl, Michigan; 8910  $\pm$  150 (GSC-614) from Ferguson Farm, Ontario, and 9568  $\pm$  1000 BP (M-282) from Lenawee, Michi-



Fig. 2. The temporal distribution of <sup>14</sup>C-dated mammoth remains

gan (Meltzer and Mead 1985); and from the Southwest United States, remains of Mammuthus columbi from Sandy, Utah dated to  $5985 \pm 210$  (SI-2341b),  $7200 \pm 190$  (RL-464) and  $8815 \pm 100$  BP (SI-2341a) (Semken 1983; Stafford *et al.* 1987; Agenbroad and Mead 1989). However, contamination of samples cannot be excluded, as has been shown by Stafford *et al.* (1987)—for different fractions from the Domebo mammoth sample, dates from 2050 to 11,490 BP were obtained. We also note that no archaeologically associated remains of mammoths younger than Clovis (*ca.* 11,000 BP) (Haynes 1993) have been replicated. It seems that the younger <sup>14</sup>C dates on mammoth remains in Northern America need replication.

More than 20 mammoth remnants from Wrangel Island collected by Vartanyan (Vartanyan, Garutt and Sher 1993; Vartanyan *et al.* 1995) were dated in the range 4–7 ka BP; the youngest dates are 3730  $\pm$  40 BP (LU-2741) and 3920  $\pm$  30 (GIN-6980). Two samples were replicated by the AMS dating facility at the University of Arizona (Long, Sher and Vartanyan 1994). Thus, the last mammoths now known lived on Wrangel Island *ca.* 4000 yr ago. The dated remains belong to a dwarf form of a mammoth that lived in isolation on the island under strictly limited food conditions (Grant 1985). One problem connected with these phenomena is that mammoths came to the island as a consummate form (Johnson 1978), but no data exist for the interval 12–8 ka BP. The mammoth refugium must have been disconnected from the continent, otherwise remains of Holocene dwarf mammoths should be found on the continent also. The dwarf form on Arctic islands is a common survival adaptation of large mammalians, such as deer, hippopotamus and elephantids. (Dwarf forms of elephantids lived on some islands of the Mediterranean Sea and on the Channel Islands of California during the Quaternary, with heights of  $\leq 0.9$  m.)

### **Mammoth Taphonomy**

Two aspects of the taphonomy of mammoth remains deserve attention. First, bones occurring in an unarticulated condition, almost without exception, indicate the redeposition of remains, typical of deposits of fluvial origin (such as alluvial, lacustrine, fluvioglacial). Therefore, as a rule the <sup>14</sup>C dating of mammoth remains from these sediments gives the maximum age of sediment formation. Inclusion of younger bones in an older frozen deposit is not possible. As for syngenetic sediments of fluvial series, we can state with certainty that their ages are younger or equal to the ages of the enclosed bones. Second, the possible delivery of the bones by carnivores must be kept in mind.

The burial of whole mammoth carcasses is obviously an infrequent process, requiring the coincidence of several conditions. To remain intact, a carcass must be covered with sediments or be isolated from carnivores very quickly. Hence, the best conditions for preserving mammoth remains were offered by talus and alluvial sediments, high icy terraces and thermokarst depressions.

Plant remains in the stomach can be used to establish the season of a mammoth's burial. All the mammoth carcasses found belonged to mammoths that perished in the summertime. Remains of mammoths that perished in the winter may have been destroyed by carnivores. This suggests that an important factor for the preservation of mammoth carcasses is the existence of permafrost conditions, during both fossilization and preservation.

# Plant Remains and Pollen Accompanying Mammoth Fossils

The majority of the finds of subfossil mammoths and other large animals in northern Eurasia are connected with polygonal ice wedge complexes. Possibly mammoths' pasturage depended directly on polygonal massifs. Palynological and plant macrofossil analyses have revealed an abundance of herbage in polygonal relief areas.

Pollen grains and plant remains in mammoths' guts (Table 1) indicate the feeding habits of the animals (Sukachev 1914; Solonevich, Tikhomriov and Ukraintseva 1977; Ukraintseva 1979; Sokolov 1982; Shilo *et al.* 1983; Guthrie 1990).

TABLE 1. Pollen and Spores in the Stomach Content of the Carcasses of Four Fossil Mammoths and a Selerican Horse (% of total content) (after Tikhomirov and Kupriyanova 1954; Kupriyanova 1957; Belorusova, Lovelius and Ukraintseva 1977; Belaya and Kisterova 1978; Ukraintseva 1979; Gorlova 1982)

Site (see Appendix)	<sup>14</sup> C age (BP), number of samples	P <sub>tr</sub> *	P <sub>sr</sub>	P <sub>hr</sub>	S
Yuribey River (Gydan Peninsula)	10 ka (4)	5–6	15-17	17–20	56-63
Shandrin River (northern Yakutia)	36 ka (1)	2	1	19	77
Kirgilyakh River (Magadan Region)	39 ka (6)	2–10	6–20	60–77	8–18
Beryosovka River (northern Yakutia)	44 ka (1)	1	1	97	1
Bolshoy Selerican (Indigirka River)	38 ka (1)	4	5	80	11

 ${}^{*}P_{tr}$  = tree pollen,  $P_{sr}$  = shrub pollen,  $P_{hr}$  = herb pollen, S = spores.

The host sediments that enclosed the Kirgilyakh baby mammoth are characterized by the predominance of pollen of herbs and shrubs (60–77%). Pollen of grass and sedges occur in equal quantities (10–25%), and other grasses are represented by 28 families such as Ranunculaceae (2–4%), Cruciferae (4–10%) and Artemisia (up to 4%). There are also pollen of hydrophilous taxa such as Potamogeton, Myriophyllum and Alismataceae. The content of Ericaceae (<2%), which is usually dominant in subfossil pollen spectra, is very small. The presence of larch pollen is evidence of larch forest at that time (Belaya and Kisterova 1978).

Pollen analyses of mammoths' digestive tracts and host sediments show a predominance of herb pollen or spores, presence of larch pollen (1-5%) and pollen of species that now live in southern areas (*e.g.*, pollen of *Ribes, Betula* sect *Albae*), and the existence of typical tundra elements in the vegetation, *e.g.*, *Dryas punctata*. The pollen and spores spectra showed some regional features, but these were evidence that mammoths lived in environments close to the modern larch forest and forest tundra.

Fossil flora found in the remains of the Yuribeyskiy mammoth (Gorlova 1982) consist of Cyperaceae (9 species), Poaceae (4 species), Salicaceae (3 species), Rosaceae (2 species), Betulaceae, Ericaceae and Pinaceae (1 species each). The present vegetation in the Arctic and the Subarctic is rather similar. However, the presence of macrofossils of *Larix sibirica* L. and *Ribes* spp. testifies to more favorable climatic conditions during the era of mammoths.

Naturally, to some extent, the content of the stomach reflects feeding preferences of mammoths. Plant remains belong to different life forms—shrubs, grasses, moss—and different habitats—dry meadows, steppe slopes, bottomland meadows, floodplain swamps, *etc.* This suggests the diversity of environmental conditions and biocoenoses in the areas where mammoths could live. Willow brushwood occurred in closed valleys, cereals and herbage occupied southern slopes of alluvial terraces, and sedges (particularly *Carex* strains) grew in meadows.

Analyses of both plant microfossils and pollen from dung and gut contents indicate a summer diet of grasses, sedges, mosses, and shoots of willow, draft birch and alder. A frozen forage mass from the stomach and gut of the Shandrin mammoth, found in Lower Indigirka, weighed *ca.* 250 kg. The

greater part of it consisted of stems and leaves of sedges, grasses and cotton-grass and the smaller part of sprouts of willow, birch and alder (Solonevich, Tikhomirov and Ukraintseva 1977; Vereshchagin 1979). There were not any ripe seeds that might testify that the animal died in summer.

Well-preserved contents of mammoths' digestive tracts have been studied from the Shandrin mammoth carcass (Ukraintseva 1979). The remains of Cyperaceae, *Eriophorum* spp., Poaceae, *Larix Daurica* Turcz., Ericaceae, *Vaccinium vitis-idaea* L., some species of *Polytrychum*, *Aulacomnium* and *Sphagnum* have been identified. The main part of the pollen spectra from the enteron consists of spores of *Bryales* and *Sphagnum* (77%). Pollen of grasses makes up 19.4%, the majority of them pollen of grasses and sedges. Pollen of *Dryas punctata*, *Valeriana capitata*, *Artemisia* spp., *Ledum* spp. and *Saxifraga* spp. are also represented. All of these species presently grow in the same area. Pollen of larch, birch and alder bush were also found (*ca.* 4%). Nowadays analogous landscapes of larch light forests occur *ca.* 200 km to the south.

Sukachev (1914) identified grasses and sedges with ripe seeds in the stomach of Beryosovka mammoth, which evidently perished in late summer. Kupriyanova analyzed pollen and spores remains from the stomach of this mammoth: 8198 pollen grains and 7 spores were found (Tikhomirov and Kupriyanova 1954; Kupriyanova 1957). These are pollen of cereals (97%), forbs (2%), trees (1%) and spores. The pollen spectra are affected by the time of the mammoth's death (second half of summer), showing a small amount of tree pollen, with predominance of the cereal pollen blooming during that period. The species list of the pollen from the mammoth's stomach evidenced a varied flora corresponding to biocoenoses that exist at present *ca*. 1000 km to the south. Species of bunchgrass steppe with forbs and wormwood (*Artemisia*), upland meadow, inundation meadow, salt meadow and herb meadow have been determined.

Zaklinskaya studied the pollen content in the Taimyr Peninsula mammoth host sediments. All pollen spectra were characterized by the predominant herb pollen. The main part of the pollen consists of herbs of meadow plant communities of polygonal tundra, with cereals and sedges dominating (Zaklinskaya 1954). The paleobotanical and palynological data evidenced no sharp changes in the vegetation features, which therefore cannot be a cause of the mammoths' extinction.

One of the details of paleobotanical characteristics was obtained from Selerican horse remains (Belorusova, Lovelius and Ukraintseva 1977; Ukraintseva 1979). Plant remains are represented by fossils of poplar, birch and mosses. Pollen spectra show a presence of hazel, juniper, spruce and elm in plant societies and *Kobresia capilliformis* as a dominant of dry meadows.

Lister and Sher (1995) pointed out that one problem of the climatic model of extinction is explaining how woolly mammoths survived an earlier interglacial. They proposed that the vegetation of the interglacial differed from that of the Holocene. In Siberia several interglacials have been recorded, but during these intervals, the vegetation differed from the vegetation of the modern larch-dominated taiga. Unfortunately, even now the climate-driven models do not show uniquely the reasons for the extinction of the mammoth population. Undoubtedly, in many cases, human involvement was important (Stuart 1991).

The pollen and plant fossils in the sediments accompanying the mammoth remains and the content of their stomachs show that the favorable season for mammoths' fossilization in permafrost areas was late summer. More detailed paleoclimatic information about the time of mammoths' existence can be provided by stable isotope data both from syngenetic sediments with ice wedges and directly from mammoth remains.

# **Isotope Records**

Variations of the oxygen isotope composition are not substantial in the Late Pleistocene permafrost syngenetic deposits where mammoth remains have been found. The interval of  $\delta^{18}$ O in the syngenetic ice wedge that formed 40–10 ka ago (Table 2) varies in the north of western Siberia from -24 to -21‰, in northern Yakutia from -34 to -29‰ and in northern Chukotka from -32 to -29‰ (Vasil'chuk 1992). As snow meltwater was the main source of the moisture for ice wedges, the oxygen isotope records reflect mainly winter precipitation temperatures.

TABLE 2. Oxygen Isotope Composition in Syngenetic Ice Wedges ( $\delta^{18}$ O, ‰), Mean Winter ( $t_{mw}$ ) and Mean January Temperatures ( $t_{mJ}$ ) 40–10 ka BP in Different Regions of Siberian Mammoths' Habitats (after Vasil'chuk 1992, 1993)

Region of Siberia	δ <sup>18</sup> Ο (‰)	t <sub>mw<sub>D, J, F</sub></sub> (°C)	$t_{mJ}$ (°C)*
Yamal and Gydan Peninsulas,	-21 to -24	-21 to -24	-32 to -36
north of western Siberia			(-22 to -28)
Bykovsky Peninsula, mouth of Lena River,	-30 to -34	-30 to -34	-45 to -49
western Yakutia			(-32 to -34)
Upper Kolyma River,	-29 to -33	-29 to -33	-43 to -49
northeastern Yakutia			(-30 to -34)
Vilyui and Aldan Rivers,	-29 to -31	-29 to -31	-43 to -46
central Yakutia			(-37 to -45)
Ayon Island,	-29 to -32	-29 to -32	-44 to -46
northern Chukotka			(-27 to -29)
Anadyr' and Mayn Rivers,	-21 to -29	-21 to -29	-31 to -43
southern Chukotka			(-21 to -27)

\*Present mean January temperatures in parentheses for comparison

Using the relationship between the mean winter temperatures and oxygen isotope records obtained by Vasil'chuk (1992), it is possible to say that the winter climate was cold, stable and unchangeable in northern Eurasia from 40 to 10 ka ago.

# Seasonal Climatic Conditions During the Time of the Mammoths

Mammoth remains are usually treated as indicators of very cold climatic conditions. The extinction of mammoths is one of the most often discussed problems in the paleogeography of the Late Quaternary. One of the most important causes of their extinction is connected with the change of climatic conditions and, therefore, the composition and production of biomass. The paleotemperature record obtained immediately from mammoth habitats, *i.e.*, detailed records of syngenetic ice wedges, which have been dated as Late Pleistocene and Early Holocene, give valuable information about the mammoths' environment. The oxygen isotope and pollen data from the same sections make it possible to reconstruct separately winter and summer temperatures. This approach is of great significance. Comparing the trends of winter and summer temperatures, we can see that winter temperatures changed especially abruptly at the Pleistocene/Holocene boundary.

An analysis of all information obtained indicates that mammoths were excellently adapted to Late Pleistocene long and cold winters without any thaws. They had long, shaggy coats and underwool, a thick layer of subcutaneous fat, tiny ears and short tails. Underwool is characterized by thick hair, which was four times thicker than that of present-day cold-adapted animals. Mammoths did not have adipose glands in their skin, so their wool would get wet when it was raining or foggy. Large tusks came in handy for scraping snow and ice both for drinking purposes (like present-day elephants) and to expose buried forage. Their large size and spreading cushioned feet on which they distributed their weight may have enabled them to cope with snow better than most large herbivores in arctic and subarctic environments. The survival of such large animals in regions with a marked seasonal temperature range requires not only abundant summer herbage but also large quantities of winter feed, probably including dead grasses and bark from shrubs and trees.

At the Pleistocene/Holocene transition, winter temperatures changed sharply. Increasing Atlantic influences caused an increase in winter temperatures and the appearance of winter thaws. If thaws occurred in February or March, they could be fatal for mammoth herds, because the resulting multilayered ice crust made it impossible to find food and to move. The animals could not move because their legs were adapted to friable and relatively shallow snow cover but not to multilayered ice crust. Moreover, mammoth hair would have quickly become covered with ice, making the animals look like terrestrial icebergs.

We have reconstructed the environment in the mammoths' time applying different methods. The oxygen isotope data in ice wedges enabled us to determine that during the interval from 40 to 10 ka BP, mean January temperatures in northern Siberia were *ca.*  $8-12^{\circ}$ C lower (in Chukotka up to 17–18°) than the modern ones (Table 2). We have established that the interval from 40 to 10 ka BP was a single cryochron (Vasil'chuk 1992, 1993) with severe winters when the oscillations of temperatures were rather small, thawing was rare and the snow cover, as a rule, quite friable. Such winters permitted mammoths to dig out the grass easily from under the snow. Interpretation of pollen data enabled us to reconstruct the mean July temperatures for the period 40–10 ka BP. They were *ca.*  $1-4^{\circ}$ C lower than the contemporary ones (almost 7°C lower in the Chukotka Peninsula). During short periods of warming, the July temperatures could have been by  $1-3^{\circ}$ C higher than modern ones (Vasil'chuk and Vasil'chuk 1995).

#### Use of Mammoth Fossils for Dating the Host Sediments

Mammoth remains have been used widely in the dating of host sediments. The high degree of validity of <sup>14</sup>C dates of mammoth bones (Sulerzhitskij 1995), enables us to determine the lower limit of the host sediment age. In several sequences (Duvannyj Yar, Zelyonyj Mys and Kular in northern Yakutia) we have produced a series of <sup>14</sup>C dates on different kinds of organic matter—plant remains, peat, roots and wood, and bones. In many cases, the <sup>14</sup>C ages of plant remains were younger than the ages of bones from the same layers. For example, the ages of plant remains from Zelyonyj Mys sequences were in the interval from 27 to 37 ka BP and those of bones from the same depth from 43.7 to >50 ka BP (Vasil'chuk 1992). The same situation occurred in the Duvannyj Yar natural exposure, where the series of <sup>14</sup>C dates of plant remains lies in the interval from 40 to 20 ka BP, with three dates of mammoth bones in a normal sequence (28.6, 33.8, 34.7) and two inversion dates of >50 and 53 ka BP (Vasil'chuk 1992). We received non-inversion dates on tusk (15.1 ka BP) in the Ledovyj Obryv natural exposure in Chukotka, which were between the dates on plant remains of 34 and 14 ka BP (Vasil'chuk 1992). These data show that the dating of the host sediments by the use of mammoth bones is, in principle, possible; however, redeposition of separate bones is typical and must be taken into account in determining the host sediments' age.

Because whole carcasses are, as a rule, not redeposited, their <sup>14</sup>C dates conform better with the age of the host formation. However, there are some exceptions. For example, the Kirgilyakh baby mammoth (which is <sup>14</sup>C-dated to 38–41 ka BP) had been redeposited together with frozen host sediments into the younger (<sup>14</sup>C-dated as Late Holocene) permafrost complex.

# Mammoth Fossils and Reconstruction of Environmental Conditions

Some important regularities in the distribution of mammoth fossils appear within the Eurasian territory. First, the temporal distribution of the mammoth remains found in Eurasia is rather even for the whole period from 10 ka BP to the older limit of the <sup>14</sup>C method (Fig. 2). Second, the spatial distribution of dated fossils shows that the southern boundary of the mammoths' distribution is very close to the southern boundary of the ice wedge cast distribution, which is located *ca.* 45°N (Fig. 1). It may be assumed that the Late Pleistocene mammoth habitat corresponds to the severe permafrost area characterized by vast polygonal ice wedge landscapes. Third, as no breaks occur in the series of data from northern Asia, the European part of Russia and western Europe including Great Britain, it seems that the mammoths lived everywhere over this vast area. Therefore, the series of <sup>14</sup>C dates from 40 to 10 ka BP on mammoth fossils from Scandinavia gives reason for critical evaluation of the scale and dynamics of the Late Pleistocene Glaciation in this region. Mammoth remains in South Sweden (from 13 to >30 ka BP), Norway (from 19 to 32 ka BP), Finland (from 15 to >43 ka BP) and Denmark (from 13 to 32 ka BP) suggest that large parts of Scandinavia were ice-free in Middle and Late Weichselian time (Donner, Jungner and Kurten (1979) reached similar conclusions).

So, for the period of the last glaciation,  ${}^{14}C$  dates on mammoth remains have been obtained from the entire territory of the supposed last glaciation area.  ${}^{14}C$  dates for the period of the last glaciation have been obtained in North America as well. Weber *et al.* (1981) received six  ${}^{14}C$  dates from bone fragments from Canyon Creek in interior Alaska—*ca.* 28, 32, 38, 39, 39 and 40 ka BP. In Canyon Creek, a portion of a tooth plate and bone fragments of *Mammuthus primigenius* and many bones of *Equus*, *Alces, Lepus, Canis, Ovis, Bison, etc.*, were sampled (Weber *et al.* 1981). Assuming that the  ${}^{14}C$  dates are trustworthy, the finds of mammoth remains show that our knowledge about the glaciation environment needs essential supplements.

# CONCLUSION

Comprehensive analyses of <sup>14</sup>C, pollen, oxygen isotope and geological data enable us to draw some conclusions about the development of the mammoth fauna and their environment. In particular our data show that:

- 1. Mammoth remains from frozen ground are a very suitable material for <sup>14</sup>C dating.
- 2. The temporal distribution of the <sup>14</sup>C dates of fossils from the vast Eurasian territory is even for the whole period from 40 to 10 ka BP.
- 3. No time breaks appear in the series of dates in northern Asia, nor in eastern and western Europe.
- 4. The southern boundary of mammoths' distribution is close to the southern boundary of the ice wedge cast distribution (and therefore close to the southern limit of severe permafrost). This demonstrates that the mammoth fauna is a typical component of Late Quaternary permafrost environments.

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#### **APPENDIX: RADIOCARBON DATES OF MAMMOTH REMAINS IN EURASIA**

No. <sup>14</sup>C date (yr BP), Lab code

#### Material dated Site

Eastern	n Europe		
1	9780 ± 260 (TA-12)	Bone	Kunda Settlement
2	11,000 ± 200 (GIN-93)	Bone	Kostenki Settlement
3	12,200 ± 300 (IGAN-282)	Molar	TimonovkaI Settlement
4	12,630 ± 360 (GIN-4137)	Molar	Eliseevichi Settlement
5	12,900 ± 200 (OxA-709)	Molar	Mezhirichi Settlement
6	12,970 ± 140 (LU-102)	Molar	Eliseevichi Settlement
7	13,650 ± 180 (LU-153)	Molar	Yudinovo Settlement
8	13,680 ± 60 (GIN-6209)	Tusk	Sevsk Town
9	13,900 ± 200 (IGAN-78)	Bone	Avdeevo Settlement
10	13,950 ± 70 (GIN-5778)	Bone	Sevsk Town
11	14,100 ± 400 (GIN-4139)	Molar	Eliseevichi Settlement
12	14,290 ± 120 (GIN-2356)	Tusk	Or'ya Settlement
13	14,320 ± 270 (QC-897)	Molar	Mezhirichi Settlement
14	14,360 ± 150 (GIN-2913)	Bone	Shatrishche Settlment
15	$14,400 \pm 250$ (OxA-712)	Molar	Mezhirichi Settlement
16	14,470 ± 100 (LU-126)	Molar	Eliseevichi Settlement
17	14,590 ± 140 (GIN-4136)	Molar	Eliseevichi Settlement
18	14,600 ± 200 (OxA-717)	Molar	Gontsy Settlement
19	14,700 ± 250 (OxA-715)	Molar	Chulatovo Settlement
20	14,700 ± 500 (GIN-2593)	Molar	Kintu Lake
21	15,100 ± 200 (OxA-719)	Molar	Mezin Settlement
22	15,100 ± 250 (OxA-716)	Molar	Berdyzh Settlement
23	15,110 ± 530 (LU-358)	Bone	Timonovka I Settlement
24	15,660 ± 180 (LU-127)	Molar	Yudinovo Settlement
25	15,245 ± 1080 (QC-900)	Molar	Mezhirichi Settlement
26	16,300 ± 700 (GIN-2002)	Molar	Timonovka I Settlement
27	16,850 ± 120 (GIN-4138)	Molar	Eliseevichi Settlement
28	17,340 ± 170 (LU-360)	Molar	Eliseevichi Settlement
29	17,930 ± 100 (LE-1432A)	Molar	Gagarino Settlement
30	18,300 ± 200 (GIN-3727)	Molar	Zaraisk Town
31	18,320 ± 280 (TA-121)	Bone	Byzovaya Settlement

32	18,690 ± 770 (LU-361)	Molar
33	19,000 ± 300 (OxA-697)	Molar
34	19,200 ± 350 (OxA-718)	Molar
35	19,200 ± 200 (LE-2946B)	Molar
36	19,280 ± 600 (KI-1058)	Bone
37	19,800 ± 350 (OxA-698)	Molar
38	20,150 ± 100 (LE-1432B)	Molar
39	20,300 ± 200 (LE-1602)	Molar
40	$20,620 \pm 100 (LE-1432C)$	Molar
41	$21,240 \pm 200 (LE-1602a)$	Molar
42	$21,600 \pm 2200$ (GIN-4)	Molar
43	$22,000 \pm 300$ (GIN-3698)	Molar
44	$22,200 \pm 300$ (GIN-3634)	Bone
45	$22,600 \pm 300 (GIN-3633)$	Bone
40	$22,800 \pm 300 (GIN-3632)$	Bone
4/	$23,430 \pm 180 (LU-104)$	Molar
48	$23,000 \pm 270 (LU-359)$	Molar
49	$23,770 \pm 1540$ (LE-2946A)	Molar
50	$24,000 \pm 150 (LE-2624)$	Molar
51	$24,950 \pm 400 (IGAN-73)$	Molar
52	$25,000 \pm 300 (GIN-2463)$	Bone
53	$25,300 \pm 400 (GIN-6143)$ $26,470 \pm 420 (UII 125)$	Molar
54	$20,470 \pm 420 (LU-125)$	Molar
56	$27,300 \pm 800$ (KI-1051)	Molar
57	$27,700 \pm 500 (GIN-5880)$	Bone
58	$30,500 \pm 900 (L0-60)$ $32,100 \pm 500 (CIN 6146)$	Bone
50	$35,100 \pm 300$ (GIN 6622)	Molar
60	$37,000 \pm 500$ (GIN 6141)	Soft fissue
61	$37,000 \pm 300$ (GIN-0141) $37,300 \pm 1000$ (GIN 6142)	Molar
62	$37,500 \pm 1000$ (GIN-0142) $37,600 \pm 400$ (GIN 3221)	Molar
63	$38400 \pm 1000$ (GIN 6148)	Molar
64	$42200 \pm 300$ (GIN-6410)	Tuels
65	$43600 \pm 1000(GIN-6145)$	I USK Moler
66	$44000 \pm 1000(GIN-6144)$	Molar
67	$44.200 \pm 1000$ (GIN-6147)	Molar
68	>47.700 (GIN-7075)	Bone
North	of Furning Bussia	Done
60	18320 + 280(TA121)	Dere
70	$20.300 \pm 200 (TA-121)$	Done
71	$>36\ 000\ (IEMAE)$	Molar
Wanta	Situate	TUSK
w este	n Siberia	_
72	$12,860 \pm 90$ (SUAN-1283)	Bone
73	$13,350 \pm 60$ (GIN-7539)	Bone
74	$13,930 \pm 80 (GIN-7541)$	Bone
76	$14,240 \pm 100 (SUAN-78)$	Bone
70	$10,000 \pm 2000 (GIN - 2862)$	Bone
79	$19,300 \pm 200 (GIN - 2859)$ 10,700 + 200 (GIN - 2851)	Bone
70	$19,700 \pm 200 (GIN - 2001)$ 19,960 + 80 (GIN 2016)	Tusk
80	$20100 \pm 100(\text{GIN}, 2962)$	Bone
81	$20,100 \pm 100$ (GIN 2017)	Tusk
82	$20,100 \pm 300 (GIN-3017)$	Bone
83	$20,400 \pm 240 (SUAN-1313)$	Tusk
84	$41900 \pm 900(\text{GIN}5337)$	Bone
N7	41,500 ± 800 (GIN-5557)	Molar
North	of western Siberia	
85	$9000 \pm 300 (VSEGINGEO)$	Soft tissue
80 07	$9/30 \pm 100 (MGU-763)$	Stomach cor
0/ 00	$10,000 \pm 70 (LU-1153)$	Stomach cor
66 20	$10,330 \pm 30 (GIN-6386)$	Molar
00	$17,700 \pm 300$ (GIN-/292)	Bone
01	$1,300 \pm 300 (GIN - /3 / 0)$ 25 400 + 300 (GIN 2210)	Molar
<b>71</b>	20,700 ± 300 (0111-2210)	Done

Pogon Settlement Radomyshl' Settlement Kirillovka Settlement Leski Settlement Mezhirichi Settlement Novgorod-Severski Town Gagarino Settlement Sagaidak I Settlement Gagarino Settlement Sagaidak I Settlement Mezin Settlement Zaraisk Town Kostenki Settlement Kostenki Settlement Kostenki Settlement Berdyzh Settlement Khotylevo Settlement Leski Settlement Anetovka II Settlement Khotylevo Settlement Nizhniy Novgorod Region Lower Kama River Yurovichi Settlement Mezin Settlement Sungir', Vladimir Region Tver' Region Lower Kama River Starun Town Lower Kama River Lower Kama River Viliya River (Neman) Lower Kama River Novopetrovskoe Settlement, Moscow Region Lower Kama River Lower Kama River Lower Kama River Pavlovsk, Voronezh Region Pechora River Bol'shezemel'skaya tundra Kanin Peninsula Irtysh River, W.S. Krasnoyarsk Town Krasnoyarsk Town Volch'ya Griva Settlement Middle Enisey River Middle Enisey River Middle Enisey River Chulym River Middle Enisey River Middle Enisey River Mogochino Settlement Chulym River Tavda River Yuribey River ntents Yuribey River Yuribey River ntents Seyakha Mutnaya River Seyakha Zelyonaya River Parisento River

Upper Yuribey River

92	27.200 ± 500 (GIN-2021b)	Molar	Yambuto Lake, Gydan
93	$29.300 \pm 300$ (GIN-6386A)	Bone	Seyakha Mutnaya River,
94	$30.250 \pm 1800 (T-298)$	Skin	Gyda River
95	$34.500 \pm 300$ (GIN-6475A)	Tusk	Shchushch'ya River, Yamal
96	$31.500 \pm 600$ (GIN-2201)	Bone	Yekaryauyakha River, Gydan
97	$33500 \pm 1100$ (T-298)	Blubber	Gyda River
98	$34500 \pm 300$ (GIN-6475A)	Tusk	Shchushch'ya River, Yamal
00	$35,500 \pm 1100$ (T-298)	Blubber	Gyda River
	- Deminant	214000	-,
1 aimy	r Peninsula	Tuck	Nizhnava Taimvra River
100	$9070 \pm 60 (GIN - 1828)$	Molar	Nizhnava Taimyra River
101	$9800 \pm 50 (GIN-1495)$	Molar	Engel'gord Lake
102	$10,100 \pm 100$ (GIN-1489)	Pone	Nizhnovo Toimyra River
103	$10,300 \pm 100 (GIN-1828K)$	Done.	Ngoposonskava Piver
104	$10,080 \pm 70 (GIN-3708)$	Malar	Toimur Lake
105	$11,140 \pm 180 (GIN-3067)$	Molar	Mamont Diver Shrenk
106	$11,450 \pm 250 (1-297)$	Soft fissue	Taiman Lake Boshure
107	$12,100 \pm 80$ (GIN-1783)	Bone	Savarrava Biyar
108	$12,260 \pm 120$ (GIN-2943r)	Bone	Severnaya River
109	$12,450 \pm 60 (GIN-3242)$	Bone	Bile de Biuer
110	$12,780 \pm 80$ (GIN-2677)	Bone	Bikada River
111	$13,340 \pm 240$ (GIN-2758a)	Bone	Bolsnaya Balachnya River
112	$16,330 \pm 100 (GIN-3130)$	Mandible	Bolsnaya Balachnya River
113	$18,680 \pm 120$ (GIN-5046)	Tusk	Boderbo-Tarida River
114	20,400 ± 100 (GIN-3952)	Tusk	Dudypta River
115	22,750 ± 150 (GIN-3089)	Bone	Taimyr Lake, Baskura
116	23,500 ± 300 (GIN-2763a)	Tusk	Bolshaya Balachnya River
117	23,800 ± 400 (GIN-1296B)	Bone	Taimyr Lake, Sabler
118	$24,900 \pm 500$ (GIN-2160)	Pelvis	Taimyr Lake, Baskura
119	$25,100 \pm 550$ (LE-612)	Soft tissue	Pyasina River
120	$26,700 \pm 700$ (GIN-1216)	Tusk	Gulya River
121	$27,300 \pm 200$ (GIN-3836)	Tusk	Logata River
122	$27,500 \pm 300$ (GIN-3929)	Tusk	Kubalakh River
123	$27,500 \pm 200$ (GIN-3505)	Scalpula	Lygiy- I urege River
124	$28,800 \pm 600 (GIN-952)$	Tusk	Chaidachtar Lake
125	$29,400 \pm 400$ (GIN-3310)	Tusk	Chaldachtar Lake
126	$29,500 \pm 300 (GIN-2155)$	TUSK	Taimyr Lake, Matuda
127	$31,800 \pm 500 (GIN - 3240a)$	Bone	Severinaya River
128	$31,900 \pm 300$ (GIN-5/26)	Bone	Balakhaya Biyar
129	$32,000 \pm 200 (GIN-3117)$	Femur	Dalakiniya Kivel
130	$32,000 \pm 500$ (GIN-2151)	Limb bone	Lagata Diver
131	$35,000 \pm 500 (GIN - 3821)$	Bone	Logata River
132	$35,800 \pm 2700$ (1-169)	Skin	Moknovaya River
133	$36,200 \pm 500$ (GIN-3822)	TUSK	Logata River
134	$36,800 \pm 500 (GIN-3122)$	TUSK	Bolsnaya Balachiya Kiver
135	$36,950 \pm 4300 (1-169)$	Skin	Moknovaya River
136	$38,000 \pm 1500 (GIN-942)$	Tusk	Knatanga River
137	$38,300 \pm 600 (GIN-3817)$	TUSK	Logata River
138	$38,400 \pm 700$ (GIN-3118)	Tusk	Bolsnaya Balachnya River
139	$38,500 \pm 500$ (GIN-3136)	Bone	Boderbo-Tarida River
140	$38,500 \pm 500 (GIN-2763B)$	Molar	Boderbo-Tarida River
141	$38,800 \pm 400 (GIN - 3476)$	Tusk	Nemudikatarida River
142	38,800 ± 1300 (GIN-1491)	Tusk	Trautfetter River
143	38,900 ± 600 (GIN-3831)	Tusk	Logata River
144	$39,100 \pm 700 (GIN-3120P)$	Bone	Bolshaya Balachnya River
145	39,300 ± 500 (GIN-3121P)	Bone	Bolshaya Balachnya River
146	39,300 ± 500 (GIN-3071)	Bone	Taimyr Lake, Baykura
147	39,800 ± 600 (GIN-3135)	Bone	Boderbo-Tarida River
148	40,200 ± 600 (GIN-3804)	Tusk	Logata River
149	40,500 ± 800 (GIN-1818P)	Bone	Engel'gard Lake
150	40,800 ± 2000 (GIN-1835)	Bone	Taimyr Lake, Gotman
151	41,200 ± 1000 (GIN-2744B)	Bone	Boderbo-Tarida River
152	41,400 ± 2000 (GIN-3941)	Tusk	Shaitan Lake
153	42,800 ± 800 (GIN-3946)	I USK	massonov, Knatanga Kiverbasin

154	43.500 ± 1000 (GIN-3072)
155	$45.000 \pm 1000$ (GIN-766)
156	$46.100 \pm 1200$ (GIN-3073)
157	$47.900 \pm 1600$ (GIN-3118a)
158	>49,500 (GIN-3080)
159	>49,500 (GIN-3092a)
160	>52,700 (GIN-2764B)
161	>53,170 (LU-1057)
Centr	al Siberia
162	$20,700 \pm 150$ (GIN-7709)
163	$21,600 \pm 200$ (GIN-7708)
164	23,600 ± 200 (GIN-5886)
165	41,100 ± 1500 (GIN-7707)
166	49,700 ± 1100 (GIN-689)
North	ern Yakutia
167	$10.370 \pm 70$ (SOAN-327)
168	$12,000 \pm 130 (LU-149)$
169	$12,240 \pm 160 (LU-139)$
170	$12,530 \pm 60$ (SOAN-2203)
171	12,570 ± 80 (MAG-826)
172	12,850 ± 110 (LU-1055)
173	13,700 ± 800 (MAG-114)
174	14,340 ± 50 (GIN-4115)
175	14,800 ± 50 (GIN-3518)
176	17,780 ± 80 (GIN-5042)
177	18,680 ± 120 (GIN-5046)
178	$18,700 \pm 100$ (GIN-6099)
179	21,260 ± 310 (LU-786)
180	$21,630 \pm 240$ (LU-1328)
181	$22,000 \pm 200$ (GIN-5574)
182	$23,100 \pm 200$ (GIN-3232)
183	$24,000 \pm 1100$ (GIN-7176)
184	$25,300 \pm 600$ (GIN-3502) 26,000 $\pm 1600$ (M-215)
185	$20,000 \pm 1000 (M0-215)$
187	$28,000 \pm 300 (GIN - 5807)$
188	$29,900 \pm 300$ (GIN 3310)
189	$29,500 \pm 3000$ (CIRC-3310)
190	$29,600 \pm 5000$ (GIN-3234)
191	$30400 \pm 300$ (GIN-6023a)
192	$31.500 \pm 2000 (\text{T-170})$
193	$31.750 \pm 2500 (T-299)$
194	31,900 ± 300 (GIN-5726)
195	32,200 (500 (SOAN-1006B)
196	32,300 ± 400 (GIN-5074)
197	32,650 ± 2500 (T-170)
198	33,800 ± 500 (GIN-3861)
199	34,450 ± 2500 (T-171)
200	34,700 ± 400 (GIN-4434)
201	35,000 ± 300 (GIN-3503)
202	35,800 ± 1200 (T-171)
203	35,830 ± 630 (LU-504)
204	$36,450 \pm 420$ (SOAN-1005)
203	$30,000 \pm 500 (GIN-5751)$
200	$37,000 \pm 500 (GIN-5750)$
201	$33,400 \pm 1000 (GIN-3517)$
200	$40,100 \pm 300 (GIN-5/26A)$
209	$70,300 \pm 400 (011N-3023)$ $40.350 \pm 990 (1.11 + 605)$
210	41 750 + 1200 (LU-375)
212	$41,000 \pm 1200 (EU-505)$ 41,900 + 800 (GIN-5224)
213	$42.400 \pm 800$ (GIN-6310)
214	$43.200 \pm 400$ (GIN-6100)

Bone Taimyr Lake, Baykura Bone Kheta River Bone Taimyr Lake, Baykura Tusk Bolshaya Balachnya River Tusk Taimyr Lake, Baykura Tusk Bolshaya Balachnya River Bone Boderbo-Tarida River Bone Bone Bone Bone Bone Tusk Bone Tusk Tusk Humerus Humerus Tusk Soft tissue Tusk Tusk Molar Tusk Tusk Mandible Carpal Molar Pelvis Tusk Bone Hair Limb Bone Tusk Tusk Soft tissue Vertebrae Bone Soft tissue Soft tissue Ribs Stomach contents Tusk Soft tissue Bone Soft tissue Bone Bone Soft tissue Skin Soft tissue Molar Molar Bone Molar Molar Stomach contents Soft tissue Tusk Molar Bone

Khatanga River Belaya River (Angara) Belaya River (Angara) MiddleAngara River Belaya River (Angara) Maimechya River, Putoran Berelyokh River Berelyokh River Berelyokh River Achchaika-Allaikha River Achchaika-Allaikha River Berelyokh River Berelyokh River Lena River Ulakhan-Yuryakh Lower Lena River Bur, Olenyok River Basin Amydai, Olenyok Basin Lower Lena River Bykovski Peninsula Anabar River, Popigay Tyung, Lena River Kular Settlment Yarasalakh River Lena River, Chekurovka Duvanniy Yar, Kolyma River Anabar River, Popigay Anabar River Lena River, Sanga-Yuryakh Tyung River, Lena basin Khamus-Yuryakh River, Kolyma Lena River, Sanga-Yuryakh Beryosovka River Anabar Gulf, Sualema River Shandrin River **Popigay River** Lena River, Sanga-Yuryakh Duvanny Yar, Kolyma River Lena River (Bykovskaya) Duvanny Yar, Kolyma River Laptev Sea coast Lena River (Bykovskaya) Tirekhtyakh River Shandrin River Anabar River, Popigay Semiriskai River, Popigay Laptev Sea coast Anabar Gulf Anabarka River, Popigay Shandrin River Shandrin River Anabarka River, Popigay Khamus-Yuryakh Riv, Kolyma Amydai, Olenyok Basin

215	43,700 ± 800 (GIN-3849)	Bone	Zelyoniy Mys Settlement
216	44,000 ± 3500 (T-170)	Soft tissue	Lena River, Sanga-Yuryakh
217	44,000 ± 3500 (T-299)	Soft tissue	Beryosovka River
218	44,540 ± 1900 (LU-1050)	Bone	Tirekhtyakh River
219	45,500 ± 1200 (GIN-6105)	Tusk	Amydai, Olenyok Basin
220	46,100 ± 1000 (GIN-3206)	Bone	Lower Kolyma River
221	49,500 (GIN-6101)	Tusk	Nekyu, Olenyok Basin
222	>50,000 (GIN-359)	Bone	Lower Lena River
223	>50,000 (GIN-5731)	Molar	Anabarka River, Popigay
224	>50,000 (SOAN-813)	Soft tissue	Tirekhtyakh River
225	>50,000 (GIN-3848)	Bone	Zelyoniy Mys Settlement
226	>50,000 (GIN-3866)	Bone	Duvanniy Yar, Kolyma River
227	50.400 ± 1300 (GIN-4114)	Tusk	Lower Lena River
228	>53.000 (GIN-3857)	Tusk	Duvanniv Yar, Kolvma River
229	>53.170 (LU-1057)	Skin	Tirekhtyakh River
Maga	dan Region		
220	$21.600 \pm 200$ (GIN 6200)	Tuck	Tanon River
230	$21,000 \pm 200 (GIN - 0509)$	Tusk	Srednekan Diver
231	$20,400 \pm 300 (0114-3090)$	Soft ticous	Kirgilyakh Diver
232	$39,570 \pm 870$ (LU-718A)	Soft tissue	Kirgilyakh River
233	$39,390 \pm 7/0 (L0-718B)$	Soft tissue	Kingilyakii Kiver
234	$40,000 \pm 700 (MAG-300A)$	Soft tissue	Kingilyakii Kiver
235	$41,000 \pm 1100 (MAG-300B)$	Soft tissue	Kingilyakii Kiver
236	$41,000 \pm 900 (MAG-576)$	Son tissue	Kirgilyakh Kiver
Chuko	otka Peninsula		
237	14,380 ± 70 (GIN-7289)	Tusk	
238	15,100 ± 70 (GIN-5370)	Tusk	Mayn River
239	31,370 ± 900 (MAG-1000A)	Soft tissue	Enmynveem River
240	31,100 ± 900 (MAG-1000B)	Soft tissue	Enmynveem River
241	32,800 ± 720 (MAG-1001A)	Soft tissue	Enmynveem River
242	32,850 ± 900 (MAG-1000)	Soft tissue	Enmynveem River
243	32,890 ± 1200 (MAG-1001B)	Soft tissue	Enmynveen River
244	32,000 ± 3000 (MAG-1124)	Soft tissue	Enmynveem River
Kamci	hatka Peninsula		
245	$12.630 \pm 50$ (GIN-3420)	Tusk	Kamchatka River, Urz
246	$21,300 \pm 400$ (GIN-2224)	Skull	Pakhtcha River
247	$21,750 \pm 150$ (GIN-5299b)	Tusk	Kamchatka River
248	$30000 \pm 300(\text{GIN}-3415)$	Tusk	Kamchatka River
249	$36000 \pm 500(\text{GIN}-3425)$	Tusk	Kamchatka River
250	$39.600 \pm 1600$ (GIN-3411)	Molar	Kamchatka River
251	$40600 \pm 600(\text{GIN}-3407)$	Tusk	Kamchatka River
4		1 USK	
Arcuc 252	$2720 \pm 40$ (LU 2741)	Tuck	Wrongel Island
252	$3/30 \pm 40 (LU-2/41)$	Tusk Tusl	Wrangel Island
255	$3920 \pm 30 (GIN-0980)$	I USK	Wrangel Island
254	$4010 \pm 50 (LU - 2798)$	Molar	Wrangel Island
200	$4040 \pm 30 (LU - 2808)$	Tootn	Wrangel Island
250	$4400 \pm 40 (LU-2756)$	Tusk	Wrangel Island
257	$4410 \pm 50 (LU-2768)$	T USK	Wrangel Island
258	$4/40 \pm 40 (LU-2556)$	I ibia	wrangel Island
259	$4900 \pm 40 (LU - 2740)$	TUSK	wrangel Island
260	$5110 \pm 40 (LU - 2794)$	Molar	wrangel Island
261	$5200 \pm 30 (LU-2745)$	TUSK	wrangel Island
262	$5250 \pm 40 (LU - 2744)$	TUSK	wrangel Island
263	$5310 \pm 90 (LU-2/42)$	I USK	wrangei Island
264	$5480 \pm 50 (LU-2535)$	TUSK	wrangei Island
265	6260 ± 50 (LU-2799)	Molar	Wrangel Island
266	6360 ± 60 (AA-11529)	Molar	Wrangel Island
267	<pre>ccia : #0 /3 ** ******</pre>		
~ ~ ~	6610 ± 50 (LU-2558)	Tusk	Wrangel Island
268	6610 ± 50 (LU-2558) 6760 ± 50 (LU-2736)	Tusk Tusk	Wrangel Island Wrangel Island
268 269	6610 ± 50 (LU-2558) 6760 ± 50 (LU-2736) 6890 ± 50 (LU-2810)	Tusk Tusk Tooth	Wrangel Island Wrangel Island Wrangel Island
268 269 270	6610 ± 50 (LU-2558) 6760 ± 50 (LU-2736) 6890 ± 50 (LU-2810) 7040 ± 60 (LU-2746)	Tusk Tusk Tooth Tusk	Wrangel Island Wrangel Island Wrangel Island Wrangel Island
268 269 270 271	$6610 \pm 50 (LU-2558)  6760 \pm 50 (LU-2736)  6890 \pm 50 (LU-2810)  7040 \pm 60 (LU-2746)  7250 \pm 60 (LU-2809)  7250 \pm 60 (LU-280)  7250 \pm$	Tusk Tusk Tooth Tusk Molar	Wrangel Island Wrangel Island Wrangel Island Wrangel Island Wrangel Island
268 269 270 271 272	$6610 \pm 50 (LU-2558)$ $6760 \pm 50 (LU-2736)$ $6890 \pm 50 (LU-2810)$ $7040 \pm 60 (LU-2746)$ $7250 \pm 60 (LU-2809)$ $7295 (95 (AA-11530)$	Tusk Tusk Tooth Tusk Molar Molar	Wrangel Island Wrangel Island Wrangel Island Wrangel Island Wrangel Island Wrangel Island

274	7390 ± 30 (LU-2444)	Tusk
275	7710 ± 40 (GIN-6995)	Tusk
276	11,500 ± 60 (LU-610)	Tusk
277	12,010 ± 110 (LU-2823)	Molar
278	12,750 ± 50 (GIN-6987)	Tusk
279	12,980 ± 80 (LU-2792)	Molar
280	$15.420 \pm 100$ (LU-1671)	Tusk
281	$19.270 \pm 300 (LU-654B)$	Tusk
282	$19.970 \pm 110$ (LU-688)	Molar
283	$19.990 \pm 110 (LU-1790)$	Tusk
284	$20000 \pm 110$ (LU-2807)	Molar
285	$20.900 \pm 100$ (GIN-5760)	Tuck
286	$25,500 \pm 100$ (CH( $\pm 5700$ )	Tibia
287	$25,000 \pm 210 (B0 + 74)B)$ 25,800 ± 200 (GIN-4710B)	Tuck
288	$28,000 \pm 200$ (GIN 4710)	Tusk
280	$20,000 \pm 200 (011-4710)$	Tusk
209	$29,020 \pm 190 (L0-1/91)$	TUSK
290	$29,100 \pm 400$ (GIN-4330)	Bone
291	$29,100 \pm 1000 (GIN-4/11)$	Tusk
292	$32,100 \pm 900 (MAG-316)$	Skin
Denm	ark	
293	13,240 +760/-690 (K-3697)	Tusk
294	21,530 ± 430 (K-3703)	Bone
295	24,190 ± 420 (K-3806)	Tusk
296	25,110 ± 440 (K-3699)	Tusk
297	25,480 +560/-520 (K-3809)	Tusk
298	25,520 +920/-830 (K-3805)	Tusk
299	25,760 +840/-770 (K-3805)	Tusk
300	26.270 +1400/-1210 (K-3805)	Tusk
301	$27.810 \pm 610 (K-4192)$	Tusk
302	28.120 + 760/-680 (K-3808)	Tusk
303	29,570 + 950 (K-3807)	Tusk
304	$31.840 \pm 1010/-870 (K_{-3696})$	Tusk
305	$32460 \pm 970/-870(K-4190)$	Tusk
306	>37,000 (V, 1101)	Tusk
307	>30,500 (K-4191)	Tusk
200	> 39,500 (K-4188)	Tusk
508	>39,000 (K-4387)	Tusk
Germa	iny	
309	15,810 ± 410 (HV-1961)	Molar
310	30,300 +2500/-1900 (Fra-5a)	Femur
Switze	rland	
311	12,270 ± 210 (Ly-877)	Tusk
312	34.600 + 2700/-1800 (Lv-751)	Tusk
Poland	· ,···· · ··· · ··· · ··· (; ····)	
212	• 20 200 + 350 (0 4 625)	Dene
313	$20,200 \pm 330 (0XA-033)$	Bone
215	$20,000 \pm 1000 (Ly-0.001)$	Bone
210	$21,000 \pm 900 (Ly-2542)$	Bone
310	$23,040 \pm 170$ (GrN-6636)	Bone
France	8	
317	12,000 ± 220 (Ly-1351)	Scapula
318	14,330 ± 260 (Ly-357)	Bone
319	$14,390 \pm 300$ (Ly-433)	Bone
320	$14,850 \pm 350$ (Ly-434)	Bone
321	$25.800 \pm 700$ (Ly-1863)	Tusk
Iroland	, (,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	
322	33,500 ± 1200 (D-122)	Molar
Great 1	Britain	
323	11,650 ± 130 (OxA-2155)	Ivory rod
324	12,080 ± 130 (OxA-1457)	Skull
325	$12,170 \pm 130$ (OxA-1890)	Ivory rod
326	12,300 ± 180 (OxA-1316)	Molar
327	$12,320 \pm 120$ (OxA-1462)	Tusk
328	$12.330 \pm 120$ (OxA-1456)	Molar
-	,	

Wrangel Island Wrangel Island Severnaya Zemlya Wrangel Island Wrangel Island Wrangel Island Kotel'niy Island Severnaya Zemlya Severnaya Zemlya Kotel'niy Island Wrangel Island Faddeevskiy Island Severnaya Zemlya Faddeevskiy Island Faddeevskiy Island Kotel'niy Island Faddeevskiy Island Faddeevskiy Island Lyakhovskiy Bolshoiy Island Rosmos 1 Myrup Banke Munke Bjergby Hadsund Ostrupgard Ny Stengaard Ny Stengaard Ny Stengaard Stengardens Grusgrav 2 Ronninge 1 Saxkobing Kiskelund Lundebjerg 1 Sonder Kollemorten Sonder Omme Rosmos 2 Kelsterbach Kelsterbach Praz Rodet Bioley-Orjulaz Spadzista Street, Krakow Spadzista Street, Krakow Spadzista Street, Krakow Spadzista Street, Krakow Etiolles La Croze-sur-Suran 1 La Columbière rockshelter La Croze-sur-Suran 2 La Mère Clochette Grotto Castlepook Cave, Cork Kent's Cavern Condover, Shropshire Cough's Cave Condover, Shropshire Robin Hood's cave

Condover, Shropshire

329	12,400 ± 160 (OxA-1455)	Tusk	Condover, Shropshire
330	12,460 ± 160 (OxA-1204)	Calcaneum	Condover, Shropshire
331	12,480 ± 96 (Birm-1273com.)	Tusk	Condover, Shropshire
332	12,700 ± 160 (OxA-1021)	Tusk	Condover, Shropshire
333	12,920 ± 390 (Birm-1273)	Tusk	Condover, Shropshire
334	18,000 +1400/-1200 (Birm-146)	Bone	Cae Gwyn Cave, Wales
335	19,300 ± 700 (Gif-1110)	Bone	Condover, Shropshire
336	26,700 ± 550 (OxA-1205)	Bone	Pin Hole Cave, Creswell
337	33,200 ± 1300 (OxA-1069)	Bone	Conningbrook, Kent
338	34,500 ± 500 (Birm-466)	Bone	Little Rissington
339	34,850 ± 1500 (OxA-1654)	Molar	King Arthur's Cave
340	35,200 ± 1600 (OxA-1610)	Bone	Conningbrook, Kent
341	37,020 +1900/-1350 (Q-2500)	Tusk	Farnham, Surrey
342	37,300 ± 1900 (OxA-1644)	Bone	Conningbrook, Kent
343	38,500 ± 2300 (OxA-1565)	Molar	King Arthur's Cave
344	38,600 ± 2400 (OxA-1611)	Bone	Conningbrook, Kent
345	38,600 +1720/-1420 (NPL-1628)	Tusk	Oxbow, Leeds
346	>39,500 (OxA-1566)	Molar	King Arthur's Cave
Finlan	ad		
347	15,500 ± 200 (Hel-1074)	Humerus	Herttoniemi
348	25,200 ± 500 (Hel-1075)	Femur	Lohtaja
349	>43,000 (Hel-1076)	Molar	Espoo
Norwa	IV		
350	19,000 ± 1200 (U-4214)	Tusk	Toten, Opland
351	$20000 \pm 250$ (K-3703)	Tusk	Favang
352	22.370 ± 980 (K-3703)	Tusk	Favang
353	24,400 ± 900 (K-3806)	Tusk	Kvam
354	28,100 +2300/-1800 (U-2766)	Tusk	Toten, Opland
355	32,100 +3100/-2300 (U-4214)	Tusk	Toten, Opland
Swede	'n		
356	$13.090 \pm 120$ (Lu-796, 2)	Tusk	Lockarp
357	$13,260 \pm 110$ (Lu-865)	Tusk	Lockarp
358	13,360 ± 95 (Lu-796)	Tusk	Lockarp
359	19,150 ± 390 (Lu-887E)	Tusk	Arrie
360	22,000 +900/-800 (Lu-887)	Tusk	Arrie
361	31,200 +3050/-2650 (Lu-746)	Tusk	Orsjo
362	36,000 +1550/-1300 (Lu-879)	Tusk	Dosebacka
363	36,100 +2000/-1600 (Lu-880)	Tusk	Orsjo

References: Berglund, Hakansson and Lagerlund 1976; Follestad and Olsson 1979; Orlova 1979; Jungner and Sonninen, 1983; Aaris-Sorensen et al. 1990; Stuart 1991; Vasil'chuk 1992; Svezhentsev and Popov 1993; Vartanyan, Garutt and Sher 1993; Vartanyan et al. 1995; Sulerzhitskij 1995.

# RADIOCARBON AMS DATING OF POLLEN CONCENTRATED FROM EOLIAN SEDIMENTS: IMPLICATIONS FOR MONSOON CLIMATE CHANGE SINCE THE LATE QUATERNARY

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ABSTRACT. Dating pollen concentrated from eolian sediments provides a new way to establish a chronological framework on the Loess Plateau of China. We show that pollen deposited simultaneously with sediment in a stable environment can provide reliable ages. We suggest that the reliability of pollen dating can be evaluated by comparison with wood cellulose or charcoal ages from the same stratigraphic level. Dating pollen concentrates from the various profiles indicates paleomonsoon precipitation variability at the loess/desert transitional belt from the late Pleistocene to the early Holocene.

# INTRODUCTION

Loess-paleosol <sup>14</sup>C dating in the loess Plateau of China has been limited by the lack of suitable dating material. Previously, datable materials included mainly organic matter in soil, which generally provides a minimum age, owing to the presence of younger organic materials brought in by agriculture or atmospheric carbon incorporated through plant growth (Head, Zhou and Zhou 1989).

In this paper, we evaluate the use of fossil pollen as a dating material. Pollen concentrated from sediments can be dated on the basis of the following postulates. 1) Pollen is produced annually and deposited *in situ* in sediment; its exine or outer layer is composed of sporopollenin, which is very stable and does not exchange carbon with sediment. 2) Pollen undergoes limited translocation in loess-paleosol, <10 cm (Wu *et al.* 1992), because at the loess/desert belt (which belongs to the arid steppe environment, with low annual precipitation and scarce vegetation cover), soil development is very weak, although mixing and percolation occur in the pedogenic process (Dimbleby 1985). 3) Pollen is an abundant plant fossil, well preserved in sediment, and accelerator mass spectrometry (AMS) technology allows the analysis of  $<500 \mu g$  of carbon. 4) Pollen is predominantly of terrestrial origin, and its age should not be influenced by reservoir effects (Regnell 1992).

Brown *et al.* (1989, 1992) showed that separation of pollen and pollen concentrates from lake sediments provides a dating material that is potentially superior to any sediment components used previously. Long, Davis and Lanois (1992) developed procedures for physically removing non-pollen contaminants from pollen concentrates, achieving high-precision dates. Based on their work, we developed and modified a procedure suitable for the extraction of pollen concentrate from eolian sediments.

#### **DESCRIPTIVE BACKGROUND**

This study focused on five profiles at the present northwestern margin of the Loess Plateau (Fig. 1), *i.e.*, at the loess/desert transitional belt. Loess-mantled hills, sandy loess platforms (*yuan*) and seasonally flowing streams characterize the landscape of this area. In winter, the area has predominantly northerly and northwesterly winds, generating frequent dust storms. In summer, the main winds are southeasterly, moving inland off the ocean and producing 80% of the annual precipitation that falls as rainstorms (Zhang and Lin 1992). Mean annual temperature is 8°C; mean annual precipitation is *ca.* 200–400 mm.



Fig. 1. Map showing study sites and the northern margin of the Loess Plateau. 1. Shiyaowan; 2. Yangtaomao; 3. Xuanheze; 4. Midiwan; 5. Baxie at the loess/desert transitional belt. The inset map shows China and the Yellow River.

The Xuanheze profile  $(37^{\circ}39'N, 108^{\circ}37'E)$ , at a mean elevation of 1400 m) has a 500-cm exposure of sediment (Figs. 1 and 2a). It is located on the second terrace of Xuanhe, a seasonal tributary of the Heiheze River, which is a second-order tributary of the Huanghe (Yellow River). The lowest 60 cm is a grayish-green silty mud. From 440 to 200 cm depth it is characterized by an eolian sand. Gray-ish-white marl occurs between 200 and 50 cm from the top of the sediment column. The top 50 cm consists of a grayish-green silty mud. The Midiwan profile (Fig. 1) occurs *ca*. 500 m to the east, located on the first terrace of Xuanhe River, with 1380 cm of peat sediment. Based on the <sup>14</sup>C chronology, peat deposition in the lake basin began *ca*. 13,000 BP, reflecting the onset of a warm-moist climate in the last deglaciation. Peat deposition reached maximum thickness *ca*. 10,000–7500 BP, during the Holocene Optimum. The section of the Midiwan profile is high-resolution and appears to be continuous. A detailed study of this profile has been given elsewhere (Zhou *et al.* 1996; Zhou 1995).

The Yangtaomao profile ( $38^{\circ}48'N$ ,  $110^{\circ}27'E$ , at a mean elevation of 1400 m) consists of sand and interstratified paleosol exposed by wind erosion at the top of a loess-mantled hill (Zhou *et al.*1996). The section of the studied profile (Figs. 1 and 2c) consists of the upper 350 cm, below which loess is found. From 350-315 cm is an immature sandy paleosol. Pale-yellow eolian sand at 315-157 cm depth contains a 38-cm-thick immature sandy paleosol (242-204 cm depth). The top 157 cm consists of a dark grayish-brown paleosol complex interstratified with an eolian sand layer 40-cm thick (78-38 cm depth). This soil complex consists of well-developed sandy steppe-type paleosol and is poorly consolidated. Located *ca*. 500 m to the west of the Yangtaomao section is a 2-m-thick exposure of the Shiyaowan profile (Figs. 1 and 2b). This section consists of eolian sand (200-150 cm) and grayish-brown paleosol (150-0 cm).

The Baxie loess section is located at the southwest margin of the Loess Plateau, 65 km southwest of Lanzhou (35°33'N, 103°35'E, 2000 m elevation), The stratigraphy of this profile was studied in detail by Zhou *et al.* (1992) and An *et al.* (1993). Here, we tried to redate one sample from the Baxie paleosol (where there was evidence for an increase of precipitation during the Younger Dryas interval), in order to compare dating of pollen concentrate with that of humic acid.

# METHODS

Separation of the pollen concentrates is based on both particle size and specific gravity (Table 1). Pollens from semi-arid to arid zones are mostly silt-sized. Before removing sand grains (> 40  $\mu$ m) from the silt fraction by sieving, we used 5% HCI to disaggregate carbonate clay. The mesh size of the sieve was also selected to eliminate organic detritus larger than most pollen. Then, we further



TABLE 1. Flowchart for Pollen Concentrate Preparation

removed CaCO<sub>3</sub> with 5% HCl, humic acid with 10% NaOH, and silica with 40% HF. Because pollen from semi-arid to arid zones ranges in specific gravity from 2.2 to 1.5, we floated pollen out of the sediment using a heavy liquid (KI + HI + Zn) with a specific gravity (sp. gr.) of 2.2 g cm<sup>-3</sup>. The liquid was then diluted with water to sp. gr. of < 1.5 to sediment the material. To increase pollen recovery, the extraction step was repeated twice, as sand, loess and paleosol are low in pollen compared to other sediments. Ke (1994) pointed out that flotation with a heavy liquid of sp. gr. ca. 2.2 is especially efficient for pollen extraction from loess-paleosol samples. Before we carried out the flotation method with a heavy liquid of sp. gr. 2.2, we tried the method proposed by Brown et al. (1992), but failed to extract enough pollen from eolian sand and paleosol for dating. Therefore, we developed a method combining particle size separation with floating in a heavy liquid, which enabled successful extraction of pollen from sand and paleosol. After pollen extraction, we found that our pollen concentrates consisted of 50% pollen (mainly Artemisia and Chenopodiaceae), 45% phytoliths (considered to be a reliable alternative source for <sup>14</sup>C dating (Wilding 1967; Mulholland 1992)) and 0-5% organic fragments. Careful microscope identification showed no soil fungi spores in the samples we studied. Because spores of soil fungi are the products of humid forest soil, generally they adhere to residues of higher plants (Cao 1980). Since the objective is to date the original carbon, any younger material adhering to the pollen particles must be removed. We used an oxidizer of 2% NaClO<sub>2</sub> for cleaning pollen concentrates. Because NaClO<sub>2</sub> is a very strong oxidizer and destroys lignin and humic acids, but is also corrosive to pollen grains, we used the weak solution for a short period, e.g., no longer than 5 min. Pollen samples were then treated further with 10% HCl, rinsed with distilled water and dried.

Ca. 500 mg of extracted pollen concentrates was combusted in a sealed tube with CuO. The  $CO_2$  was reduced to graphite (Slota *et al.* 1987) for AMS dating (Donahue, Jull and Zabel 1984).

# **RESULTS AND DISCUSSION**

Pollen dating results from the various profiles are shown in Tables 2–4. In order to evaluate the reliability of pollen concentrate dating, we re-collected four samples. Three samples are from the well-dated Midiwan peat profile (Zhou *et al.* 1996), containing wood, charcoal, and peat (Table 2) and the others from Baxie paleosol for pollen concentrate dating. We can compare ages of wood cellulose and pollen (958 cm, 10,370  $\pm$  70 (AA-12190); 10,360  $\pm$  65 (AA-12186)), peat and pollen (1112 cm, 11,755  $\pm$  95 (AA-16691); 11,765  $\pm$  80 (AA-12188)), and charcoal and pollen (1143 cm, 12,750  $\pm$  95

Shiel Dating Material Henri Materia				
Lab Code*	Depth (cm)	Material dated	<sup>14</sup> C age (yr BP)	
AA-12190	958	Wood cellulose	10,370 ± 70	
AA-12186†	958	Pollen concentrate	10,360 ± 65	
AA-16691	1112	Fine peat	11,755 ± 95	
AA-12188†	1112	Pollen concentrate	11,765 ± 80	
AA-12196†	1143	Pollen concentrate	12,390 ± 100	
AA-16393	1143	Charcoal	12,750 ± 95	
AA-16389	973	Charcoal	10,610 ± 80	
AA-6442	925	Humic acid	9695 ± 95‡	
M12566†	925	Pollen concentrate	10,630 ± 70‡	

TABLE 2. AMS Result Comparison of Pollen Concentrates with Other Dating Material from Midiwan Peat and Baxie Loess Profiles

\*AA- = NSF-AMS Facility, The University of Arizona

†Re-collected samples for pollen dating research

tfrom Baxie loess profile

(AA-16393); 12,390  $\pm$  100 (AA-12196)). These values are in good agreement and indistinguishable within each pair within one standard deviation, indicating that pollen concentrate dating is reproducible. As for the Baxie paleosol (An *et al.*1993), which indicates a precipitation increase during the Younger Dryas on the Loess Plateau, the new pollen-concentrate age (935 cm depth, 10,630  $\pm$  70 BP (AA-12566)) is *ca.* 900 yr older than the previous humic-acid age (925 cm depth, 9695  $\pm$  95 (AA-6442)), although there is 10 cm difference in sample depth. The new pollen concentrate age on Baxie paleosol agrees with a recent charcoal age on the Midiwan peat, which is regarded as an indicator of increased precipitation in the mid Younger Dryas (AA-16389, 10,610  $\pm$  80; Zhou *et al.* 1996). This obviously suggests that pollen concentrate dating improves upon the age of humic acid from paleosol.

We applied this method to the chronology of these various profiles, with very interesting results (Tables 3 and 4; Fig. 2). Terrace two in the Xuanheze profile (Fig. 2a) shows that pollen concentrate ages form a good age sequence, except for one date from eolian sand: at 330 cm depth, AA-16120, the pollen concentrate dated *ca.* 10,000 yr younger than the one above at 200 cm depth, AA-16119. Since the sand layer generally deposits rapidly, it is hard to believe that it took *ca.* 10,000 yr for the deposition of *ca.* 250 cm-thick sand. Accordingly, sample AA-16120 must contain younger pollen or organic detritus, and it should be discarded. The silty mud layer of the profile may correspond to the mud layer of the Beizhuangcun profile from the Loess Plateau (28,400  $\pm$  340 BP, XLLQ28, Zhou, Zhou and Head 1990) and possibly further to the later oxygen isotope stage 3 of deep sea cores (Imbrie *et al.* 1984), reflecting cool and wet climatic conditions in East Asia. Moreover, we expect that dating of pollen concentrates from the Shiyaowan paleosol (Fig. 2b and Table 4) should provide reliable ages. These results generally agree with those from Midiwan peat (Zhou *et al.* 1996), indicating that warm and moist monsoon climate was favorable to paleosol development between *ca.* 11,680 and 13,700 BP, during the climatic conditions of the last deglaciation.

Lab code	Depth (cm)	Material dated	<sup>14</sup> C age (yr BP)
AA-16118 AA-16119 AA-16120 AA-16121	170 200 330 490	Pollen concentrate Pollen concentrate Pollen concentrate Pollen concentrate	$27,910 \pm 330 27,540 \pm 320 17,315 \pm 130 28,730 \pm 790$

 TABLE 3. AMS Dating Result of Pollen from Xuanheze Eolian

 Sand-Paleosol Profile

Table 4 and Figure 2c show pollen concentrate ages obtained from Yangtaomao paleosol and eolian sand. One sample at 158 cm depth gives a pollen concentrate age *ca.* 3000 yr too old, and another one at 258 cm depth is *ca.* 1600 years too young, again providing evidence that pollen concentrated from eolian sand contains either younger or older materials. However, the rest of the six dates from paleosol have a good age sequence with depth and can be regarded as reliable. Microscopic examination combined with dating shows that pollen concentrate consists of two fractions: 1) pollen deposited simultaneously with sediment, and 2) reworked pollen including older (fossilized) spore pollen preceding sediment deposition *in situ*, or younger pollen transported through hydrological or agricultural activities; both younger and older pollen may come from either distant or local regions. Pollen reworking is generally an indicator of an unstable environment (Zhou *et al.* 1996).

The history of Asian monsoon variation at the loess/desert transitional belt at the end of the last glaciation has been reconstructed on the basis of the <sup>14</sup>C chronostratigraphy in Yangtaomao. The period from *ca.* 11,200 to 10,000 BP (interpolated age) was dominated by a climate marked by cold and dry



Fig. 2. (a) Xuanheze mud-eolian sand, (b) Shiyaowan loess-paleosol, and (c) Yangtaomao eolian sand-paleosol profiles, with AMS <sup>14</sup>C dates of pollen concentrate (*Artemisia*, Chenopodiaceae, *Pinus*, and organic residues). Ages in brackets are derived by interpolation. Dating of pollen concentrates extracted from paleosol or mud creates good age sequences with depth, except on those from eolian sand. Based on dating and lithology, several wet phases are marked by mud or paleosol around the late Pleistocene, last deglaciation, mid Younger Dryas and early Holocene.

Lab code	Depth (cm)	Dating material	<sup>14</sup> C age (yr BP)			
AA-12317	11	Pollen concentrate	<u>6950 ± 60</u>			
AA-12311	22	Pollen concentrate	$7790 \pm 70$			
M-12313	102	Pollen concentrate	9030 + 70			
AA-12315	158	Pollen concentrate	$13.260 \pm 90$			
AA-12312	258	Pollen concentrate	9105 + 70			
AA-12316	325	Pollen concentrate	$11.250 \pm 80$			
AA-12306*	12	Pollen concentrate	11.680 + 80			
M-12307*	140	Pollen concentrate	$13.720 \pm 130$			

TABLE 4. AMS Dating Results of Pollen from Yangtaomao and Shiyaowan Eolian Sand-Loess-Paleosol Profiles

\*Samples collected from Shiyaowan profile

winter monsoon during which 167 cm of eolian sand was deposited. However, the continued influx of an accretionary sandy soil suggests the continuation of strong winter monsoon conditions and increased precipitation. From 10,000–9000 BP, the well-developed paleosol reflects strengthening summer and weakening winter monsoon climates. A rapid decrease in the strength of the summer monsoon from ca. 9000 until 8000 BP, as recorded in the deposition of an eolian sand unit, indicates the return of the strong winter monsoon. After ca. 7000 BP, the profile was eroded. The late-glacial climatic oscillation evidence described here supports the pollen analysis results obtained from Midiwan profile (Zhou *et al.* 1996).

# CONCLUSION

1. The comparison of <sup>14</sup>C ages of pollen concentrate with wood, charcoal, peat and paleosol humic acid clearly demonstrates that pollen dating provides useful age information for the Loess Plateau chronology. The newly obtained dating of pollen concentrate from the Baxie paleosol (10,630  $\pm$  70 BP) agrees with charcoal dating of Midiwan peat (10,610  $\pm$  80 BP). Our study shows that pollen concentrates may be a better material for dating paleosol than humic acid on the condition that pollen was deposited simultaneously with sediment in a stable sedimentary environment, such as pollen from paleosol or peat, whereas pollen preserved in an unstable environment (*e.g.*, pollen from eolian sand or loess in an area where the winter monsoon is predominant and strong) does not necessarily provide reliable data. Therefore, a reliable chronology of the loess-paleosol sequence cannot be established without pollen separation, where suitable dating materials such as wood and charcoal are lacking. We intend to experiment further with loess-paleosol sequences for which detailed stratigraphic work has been done in order to improve the chronology of the Loess Plateau.

2. Four major wet phases have been dated from the Late Pleistocene to the early Holocene. Phase 1 is marked by lacustrine mud, which developed in Xuanheze *ca.* 29,000 yr BP; a similar sedimentary phase can be found in many locations on the Loess Plateau, reflecting a wet-cool climate (based on finding pollen of *Abies* and *Pinus* in Beizhuangcun mud (Zhou, Zhou and Head 1990)). In phase 2, *ca.* 13,700 BP, paleosol started to develop as shown in Shiyaowan, indicating the arrival of the last deglaciation. Phase 3 is recorded in Yangtaomao sandy paleosol (10,500–10,200 BP) during the Younger Dryas interval from 11,200 to 10,000 BP, indicating increased precipitation. In phase 4, from 10,000 to 9000 BP, paleosol was well developed, reflecting warm-moist climatic conditions and arrival of the Holocene.

Precipitation variability is especially important for the understanding of paleomonsoon climate change at the loess/desert transitional belt of East Asia and can be used as a historic analog for climate prediction.

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# CALIBRATION OF RADIOCARBON DATES FOR THE LATE PLEISTOCENE USING U/Th DATES ON STALAGMITES

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**ABSTRACT.** Twenty paired <sup>14</sup>C and U/Th dates covering most of the past 50,000 yr have been obtained on a stalagmite from the Cango Caves in South Africa as well as some additional age-pairs on two stalagmites from Tasmania that partially fill a gap between 7 ka and 17 ka ago. After allowance is made for the initial apparent <sup>14</sup>C ages, the age-pairs between 7 ka and 20 ka show satisfactory agreement with the coral data of Bard *et al.* (1990, 1993). The results for the Cango stalagmite between 25 ka and 50 ka show the <sup>14</sup>C dates to be substantially younger than the U/Th dates except at 49 ka and 29 ka, where near correspondence occurs. The discrepancies may be explained by variations in <sup>14</sup>C production caused by changes in the magnetic dipole field of the Earth. A tentative calibration curve for this period is offered.

# INTRODUCTION

At the International Radiocarbon Conference in Bern in 1978, it was pointed out that certain <sup>14</sup>C dates for the Late Pleistocene appear to be several thousand years too young when compared with corresponding uranium series dates (Vogel 1980; Barbetti 1980). This prompted us to undertake a more detailed comparison of the two dating techniques using a slender 2.8 m stalagmite from the Cango Caves in the Cape Province of South Africa. This specimen, which stood at the far end of Cave II, was sawed into sections and removed during several visits over a period of four years. It proved to be ideal for the purpose since it had a coarse crystalline nonporous structure that ensured closed-system conditions. Its base turned out to date back nearly 50,000 yr; water was dripping onto the tip so that it was still growing at the time of removal. Preliminary results for the comparison were reported at the next Radiocarbon Conference in Seattle in 1982 (Vogel 1983).

Unfortunately there is a gap in the sequence, because the stalagmite's growth halted between *ca*. 17,000 and 7000 yr ago. This gap could partially be filled by samples of two similarly crystalline stalagmites from Lynd's Cave in Tasmania (Vogel 1987; Goede and Vogel 1991).

# Method

We cut samples along the growth axis of the three stalagmites and performed the <sup>14</sup>C and U/Th analyses using the same pieces in each case. Carbon dioxide for measurement of the <sup>14</sup>C in a CO<sub>2</sub> proportional counter was generated by adding acid, and the remaining solution was processed for the U/ Th isotope analysis, using standard  $\alpha$ -counting techniques.

# RESULTS

The results for the Cango stalagmite are given in Table 1, and those for Lynd's Cave are listed in Table 2. Two adjustments need to be made to the dates.

1. The <sup>14</sup>C ages must be corrected for the apparent initial age of the calcite when it is precipitated from the dripping groundwater.

As previously shown (Vogel 1983), the first six  $^{14}$ C dates for the upper 95 cm of the Cango stalagmite show complete concordance with the U/Th dates if 1450 yr are subtracted from the initial age and the results are calibrated with the tree-ring calibration curve (*e.g.*, Stuiver and Pearson 1993). This correction of 1450 yr is thus adapted for the older sections, too. It is, of course, possible that the apparent initial age varied with differing climatic conditions, but the shift would not have been more than a few hundred years, since this figure has been found to apply to groundwater in limestone areas worldwide. The adjustment for the Lynd's Cave stalagmites is less certain. 1500 yr are subtracted from these  $^{14}$ C dates, but an error of ±500 should be assumed. When these results were first presented (Vogel 1987) 2160 yr was subtracted, based on a comparison with parallel electron spin resonance (ESR) dates, but subsequent tree-ring calibration data for the early Holocene (Kromer and Becker 1993) have shown this correction to be too large.

2. The U/Th ages need to be corrected for any initial <sup>230</sup>Th that may have been incorporated into the stalagmite during formation.

Most of the samples had very small amounts of  $^{232}$ Th. For those that did show measurable amounts of the isotope, the adjustment was <230 yr, assuming an initial  $^{230}$ Th/ $^{232}$ Th activity ratio of  $1 \pm 0.5$ . An exception was the Lynd's Cave stalagmite L1, which had a very low uranium content (*ca.* 0.02 ppm) and consequently required a correction of 620 yr in one case. The U/Th results from this specimen are therefore less precise than the rest.

TABLE 1. <sup>14</sup>C and U/Th data for Cango stalagmite V3. Column 2 gives the distance of the sample from the tip of the stalagmite; column 4 the apparent <sup>14</sup>C age adjusted for isotope fractionation using the <sup>13</sup>C/<sup>12</sup>C ratio. Columns 7, 8 and 9 list the activity ratios of the U and Th isotopes and column 10 the U-series ages adjusted for initial Th.

	Distance	I ab code	Apparent <sup>14</sup> C	Anal.	U cons	<sup>234</sup> U	<sup>230</sup> Th	<sup>230</sup> Th	U/Th
Sample	(mm)	(Pta-)	age (yr BP)	no.	(ppm)	<sup>238</sup> U	<sup>232</sup> Th	<sup>234</sup> U	age (yr BP)
V3/1a	5	2256	1530 ± 50	U-003	0.068	3.70			recent
V3/1h	14	2435	1490 ± 55						
V3/1c	314	2579	3270 ± 45	U-107	0.062	3.43	10	0.019	$1870 \pm 220$
V3/1d	492	2289	4660 ± 60						
V3/2a	715	2580	5730 ± 50	U-108	0.098	3.61	28	0.044	$4710 \pm 350$
V3/2h	887	2581	6380 ± 60	U-109	0.109	3.59	101	0.057	$6240 \pm 400$
V3/2c	974	2436	$15,400 \pm 140$	U-112	0.091	3.14	217	0.152	$17,400 \pm 800$
V3/6a	989	3081	$15,790 \pm 50$	U-263	0.109	3.15	>>	0.166	19,300 ± 800
V3/6h	1157	3087	19,180 ± 50						
V3/6c	1304	3090	$21,610 \pm 70$	U-242	0.140	2.65	>>	0.227	$27,200 \pm 3000$
V3/3m	1517	3084	$26.710 \pm 110$	U-265	0.078	3.18	103	0.227	$27,200 \pm 1300$
V3/3h	1598	3277	$28.380 \pm 150$	U-282	0.101	3.10	256	0.264	$32,100 \pm 2100$
V3/4a	1679	2657	30,300 ± 610	U-118	0.106	3.02	>>	0.242	$29,200 \pm 800$
V3/4c	1759	3078	$30,130 \pm 180$	U-264	0.083	3.22	>>	0.243	$29,300 \pm 3000$
V3/4d	1842	3077	$30,170 \pm 180$	U-266	0.073	2.99	153	0.282	$34,400 \pm 2200$
V3/4h	2030	2658	$30,110 \pm 500$	U-119	0.083	3.18	173	0.287	$35,200 \pm 1000$
V3/5a	2101	3279	$32,670 \pm 330$	U-269	0.077	3.03	>>	0.294	$36,400 \pm 1800$
V3/5c	2181	3893	33,870 ± 330	U-325	0.087	3.05	155	0.305	$37,700 \pm 1200$
V3/5b	2288	2689	35,900 ± 280	U-136	0.058	3.11	255	0.311	$38,700 \pm 900$
V3/7a	2409	3278	$36,520 \pm 300$	U-270	0.109	2.84	254	0.329	$41,300 \pm 1200$
V3/7d	2491	3887	37,580 ± 490	U-324	0.117	2.85	200	0.348	$44,100 \pm 1400$
V3/7c	2589	3884	41,330 ± 630	U-323	0.107	2.90	200	0.352	$44,700 \pm 1400$
V3/7b	2698	3280	47,660 ± 1120	U-283	0.119	2.73	420	0.381	49,200 ± 1300

	Distance	Lab code	Apparent <sup>14</sup> C	Anal.	U cons	<sup>234</sup> U	<sup>230</sup> Th	<sup>230</sup> Th	U/Th
Sample	(mm)	(Pta-)	age (yr BP)	no.	(ppm)	<sup>238</sup> U	<sup>232</sup> Th	<sup>234</sup> U	age (yr BP)
Lynd's C	Cave stalag	gmite L2							
L2/1	717	3792	10,980 ± 110	U-305	0.307	3.12	171	0.104	$11.740 \pm 500$
L2/2	515	3791	$11,800 \pm 100$						,,
L2/4	334	3707	12,770 ± 100	U-293	0.262	2.97	236	0.117	$13,300 \pm 360$
L2/3	120	3708	$13,450 \pm 130$	U-294	0.305	2.85	442	0.128	$14,570 \pm 420$
Lynd's Cave stalagmite L1									
L1/3	1380	3762	8320 ± 90	U-301	0.019	2.32	12	0.069	$7070 \pm 570$
L1/6	980	3731	10,970 ± 100	U-299	0.016	2.69	64	0.099	$11,080 \pm 800$
L1/7	614	3797	$12,400 \pm 110$						,
L1/9	79	3713	14,170 ± 60	U-295	0.024	2.80	44	0.114	12,840 ± 750

TABLE 2. <sup>14</sup>C and U/Th data for the Tasmanian stalagmites. (See Table 1 for explanation.)

# DISCUSSION

With these two adjustments to the ages, the results for the Cango stalagmite are plotted (Fig. 1.) against the distance from the tip. A few of the U/Th analyses had relatively large margins of error, but on the whole they show a nearly linear growth of the stalagmite between 44 ka and 18 ka ago, with a somewhat slower growth rate between 49 ka and 44 ka. The <sup>14</sup>C ages, on the other hand, show major deviations from this linear growth curve, especially for the period beyond 30,000 yr BP, where they are consistently several thousand years younger than the uranium series dates.



Fig. 1. <sup>14</sup>C dates ( $\times$ ) adjusted for initial age of 1450 yr and parallel U-series dates (+) for the Cango stalagmite showing the large discrepancy between age-pairs between 30 ka and 45 ka

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Now, it is well known that samples older than 30,000 yr are prone to produce <sup>14</sup>C ages that are too young due to contamination by small amounts of more recent carbon. The fact that the <sup>14</sup>C date for the bottommost sample agrees with the U/Th age to within 1 $\sigma$  if the correct <sup>14</sup>C half-life (5730 yr) is used (49,200 ± 1300 (U/Th) – 47,550 ± 1120 (<sup>14</sup>C) = 1650 ± 1720), indicates, however, that contamination cannot in this case explain the discrepancies between the two methods. Of special significance is near identity between the <sup>14</sup>C and U/Th ages *ca.* 29,000 yr ago. This will be discussed briefly below.

For the period between 10 and 20 ka we need to consider the age-pairs derived from the two Lynd's Cave stalagmites (Table 2). As previously noted (Vogel 1987), these data also show considerable discrepancies between the <sup>14</sup>C ages and the U/Th ages.

More recently, a comprehensive set of age comparisons has been published using coral from Barbados (Bard *et al.* 1990, 1993), which confirm that <sup>14</sup>C dates between 12 ka and 22 ka are several thousand years too young. The U/Th dates were produced by thermal ionization mass spectrometry (TIMS), which gives greater precision than the conventional  $\alpha$ -counting technique. In Figure 2 these results are presented together with our measurements in this time range; two pairs from Cango (V3) are also included to extend the age range. All except two of our data points correspond to the TIMS results to within one sigma, thus confirming that the <sup>14</sup>C age discrepancy is a worldwide phenomenon and that the stalagmite data do represent the actual situation.



Fig. 2. Calibration curve for conventional <sup>14</sup>C dates after Bard *et al.* (1993) (×) with our age-pairs for the Lynd's Cave stalagmites (L1 and L2) (Table 2) and the two Cango Cave (V3) age-pairs in this time range (Table 1)
Using the comparisons obtained from the Cango Cave stalagmite, a calibration curve for the period 25 ka to 50 ka is plotted in Figure 3. The four samples with U/Th error margins greater than  $\pm$  2000 yr (V3/6c, V3/3b, V3/4c and V3/4d) are not included in the figure because they are not very meaningful. The margins of error of this curve are still relatively large, but for the present it can be used in cases where absolute date estimates are required rather than <sup>14</sup>C ages. It is evident from this figure that <sup>14</sup>C dates between 35 ka and 45 ka are *ca*. 5000 yr too young. Recently, one additional age comparison has been published (Bischoff *et al.* 1994) that confirms this discrepancy.



Fig. 3. Calibration curve for conventional <sup>14</sup>C dates between 20 ka and 50 ka using the data for the Cango Cave stalagmite, V3 (Table 1)

The authors find that their <sup>14</sup>C ages on charcoal  $(37 \pm 1 \text{ ka})$  are much younger than U/Th ages on bracketing flowstone  $(42.6 \pm 1.1 \text{ ka})$  in a cave near Barcelona, Spain. If the <sup>14</sup>C date is calibrated, using Figure 3, it overlaps with the U/Th date.

Not shown in Figure 2 is the isolated coral sample of Bard *et al.* (1993) that gave an age-pair at variance with our results at *ca.* 30 ka (Figure 3). Their measurements show the <sup>14</sup>C date to be  $4780 \pm 380$  (2 $\sigma$ ) yr younger than the U/Th date of  $30,225 \pm 160$  yr, whereas our age-pair at 29,000 yr shows almost no difference between the two techniques: at  $29,200 \pm 800$  yr (U/Th) the <sup>14</sup>C age is only 350 yr younger. Other age-pairs in this time range, on the other hand, do seem to support our finding: the five comparisons listed by Barbetti (1980) between 26,000 yr and 32,000 yr individually and collectively show virtually no discrepancy between U/Th and <sup>14</sup>C ages. The weighted averages for these six samples give:

$$29,690 \pm 680 \text{ yr} (\text{U/Th}) - 29,800 \pm 760 \text{ yr} (^{14}\text{C}) = -110 \pm 1020 \text{ yr}$$
.

#### CONCLUSION

If <sup>14</sup>C dating gives the correct age at 29,000 yr, it implies that a drastic decrease in the <sup>14</sup>C levels of the atmosphere occurred. This seems to be indicated by the very slow decrease in the <sup>14</sup>C ages between the U/Th dates of 35,000 and 29,000 yr (Fig. 1). The effect could have been caused by a major increase in the strength of the Earth's dipole field, and there is some evidence that this did happen. Most estimates of the global dipole field intensity during the last 120,000 yr indicate considerably lower values than today, but at least one of them (Tric *et al.* 1992) shows near-normal dipole intensity between 55 and 47 ka, between 33 and 25 ka and after 12 ka, with distinctly lower values in between. The geomagnetic modulation of the <sup>14</sup>C production rate could thus explain qualitatively, at least, the general trend of the data presented here.

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# BOMB <sup>14</sup>C RECORDED IN LAMINATED SPELEOTHEMS: CALCULATION OF DEAD CARBON PROPORTION

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ABSTRACT. We performed radiocarbon measurements using accelerator mass spectrometry (AMS) on 6 stalagmites, 3 stalactites and 7 seepage waters from four different caves in Southwest France and Belgium in order to calculate the dead carbon proportion (dcp). All the speleothems studied are modern and annually laminated, which offers the advantage of an accurate chronology, with better than one-year resolution. Coupled with the fact that very little calcite is necessary for an AMS measurement (between 1.5 and 7 yr of calcite deposit), we obtained dead carbon values within an uncertainty limit of  $\pm 1.5\%$ . Results show that the dead carbon proportion varies from 9.2% to 21.9% for calcite deposits and from 3.6% to 21.9% for water. In each sampling site, the dcp is homogeneous. Although the inter-site dcp varies by >11%, its average value of 15.5%  $\pm 4.4$  still lies within the uncertainty range of the accepted value of 15%  $\pm 5$  (dilution factor of 0.85  $\pm$  0.5). We compare the average dcp of each site with the local geology, vegetation and climate. Given similar geology and temperature, the highest dcp values are found under forest cover; dcp difference is up to 9%. However, the Belgian site, which is also under a forest, shows a dcp very close to the dcp found under grassland sites of Southwest France, which proves that other unknown factors may play an important role in dissolution processes. Secondary calcite deposition and redissolution in the soil zone or more likely in the fracture system before reaching the cave itself could also explain the inter-site differences. The IAEA isotopic model (Pearson model adapted for open systems) is in good agreement with the measured activities.

### INTRODUCTION

Recent studies have shown that many speleothems possess annually deposited growth laminae; they can be visible (Genty 1993; Genty and Quinif 1996) or luminescent (Baker *et al.* 1993; Shopov and Dermendjiev 1990). The alternation of white porous and dark compact laminae represents the calcite deposit of one year, which gives modern laminated speleothems a very accurate chronology. It is not rare to have speleothems with continuous laminae for >100 yr, but in the case of longer durations, laminae are frequently interrupted by discontinuities. The search for paleoclimatic signals from these laminae has already started: correlation has been found between the laminae thickness and the pluviometry (Genty and Quinif 1996; Railsbak *et al.* 1994), and a stable isotopic seasonal signal has been observed in several stalagmites (Genty, Quinif and Keppens 1995).

Speleothem carbon has two principal sources: 1) the  $CO_2$  of the soil above the cave; 2) the carbonate of the limestone where the cave developed and/or of secondary calcite deposits of the unsaturated zone. The soil  $CO_2$  comes from root respiration and microbial decomposition of organic matter; it is thus linked to atmospheric  $CO_2$  (Dörr and Münnich 1986). Carbon from limestone is generally so old as to have a negligible <sup>14</sup>C activity; however, if it comes from secondary deposits, its activity can be significant.

The determination of speleothem <sup>14</sup>C activity has at least two essential purposes:

 To estimate the age of the deposit. Even if it is not now the most appropriate method precisely because of the unknown dead carbon proportion, it has been the only feasible method for a long time (Broecker and Olson 1960; Geyh and Franke 1970). It is still used because of its cost advantage over the U/Th mass spectrometric method and because it is a means of checking U/ Th ages for the last 40 ky (Bastin and Gewelt 1986; Brook and Nickmann 1996; Genty *et al.* 1996; Gewelt, 1985; Geyh and Henning 1986; Goede and Vogel 1991; Holmgren, Lauritzen and Possnert 1994; Issar *et al.* 1992; Pazdur, Pazdur and Pawlyta 1995; Railsback *et al.* 1996; Talma and Vogel 1992; Vogel 1983).

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2. To reconstruct atmospheric <sup>14</sup>C activity through time; in this case, the chronology is given by U/Th ages (Holmgren, Lauritzen and Possnert 1994; Vogel 1983). Atmospheric <sup>14</sup>C wiggles observed during the Holocene in tree-ring dated wood (Wigley and Kelly 1990) are probably due to the Earth's geomagnetic field changes (Bard, Hamelin and Fairbanks 1990; Mazaud *et al.* 1991; Vogel 1983), solar activity variations (Suess and Linick 1990) and the capacity of the ocean to absorb CO<sub>2</sub> (Goslar *et al.* 1995). During historical time, <sup>14</sup>C variations include an abrupt increase between AD 1500 and 1700 (the de Vries effect), a slight decrease of 2% since the beginning of the industrial period (AD 1850; the Suess effect), and finally, a massive production of <sup>14</sup>C in the atmosphere because of nuclear bomb tests (maximum increase of >100% in 1963–1964; Levin *et al.* 1992).

There are major problems in both cases: 1) the initial activity is not known; in other words the dcp must be estimated (or the dilution factor q that is its complement; Gewelt 1986); 2) the dcp possibly varies with time.

The dcp has been calculated in several different ways (Table 1):

- by measurement of the <sup>14</sup>C activity on the top of still-dripping stalagmites (Vogel 1983; Gewelt 1985, 1986; Bastin and Gewelt 1986); using the classical  $\beta$ -counting method, one must sample >20 g of calcite (more often between 30 and 100 g); thus, the error margin is large because sampling takes into account deposits of different ages (*e.g.*, a dcp of 30% was found on the top of three active Belgian stalagmites, whereas the value taken into account was 15% (Gewelt 1986));
- by age-distance interpolation up to the top of the stalagmite (linear least-square fit); the hypothesis of a constant growth rate must be considered critically here, because stalagmite growth rates are highly variable even over short periods (*i.e.*, from 0.1 to 1.0 mm yr<sup>-1</sup>; Genty and Quinif 1996; Genty *et al.* 1996);
- by comparison with pollen extracted from the speleothem; this technique can be used in areas where the emergence or disappearance of a taxon is abrupt (*i.e.*, emergence of *Tilia* at the Atlantic in Belgium; Bastin and Gewelt 1986); but this is not an accurate method because there is very little pollen in speleothem calcite (up to 5 grains gm<sup>-1</sup> of CaCO<sub>3</sub>) and a great quantity of calcite is required (>100 g);
- by comparison with U/Th ages (Vogel 1983; Holmgren, Lauritzen and Possnert 1994); the accuracy depends on the U/Th errors, which are great for recent deposits (very little thorium).

In order to attempt more accurate dead carbon inferences we have combined these methods, using 1) annually laminated speleothems, which give an excellent chronology for the last few decades, and 2) AMS techniques that permit the analysis of *ca.* 10 mg of calcite, which is, in the most favorable cases, 1 yr of deposits, and more often, 2-7 yr.

Our purpose is to compare dcp's in modern deposits and water from four sites and to explain the observed variability in relation to the geology, vegetation and climate.

## SITE AND SAMPLE DESCRIPTIONS

Samples come from caves at Han-sur-Lesse cave (Belgium; 50°08'N, 5°10'E, elevation 180 m); Villars (Dordogne, France; 45°30'N, 0°50'E, elevation 175 m), La Faurie (Dordogne, France; 45°08'N, 1°11'E, elevation 225 m) and Proumeyssac (Dordogne, France; 44°55'N, 0°56'E, elevation 155 m).

	Dibution		
Speleothem type, locale	factor q*	Method	Reference
Stalagmites – Belgium	0.85 ± 0.05	Pollen correlation	Bastin and Gewelt 1986
Stalagmite crust on a bone (called travertine)-Moaning Cave, California, USA	0.915	<sup>14</sup> C age-distance interpolation (linear least square fit)	Broecker and Olson 1960
Stalagmites and stalactites – Red Spider Cave, Georgia, USA	0.85	Bibliography†	Brook and Nickmann 1996
Stalagmites and flowstones – Belgium	0.85	Pollen correlation, bibliography	Gewelt 1985
Stalagmites and flowstones – Belgium	0.85 (est.); 0.69; 0.72; 0.69 (measured)	Pollen correlation, bibliography	Gewelt 1986
Stalagmite – Central Europe	0.85	Bibliography	Geyh and Franke 1970
Flowstone – Heggen Cave, Germany	0.81	<sup>14</sup> C age-distance interpolation (linear least square fit)	Geyh and Hennig 1986
Stalagmite – Tasmania	0.83 ± 0.05	Bibliography	Goede and Vogel 1991
Stalagmites – Galilee, Israel	0.89	Bibliography	Issar et al. 1990
Speleothems – Cracow– Wielun, Poland	0.83	Bibliography	Pazdur et al. 1995
Stalagmite – Drotsky's Cave, Botswana	0.86	<sup>14</sup> C age-distance interpolation (linear least square fit)	Railsback <i>et al</i> . 1994
Stalagmite – Cango Cave, South Africa	0.83	<sup>14</sup> C age-distance interpolation (linear least square fit)	Talma and Vogel 1992
Stalagmite – Cango Cave, South Africa	0.83	<sup>14</sup> C age of the still active top, comparison with U/Th ages	Vogel 1983

TABLE 1. Examples of Dilution Factors Found in Speleothems

\*q = 1 - (dead carbon proportion) / 100; note that all values are between 0.81 and 0.91, meaning that the dead carbon proportion is between 9 and 19%.

+"Bibliography" = author(s) used former works for their dilution factor q.

All these caves are horizontal and developed in hilly areas. Their entrances are on hill flanks, which explains the low thickness of the terrain above them (<50 m). Each site has characteristic geology, vegetation type and density, soil and limestone thicknesses and climate (Table 2). Examination of aerial photographs and of maps permitted us to estimate the grassland/forest ratio of the watershed above the caves; the Han-sur-Lesse and Villars caves are under forest while La Faurie and Proumeyssac are under grassland.

AMS <sup>14</sup>C activity measurements were performed on 6 stalagmites, 3 stalactites and 7 seepage waters. Except for Proumeyssac (station #1), waters and calcite deposits are from different systems (different places inside the cave). In the Han-sur-Lesse cave, the two stalactites studied correspond to the two stalagmites, which permits the comparison of the isotopic content of stalactite and stalagmite.

<b>TABLE 2. Compa</b>	rison of the I	<b>Dead Carbon</b>	Proportion w	vith the Geolog	3y, Vegetation and Cl	limate Chara	acteristics of th	ne Four Studi	ed Sites
			Limestone			Cumbred /	Cail thinkness	leunne neeM	Mean temn
	dcp(%) in:		thickness		Vegetation types	Grassianu/ forest ratio	ohove cave	mecinitation	Ian. May. Sent
Site	speleothems (waters)	Stratigraphy	above cave (m)	ار%) (%)	and nee density of forest cover	101051 1au (%)	(cm)	(mm)	(°C)
Han-sur-Lesse	13.8 ± 1.5	Givetian	50	0.03	Hornbeams, limes,	0	5–30	787	2.5 - 9.1 - 14.8
(Belgium)	(n = 4)	(Devonian)		(50 m near the	oaks, hazels			(6 yr)	
				entrance, large gallery)					
Villars (Dor-	$21.1 \pm 1.5$	Bajocian	10-40	0.16	Oaks (13 oaks / 100	5±2	0–15	1048	4.1 - 14.0 - 16.0
dogne, France)	(u = 3)	(Jurassic)		(n=2; 3 Jan 1997)	m <sup>2</sup> ; mean dia. = 17 cm), hazels, juni-			(10 yr)	
	$(20.0 \pm 1.5;$ n = 3)				pers, mosses, grass				
La Faurie (Dor-	12.2 ± 1.5	Bajocian	15	0.08	Grass, mosses,	90 ± 5	0-20	861	4.6 - 14.1 - 16.2
dogne, France)	(n = 1)	(Jurassic)		(n=1; 31 Dec 1996)	oaks (20 oaks / 100 m <sup>2</sup> ; mean dia. = 10			(10 yr)	
	$(3.7 \pm 1.5; n=2)$				cm)				
Proumeyssac (Dordogne,	9.2 ± 1.5 (n = 1)	Maastrich- tian (Creta-	20	0.04 (n=1; 2 Jan	Grass, mosses, oaks (9 oaks / 100	85±5	10–30	889 (23 yr)	4.6 - 14.1 - 16.2
France)		ceous)		1997)	$m^2$ ; mean dia. = 24				
	$(5.5 \pm 1.5; n = 2)$				cm)				

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## **METHODS**

## **Analytical Procedures**

Between 19 and 82.3 mg of  $CaCO_3$  were sampled with a microdrill for measurements on speleothems. This corresponds to 1.5 to 7 annual alternations. Laminae counting determined the exact date of the deposition of each sample (Table 3). Water analysis was performed on volumes of 100 to 200 ml.

Calcite powders were reacted with  $H_3PO_4$  in order to obtain  $CO_2$ . The  $CO_2$  gas was graphitized on iron with hydrogen at 650°C for 100 min; residual gas was used for stable isotope measurements on a SIRA spectrometer. Carbon atoms were counted with an accelerator mass spectrometer (Tandetron) at Gif-sur-Yvette (Centre des Faibles Radioactivités, France). Water samples were reacted with  $H_3PO_4$  and degassed  $CO_2$  was used as above for <sup>14</sup>C activity and  $\delta^{13}C$  measurements.

Analytical errors, including laboratory errors, are  $\pm 0.1\%$  for  $\delta^{13}$ C and between 0.6 and 0.9 pMC for  $^{14}$ C activity. Calcite powders are from seasonal laminae, and because two laminae deposit in one year, accuracy of sampling is less than one year.

## **Dead Carbon Calculation**

Dead carbon calculation requires three values: 1) the deposit <sup>14</sup>C activity; 2) the deposit age; 3) the atmospheric <sup>14</sup>C activity during the deposit formation. Because the latter has varied greatly during recent decades, due to nuclear bomb tests, it is necessary to have the most accurate date for the calcite deposits ( $\Delta^{14}$ C varied from 200‰ in 1960 to >1000‰ in 1963 in the Northern Hemisphere; Levin et al. 1992). This is possible with annually laminated speleothems. One must be cautious in attributing an age to a karst water because the latter may be composed of waters of different residence times: typically, a rapid transit occurs a few hours to a few days after a storm event, whereas in dry periods water comes mostly from porous media storage whose age is difficult to know (Ford and Williams 1989). Spring hydrographic analysis can tell us about the storage characteristics of an aquifer (Mangin 1975). Models with different water flow characteristics have been developed: porous (or diffuse), fissure and conduit flows permit plotting of a karst aquifer on a three end-member diagram (Atkinson 1985; Hobbs and Smart 1986). However, very little is known about stalactite flow characteristics; the few published measurements also show the existence of a fast and a slow component (Pitty 1966; Baker et al. 1997; Destombes et al. 1997; Genty et al. in press; Reicher and Trimborn 1995), but only the continuous dripping flow rate and the chemistry under several stalactites will permit quantification of these two components (work in progress). For all the waters, it is assumed that the sampling date corresponds to the water age because of the low thickness of the terrain above the caves and because it has been observed that after a rain storm, flow rate increases in the caves after a few hours or a few days. However, we must keep in mind that dilution with older water is possible.

Because the calcite samples correspond to a depositional time span between 1.5 and 7 yr, we have considered the mean atmospheric <sup>14</sup>C activity of the corresponding period. No direct atmospheric <sup>14</sup>C activity measurements near the studied sites are available. This is why we used the published results of European sites, particularly those from Germany (Levin *et al.* 1992; Levin, Graul and Trivett 1995). For the most recent years, we have extrapolated data from the modeling, with an exponential representation of the decrease of the atmospheric <sup>14</sup>C activity measured at Schauinsland (Germany; Levin, Graul and Trivett 1995).

IADLE J. C JULI		ada to cunco		0 1					a <sup>14</sup> C,	Error	8 <sup>13</sup> C
Samule	Tvpe	Posit	tion		Quantity	Duration o	covered by sa	mple	(pMC)	(pMC)	(‰ PDB ±0.1)
	-1	I I ad			10 mo	1985-199	2: 7 alternatic	ns	9.96	9.6	-8.9
Vil-stm1 14C-C	Stalagmite		ci ure top		50 0 mg	1001-100	5: 3-4 alterna	tions	91.1	0.6	-9.7
Vil-stm5 14C-A	Statagmite				31 5 mg	1958-196	5 (?): 7 altern	ations	118.6	0.6	-10.1
Vil-stm4 14C-A	Stalagmite					0 Can 100	5 / 3 h		89.7	0.6	-11.8
Vil-eau 14C-A	Water	Statu	91# UO				0, ~0 m		03.7	0.8	7.6-
Vil-eau 14C-B	Water	Stati	on #4			<1 Sur 22	90; < 5 h		01.6	80	-10.9
Vil-eau 14C-C	Water	Stati	on #1B		100 ml	22 Aug 15	yo; < 3 n		0.1%		10.0
Fair-eff1 14C-A	Stalactite	Tip (	6 mm). thin	wall	59 mg	1990-199	6; 6 alternatic	SUC	102.6	8.0 9	-10.9
Equipart 14C-A	Water	Stati	on #2		120 ml	24 Aug 19	96; < 48 h		110.3	8.0	-11-
Four-out 14C-R	Water	Stati	on #12		120 ml	24 Aug 19	96; < 48 h		110.0	0.7	-11.7
	Ctala anitia	int Cro	a ortare	tation #1	63 mø	Anr 1996	– Aug 1996		104.0	6.0	-11.2
Prou-poil 14C-A	Statagmuc		will suitace, s	1 11 11 11 11 11 11 11 11 11 11 11 11 1	100 m	26 Aup 19	96: < 0.3 h		108.2	0.7	-10.7
Prou-eau 14C-B	Water	Stat	ion #7		120 ml	26 Aug 19	96; < 0.5 h		108.2	0.7	-14.0
Prou-eau 140-A	water	0141	7.1.10				05. 1 5 alt	arnatione	080	8.0	-9.3
Han-stm4 14C-A	Stalagmite	Gro	wth surface		gm 8.21		Nov. 05. 1 5 al	ternations Iternations	100.5	9.0	-9.1
Han-stm5 14C-A	Stalagmite	ē C	wth surface	:	44.1 mg		Nov 20, 1.0 a Nov 6 alternativ		100-1	0.9	-10.2
Han-stt4 14C-A	Stalactite		(6 mm), thin (4 mm), code	wall ctrow 2/2 filled	рш 7.0С р 3 то	1001-100	5. 4 alternati	ons	9.66	0.8	-10.8
Han-stt5 14C-A	Stalactite	dır	(4 mm), soua	MAIN CH MPINS-	9m 0.70						
TABLE 3 – continua	tion						14.0		5	ULC fron	IAEA model
	$\Delta^{14}C_{atm}$	$a^{14}C_{atm}$	dcp	ð <sup>18</sup> O	$a^{14}C_g$	co <sub>2</sub> /DIC	artCDIC	Avg. cave temp			
Sample	(096)	(pMC ±0.7)	(% ±1.5)	(‰ PDB ±0.1)	(pMC)	frac. (pMC)	(pMC)	(°C ±0.2)	(%oP	DB ±0.1)	(CII TOMO)
1/21 ctm1 140 0	171.0	121 2	20.3	48	117.4	3.0	120.4	1		1	1
	0.1/1	1167	2010 210	-51	112.9	2.8	115.8	1		1	1
	0.121	1501	21.0	, 4 , 1	145.3	2.7	148.1	ł		1	1
VII-Stm4 14C-A	0.001	1.001	0.12		1111	5.0	1135	11.3		-9.45	104.7
Vil-eau 14C-A	109.0	114.8	6.12	ł	8 011	2 C	113.7	11.3	•	-9.45	85.4
Vil-eau 14C-B	100.0	C.411	10.2	1	0.011	9 Y C	113.4	11.3		-9.45	95.8
Vil-eau 14C-C	106.0	C.411	20.0		0.011	0.7	1157				
Fau-stt1 14C-A	129.0	116.9	12.2	4.8	113.1	0.7	/.011			92.0	101.5
Fau-eau 14C-A	106.0	114.5	3.6	:	110.8	4-7	7.011	1.7T		90.0	101 5
Fau-eau 14C-B	106.0	114.5	3.9	1	110.8	2.4	113.2	14.7		07.6	
Proli-not1 14C-A	106.0	114.5	9.2	-5.3	110.8	2.5	113.3	1			: 2
Duoi poir 14C-R	106.0	1145	5.5	;	110.8	2.6	113.4	13.0		2.6	9.26
Prou-cau 14C-D	106.0	114.5	5.5	1	110.8	1.8	112.7	13.0		-9.25	121.7
Uen etm/ 1AC-A	116.0	1155	14.4	-5.9	111.8	2.9	114.8	1		;	:
Uon stm5 140-A	116.0	1155	13.0	4 6	111.8	3.0	114.8	ł		١	1
	137.0	1177	13.2	-5.5	114.0	2.7	116.7	1		ł	:
	0.101	1168	14.7	6.5-	113.0	2.6	115.6	ł		1	:
Han-Stid 14C-A	170.0	0.011							513 CV -	( DC 0	. 14C - soil CD-
$*a^{14}C_m = \text{sample } m$	easured 14C a	ctivity; a <sup>14</sup> Cat	m = atmosphe	ric <sup>14</sup> C activity (c	alculated w	ith A <sup>14</sup> C publ	ished data and	an atmospheric		(mar	, u vg - 500 002
14C activity. CO./I	DIC frac. = so	il CO, / DIC	fractionation	factors; a <sup>14</sup> C <sub>DIC</sub> =	EDIC <sup>14</sup> C ac	stivity, IAEA	model = initia	I DIC 1*C activ	rity calcula	IED WIII UNI	
- Cacuviry, ~~ "						•					

TABLE 3. 14C Activity and <sup>13</sup>C Results of Speleothems and Seepage Waters\*

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The dcp is calculated using the measured <sup>14</sup>C activity of the deposit  $(a^{14}C_m)$  and the mean atmospheric <sup>14</sup>C activity of the period covering the calcite formation  $(a^{14}C_{atm}; Table 3)$ :

dcp = 
$$\left(1 - \frac{a^{14}C_m}{a^{14}C_{atm}}\right)$$
 100 % . (1)

Because we compare only groundwaters, no normalization (for a  $\delta^{13}$ C of -25%; Stuiver and Polach 1977; Mook 1980) was done (Wigley and Muller 1981).

The dilution factor (q) connected to the "hard water effect" and used by several authors (Thorpe, Otlet and Sweting 1980; Vogel 1983; Gewelt 1985; Bastin and Gewelt 1986; Fontes 1992) is

$$q = (100 - dcp) / 100.$$
 (2)

From  $a^{14}C_m$  and  $\delta^{13}C$  uncertainties (average uncertainty of ±0.7 pMC and ±0.1‰, respectively) and assuming an average error of ±0.7‰ for the  $a^{14}C_m$ , we estimated the total dcp error at ±1.5%.

Dead carbon calculation, as presented above, is actually an apparent proportion because it is deduced from direct atmospheric and carbonate or water <sup>14</sup>C activity measurements and does not take into account fractionation processes. The decrease of the seepage water <sup>14</sup>C activity due to limestone dissolution (the actual "dead carbon effect") should lie between the dissolved inorganic carbon (DIC) <sup>14</sup>C activity of the water before and after dissolution. Before dissolution, DIC activity is in equilibrium with the soil CO<sub>2</sub>; after dissolution, DIC activity is similar to the measured activity of water bicarbonates and of calcite, within uncertainties due to fractionation processes. Thus, it would be more rigorous to consider isotopic fractionations that occur at different stages (Fig. 1):

- 1. exchange between atmospheric and soil CO<sub>2</sub>;
- 2. CO<sub>2</sub> dissolution and bicarbonate formation;
- 3. limestone dissolution (dead carbon injection);
- 4. seepage;
- 5. CO<sub>2</sub> degassing and CaCO<sub>3</sub> precipitation at the time of seepage into the cave.

Based on this simplified course, the dead carbon arrives in the water during the third stage and it is necessary to know the processes during the two previous stages.

## Exchange between Atmospheric and Soil CO2

A global formula takes into account <sup>14</sup>C fractionation due to photosynthesis, decarboxylation and diffusion (Fontes 1992):

$$a^{14}C_{g} = a^{14}C_{atm}\left(\frac{1-2.3(\delta^{13}C_{atm}-\delta^{13}C_{g})}{1000}\right) pMC$$
, (3)

where "g" signifies soil CO<sub>2</sub> and "atm" atmospheric CO<sub>2</sub>. The latter is known  $(a^{14}C_{atm})$  is variable and  $\delta^{13}C_{atm} = -8.2\%$ , from Levin, Graul and Trivett (1995)), so we must estimate  $\delta^{13}C_g$ . Above the caves studied, vegetation consisting of oak trees and Graminaceae is typical of the C<sub>3</sub> type (Calvin-Benson photosynthetic pathway) which produces a  $\delta^{13}C_g$  between -20 and -25‰ (Fritz *et al.* 1978; Dörr and Münnich 1986, etc.). Direct measurements on soils above limestone yield values between -20.8 and -23.1‰ for Moulis (Pyrenees, France; Fleyfel 1979) and -22‰ for the Champagne area



Fig. 1. Schematic <sup>14</sup>C activity evolution from atmosphere toward cave deposit. Numbers in % represent the average relative <sup>14</sup>C activity variations due to fractionation processes (see text and Table 3). Note that the <sup>14</sup>C activity decrease due to soil/atmosphere exchange is compensated by the CO<sub>2</sub>/DIC fractionation within the uncertainty limits ( $\pm$  1.5%).

(France; Dever *et al.* 1982). We can reasonably consider a mean value of -22%. Results show that soil CO<sub>2</sub> activity is, on average, 3.6 pMC lower than atmospheric activity (Table 3). Karstic soils are generally very thin, and it can be assumed that there is no significant delay between atmosphere and soil CO<sub>2</sub> (Dever *et al.* 1982).

## CO<sub>2</sub> Dissolution and Bicarbonate Formation

Assuming equilibrium between soil  $CO_2$  and the DIC, the activity of the latter is given by (Saliège and Fontes 1984):

$$a^{14}C_{\text{DIC}} = a^{14}C_{\text{g}} + 0.23(\delta^{13}C_{\text{DIC}} - \delta^{13}C_{\text{g}}) \text{ pMC}$$
 (4)

Results show an average  ${}^{14}C_{DIC}$  enrichment of 2.6 pMC (Table 3).

The net isotopic balance of stages 1 and 2 is a decrease of about 1 pMC between atmosphere and DIC activities, which is similar to the uncertainty range  $\pm 1.5$  pMC. Thus, depending on whether we consider atmospheric activity or DIC activity, it is very likely that we will obtain similar values for the dead carbon proportion.

Several models have been developed in order to estimate the hard water effect (connected to the dead carbon effect by Equation (2)) by the calculation of correction for initial water activity: *e.g.*, Tamers's chemical model (1967), Pearson's isotopic model (1965), the IAEA isotopic model (Salem *et al.* 1980), Plummer's chemical and isotopic model (1977) and Fontes and Garnier's chemical and isotopic model (1977). We have applied the IAEA model, which corresponds to the Pearson model corrected by the  $CO_2/DIC$  fractionation factor  $\varepsilon g$ -b. This authorizes its use for open systems, which is more adapted to the karstic unsaturated zone:

$$a^{14}C_{\rm DIC} = (a^{14}C_{\rm g} - a^{14}C_{\rm c}) \cdot (\delta^{13}C_{\rm DIC} - \delta^{13}C_{\rm c}) / (\delta^{13}C_{\rm g} - \epsilon g - b - \delta^{13}C_{\rm c}) \, pMC \,, \tag{5}$$

where  $a^{14}C_c$  and  $\delta^{13}C_c$  relate to the limestone of the rock formation,

and  $\varepsilon g - b = -9483 T^{-1} + 23.89\%$ 

with T = temperature in K (Mook, Bommerson and Staverman 1974).

## **RESULTS AND DISCUSSION**

#### <sup>13</sup>C Results

Measured  $\delta^{13}$ C values (Table 3) are typically in the range of published results for speleothems and demonstrate that the main source of carbon from dissolved inorganic carbon comes from an equilibration with soil CO<sub>2</sub> (*i.e.*, with  $\delta^{13}C_g = -22\%$  and T = 15°C, the enrichment factor = -9\%, then  $\delta^{13}C_{DIC} = -13\%$ ; Mook, Bommerson and Staverman 1974; Schwarcz 1986; Dulinski and Rozanski 1990; Gascoyne 1992). Calcite values are slightly higher than water values. Average differences are 1.2% for Villars, 0.8% for La Faurie and 1.1% for Proumeyssac. For the cave temperatures (between 10.5 and 15°C), the enrichment factor between DIC and calcite is much lower than the observed difference (between 0.41 and 0.18‰; Mook 1980). However, these differences cannot be interpreted in terms of isotopic equilibrium because waters and calcite do not come from the same systems, and we know that variations from place to place in the same cave can be very great, as noticed in Lower Cave in England (variations up to 4.7‰ between modern deposits; Baker et al. 1996). Another possibility is seasonal variation: because calcite develops over several years, mostly in winter when the drip rate is high (Genty et al. 1996, 1997), and because all the waters studied were sampled in summer, it may be possible that the observed differences reflect seasonal variations in the isotopic content of seepage water. Similar variations have been observed in the seasonal growth laminae of stalagmites in Belgium, possibly due to the isotopic content variation of the seepage water (Genty, Quinif and Keppens 1995). The other possibility is that water is lighter than cal-

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cite because of a slight kinetic fractionation due to a fast  $CO_2$  degassing during CaCO<sub>3</sub> precipitation; however, if this is the case here, it would provoke a negligible <sup>14</sup>C fractionation (Hendy 1971).

In the Han-sur-Lesse cave, analyzed samples consist of two pairs of stalagmites and stalactites; each pair belongs to the same system. A <sup>13</sup>C enrichment of 0.9‰ and 1.7‰ is observed between stalactites and stalagmites for both stations, respectively. Differences up to 2.8‰ have been observed between stalactites and stalagmites of Lower Cave, England (Baker, Barnes and Smart 1996); evaporation could be one of the causes. Here, the  $\delta^{18}$ O results do not testify in favor of evaporation, at least for the Han-stt4/Han-stm4 couple where the difference is -0.4% (Table 3); then kinetic isotopic fractionation due to a fast CO<sub>2</sub> escape could explain these differences (Hendy 1971). In any case, differences here are small and will have no important consequences on <sup>14</sup>C fractionation (*i.e.*, if we assume a <sup>14</sup>C fractionation twice that of the <sup>13</sup>C fractionation, maximum <sup>14</sup>C enrichment will be 3.4%, which is within the <sup>14</sup>C activity measurement uncertainty).

#### <sup>14</sup>C Results

Calcite and DIC AMS <sup>14</sup>C contents are between 89.7 and 118.6 pMC (Table 3). These high values confirm the continued importance of bomb <sup>14</sup>C contamination. The dead carbon proportion is between 9.2% and 21.9% for the calcite deposits and between 3.6% and 21.9% for the DIC of seepage waters. From these results we can make two important inferences: 1) for the same sampled site, waters have a systematically lower dead carbon proportion than calcite; 2) for the same site and where we analyzed several calcite samples, the dead carbon proportion is homogeneous.

## <sup>14</sup>C Activity Comparison between Water (DIC) and Calcite

Villars cave samples show the lowest dcp difference between water and calcite (avg. = 1.1%), which is within the uncertainty limits ( $\pm$  1.5%). For the La Faurie and Proumeyssac caves, the difference is greater: 8.5% and 3.7%, respectively. Table 4 includes factors capable of having an influence on <sup>14</sup>C content in water. These show that:

- Neither the time elapsed between sampling and analysis, nor the time during which water was sampled, has any effect on dcp: *i.e.*, similar dcp is found for a low drip rate (La Faurie) and for a high drip rate (Proumeyssac);
- δ<sup>13</sup>C values do not show any possible equilibrium with cave air, whose composition must be similar to that of the atmosphere, but are characteristic of DIC that has been in equilibrium with soil CO<sub>2</sub> (according to the pCO<sub>2</sub>, which is close to that of the atmosphere; however, measurements will be necessary to verify this);
- pH values are typical of karstic waters; note, however, the higher pH (8) in the Proumeyssac cave.

Explanation of water-calcite <sup>14</sup>C activity differences must be found elsewhere. Seasonal variations in the <sup>14</sup>C content of soil CO<sub>2</sub> have been detected in Germany: it is lower in winter because of more intense organic matter decomposition and it is higher in summer because plant root respiration is the dominant factor; the winter/summer  $\Delta^{14}$ C variation varies from 50‰ under grassland to 100‰ under forest (Dörr and Münnich 1986). Water that feeds speleothems in temperate areas like Europe infiltrates mostly in winter between November and March (Genty *et al.* 1997); then it is likely that calcite deposits more closely reflect the winter season. Conversely, water that has been collected in August must reflect the summer season. Thus, the differences observed between the <sup>14</sup>C activity of water and calcite could be explained by a simple seasonal variation in the <sup>14</sup>C content of the soil above the cave; but more measurements are needed to confirm this hypothesis.

Sample and station numbers	dcp (%)	Type of bottle	ND*	TE†	MFR‡	pН	δ <sup>13</sup> C (‰ PDB)	Cave air pCO <sub>2</sub> (%)§
Vil-eau 14C-A - #1B	21.9	Plastic 250ml	10	<3	4.7	?	-11.8	0.16
Vil-eau 14C-B - #4	18.2	Plastic 250ml	49	<3	3.8	7.5	-9.7	0.16
Vil-eau 14C-C - #1B	20.0	Plastic 250ml	49	<4	6	7.5	-10.8	0.16
Fau-eau 14C-A - #2	3.6	Glass 150ml	47	<48	95	7.1	-11.7	0.08
Fau-eau 14C-B - #12	3.9	Glass 150ml	47	<48	100	7.1	-11.7	0.08
Prou-eau 14C-B - #1	5.5	Glass 150ml	47	<0.3	continuous water flow	8.0	-10.7	0.04
Prou-eau 14C-A - #2	5.5	Glass	47	<0.5	<2	8.0	-14.0	0.04

TABLE 4. Factors Capable of Influencing Content of <sup>14</sup>C in Water

\*ND = No. of days between sampling and analysis

†TE = Time elapsed during dripping sampling, hours

\$MFR = Mean flow rate, seconds/drip

§Measured January 1997

## Inter-Site<sup>14</sup>C Activity Comparison

For each site, and where at least several analyses have been made, it appears that the calcite dcp is homogenous (Fig. 2). Villars shows the highest values (avg. = 21.1%,  $\sigma = 0.8\%$ ; n = 3), followed by Han-sur-Lesse (avg. = 13.8%,  $\sigma = 0.8\%$ ; n = 4), La Faurie (12.2%; n = 1) and Proumeyssac (9.2%; n = 1). Comparison of these results with environmental conditions—vegetation type and density, soil thickness, geology and limestone formation thickness, pluviometry and temperature (Table 2)—leads to the following remarks:

- Rock formation thickness does not correlate with dcp; moreover, in the same cave a similar dcp was found for two samples taken at a 10 m altitude difference (Villars, #1B and #4);
- soil thicknesses, which are typical of karstic areas, also do not correlate with dcp;
- the lowest dcp (calcite and water) is found under grassland (La Faurie and Proumeyssac), whereas the highest is found under forest (Villars and Han-sur-Lesse);
- however, the Han-sur-Lesse site (Belgium), also covered by a continuous forest, shows a calcite dcp close to the dcp found under grassland in Southwestern France sites; note also that this site is about 3°C colder than the French ones;
- the difference observed between La Faurie and Villars, which are 60 km apart and have similar geology and climate, is possibly due to the forest cover of the Villars site.

The dcp is theoretically linked to the dissolution intensity: the more intense the dissolution, the higher the dcp. Factors that control carbonate dissolution are temperature and, most importantly, soil  $CO_2$ partial pressure (Dever *et al.* 1982; Dörr and Münnich 1986, *etc.*). Soil  $CO_2$  has two major sources: root respiration and microbial organic matter decomposition. Both sources will be higher in a forest soil than under a grassland. This can explain, for the most part, why dcp is higher under forest covers like the Villars and Han-sur-Lesse caves. It is possible that the 3°C lower temperature of the Belgian site yields a lower vegetation activity and thus a lower dissolution and dcp.



Fig. 2. Dead carbon proportion (dcp) vs.  $\delta^{13}$ C. Note that all samples from the same site are grouped together; Villars cave has the highest dcp, followed by Han-sur-Lesse cave, both under a forest cover. La Faurie and Proumeyssac, which are under grassland, show the lowest dcp.

But we must consider that other unquantified factors may possibly intervene. The possible mixing of fast and slow transit waters can change the theoretical ages that we have considered; only the hydrographic analysis of stalactite dripping flows in the different sites studied will inform us as to the possible age spectrum of the waters (work in progress). Another complicating factor can be the fracture density in the rock formation through which water infiltrates. During seepage, both water and gas infiltrate (two-phase seepage) as has been shown in a Southern France karstic area (Mangin 1975; Fleyfel 1979; Fleyfel and Bakalowicz 1980). Such a flow, combined with an intense fracture system, will favor dissolution-reprecipitation processes and thus secondary calcite deposits before reaching the cave itself (not to mention the higher possibility of isotope exchanges). This will have major consequences on speleothem isotope measurements, particularly in modern deposits. For example, a period of intense dissolution will have opposite effects on dcp:

- a decrease in the DIC <sup>14</sup>C activity, and consequently a higher dcp, if the dissolved carbonate is old limestone;
- an increase in the DIC <sup>14</sup>C activity, and consequently a lower dcp, if the dissolved carbonate is a secondary calcite deposit that was precipitated in the 1960s, when the atmospheric <sup>14</sup>C activity was very high.

The actual process probably occupies a midpoint between these two hypotheses: both old limestone with 0 pMC activity and secondary calcite deposits with a significant <sup>14</sup>C activity are dissolved during infiltration.

# Comparison Between Measured <sup>14</sup>C Activity and the IAEA Model Initial Activity Calculation

Water mixing models could be helpful in understanding dissolution processes. Because we do not have enough chemical data, we have only tested the corrected Pearson isotopic model, known as the IAEA model (Salem *et al.* 1980), on three sites: La Faurie, Villars and Proumeyssac (Table 3). Because the original Pearson model is designed for closed systems (Pearson 1965) and because the karstic unsaturated zone cannot be considered as a closed system (CO<sub>2</sub> is mostly dissolved in the presence of carbonates, at least in our sites where the soil thickness is only a few centimeters; Krajcar-Bronić *et al.* 1986, 1992; Ek, Hilaire-Marcel and Trudel 1981; Fontes 1992), it must be corrected by the CO<sub>2</sub>/DIC fractionation factor. Considering a soil CO<sub>2</sub>  $\delta^{13}$ C = -22‰ and a 0 pMC <sup>14</sup>C activity for the limestone formation, the average calculated DIC activities show quite good agreement (±15 pMC) with measured activities (Table 3). The average difference between the IAEA and the measured activities is -1.2 pMC ( $\sigma$  = 12.0), which confirms the validity of this model in different geological contexts (Fontes, 1985).

## **CONCLUSIONS**

- 1. Thanks to annually laminated speleothems, the dcp due to old limestone dissolution can be calculated within an uncertainty range of ±1.5%.
- 2. The dcp varies from 9.2% to 21.9% for calcite deposits and from 3.6% to 21.9% for DIC in water.
- 3. At the same sampling site, and where several measurements were made, the dcp is homogeneous.
- 4. Inter-site dcp variations are up to 11%; the average dcp in our sites is 15.5% ± 4.4, within the uncertainty range of former measurements and assumptions (15% ± 5; equivalent to a dilution factor of 0.85 ± 0.5).
- 5. For the same site, water has a lower dcp than calcite deposits (mean differences are between 1.1 and 8.5%); this could be because all the waters studied were sampled in summer, when soil CO<sub>2</sub> activity is higher, while speleothems mostly develop in winter and spring when soil CO<sub>2</sub> activity is lower; but more measurements are needed to confirm this hypothesis.
- 6. For a similar environment, the dcp is higher under forest cover (e.g., at the Dordogne sites, dcp = 21.1% under forest and dcp = 12.2% under grassland); this may be the consequence of more intense dissolution processes due to root respiration and microbial organic matter decomposition. However, the Belgian site, also under a forest cover, shows a dcp very close to Southwestern France sites that are under grassland; the 3°C lower temperature may be one of the causes. The possible mixing of rapid and slow (old) water flows must also be considered as playing a role in dcp results; only stalactite hydrographic analyses in each site will bring insight into the nature of the drainage.

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7. The IAEA isotopic water mixing model (Pearson model corrected for an open system) is in good agreement with measured activities.

If dcp is as varied as we observed from one site to another, and if those variations are due to vegetation and soil conditions, it is very likely that dcp changed at any given site during the past when vegetation altered because of human activities and/or climate changes. From our study, dcp can vary by 11%; this yields an age difference of 970 yr, which is not negligible for the Holocene or Late Glacial chronology. Thus it is highly recommended to calculate the dcp at the sample site first before doing any age calculation based on <sup>14</sup>C method.

If vegetation seems to be a factor that influences the dcp, it is likely that environmental factors that favor secondary calcite precipitation in the unsaturated zone, such as fracture density, also play an important role. The high measured activities could accordingly be explained by the dissolution of secondary calcite deposits that precipitated in the 1960s or 1970s, when the atmospheric activity was very high. Thus, the dcp can be tied both to vegetation and to the local potential of secondary calcite deposit. The possible existence of such a process greatly complicates the interpretation of the dcp; work in progress on a single laminated stalagmite will help shed light on the importance of dissolution-precipitation processes above a cave.

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# CHARACTERIZATION OF GROUNDWATER IN THE CARIRI (CEARÁ, BRAZIL) BY ENVIRONMENTAL ISOTOPES AND ELECTRIC CONDUCTIVITY

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ABSTRACT. The Cariri region is the largest sedimentary basin in the state of Ceará, Brazil. Located in the southern portion of the state, it comprises the Araripe Plateau and the Cariri Valley on its northern foot. The region's groundwaters are being heavily exploited. Using electric conductivity (EC) and <sup>18</sup>O, <sup>14</sup>C and <sup>3</sup>H data, we differentiate groundwaters from various origins. We identified three horizons of springs on the slope of the Plateau through their geologic environment and the EC of their waters. Groundwaters from wells in the Cariri Valley are classified according to the aquifers exploited as indicated by the drilling profiles. However, strong tectonic features and intense fracturing in the Valley produce a great many horizontal discontinuities, which result in a mixing of groundwaters from different aquifers. Mixing systems are described in terms of  $\delta^{18}O^{-14}C$  and EC<sup>-14</sup>C linear trends.

#### INTRODUCTION

The Cariri region is located in the southern portion of the state of Ceará in the semiarid northeast of Brazil. The area is a green "oasis" embedded in a barren landscape of mostly crystalline bedrock outcroppings. The region consists of a sedimentary plateau (the Araripe Plateau) with a mean elevation of 750 m and an area of ca. 7500 km<sup>2</sup>, and a sedimentary basin (the Cariri Valley), measuring ca. 3500 km<sup>2</sup>, on its northern foot.

Rich groundwater resources are the driving agents for the region's important economy, which is second only to that of the capital of the state. The resources provide an exclusive water source for cattle raising and domestic, industrial and agricultural water supplies and are being intensively exploited mainly for large-scale irrigation of sugar cane. However, the origin of the groundwater and its recharge areas are still largely unknown.

Our research in the Cariri was initiated because of popular complaints denouncing the environmental damage caused by deforestation at the top of the Araripe Plateau, as well as overexploitation of the Cariri Valley aquifers. The discharge from one of the major springs emerging from the cliffs of the Plateau has dropped during the period from 1920 to 1993 from *ca.* 1300 m<sup>3</sup> h<sup>-1</sup> to 380 m<sup>3</sup> h<sup>-1</sup> (Kemper *et al.* 1995). In years with poor rainfall the water table in the Valley lowered dramatically. A great number of private wells (with depths <20 m) fell dry and natural lakes disappeared without returning to their normal extension in the following years. Based on environmental isotope data (<sup>14</sup>C, <sup>18</sup>O, <sup>3</sup>H) and EC measurements, we have characterized the waters from the different aquifers to evaluate regional groundwater dynamics.

## **DESCRIPTION OF THE AREA**

The area of this study comprises mainly the townships of Crato, Juazeiro do Norte, Barbalha and Missão Velha (Fig. 1). In this part of the Cariri, mean annual rainfall is *ca*. 1000 mm, surpassing that of many coastal areas of the state. Differing from the rest of Ceará, where the southerly shifting of the Zone of Intertropical Convergence causes the single rainy period around April (lasting three to four months), the Cariri is favored with two distinct rainfall regimens that produce a two-humped distribution of precipitation: in addition to the main peak in April, a precursing one appears in January. This is due to cyclonic events penetrating from the south. Because of this, the growth period of vegetation is greatly increased as compared to the central and northern parts of Ceará.

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Structurally, the Cariri region is characterized by pronounced tectonic features. Mont'Alverne *et al.* (1995) describe successive grabens and horsts along sections AB and CD in Figure 1A. Chignone *et al.* (1986) individualized seven structural blocks in the Cariri Basin, the most important of which are: 1) the block of Crato/Corredores, with arched grabens and horsts; 2) the graben of the Serra das Cacimbas; and in between these, 3) the regional, central horst of Mauriti.

A drilling (P1 in Fig. 1B) by Petrobras in the township of Araripina/Pernambuco, on the Plateau, revealed the following sequence of formations (thicknesses are in parentheses): Exu (228 m), Arajara (165 m), Santana (112 m), Rio da Batateira (198 m), Abaiara (124 m), Missão Velha (190 m), Brejo Santo (431 m) and Mauriti (41 m). Exu, Arajara, Santana and Rio da Batateira are all from the Middle Cretaceous Period, Abaiara is from the Cretaceous, Missão Velha and Brejo Santo from the Upper Jurassic and, finally, Mauriti, which is in contact with the crystalline basement, is from the Silurian/Devonian Period. The first three formations constitute the Araripe Plateau and are absent in the Cariri Valley. Arajara, Santana and Brejo Santo formations are aquitards and the others are aquifers.

A drilling by the Departamento Nacional de Produção Mineral (DNPM) in the township of Bodocó/ Pernambuco, *ca.* 15 km southwest of the Petrobras, showed that the formations Abaiara, Missão Velha and Mauriti are absent at this location. Sequence and thicknesses are as follows: Exu (190 m), Arajara (40 m), Santana (242 m), Rio da Batateira (59 m), Brejo Santo (380 m) and Mauriti (32 m). The piezometric level for the Rio da Batateira Formation was 370 m by this drilling.

Due to sandy soils on top of the Plateau and, possibly, open fractures in the Exu Formation, infiltration occurs very quickly there. A surface drainage system is only faintly developed. Even several hours after heavy rainfall, no accumulation of surface water is found. There are no producing deep wells on the Plateau, with the exception of one in the Fazenda Janaguba, near the edge of the Plateau, that taps the saturated layer (possibly 60 m thick) at the contact of the formations Exu and Arajara. However, there are some localized perched aquifers, such as in a settlement called "Cacimbas". (The name refers to a type of shallow, dug well found in the area.)

The Exu/Arajara and Arajara/Santana contacts constitute discontinuities (from high to low) of hydraulic conductivity and define the horizons of some hundreds of springs that emerge at roughly half of the height of the Plateau above the Basin. They appear mainly in gorges and other locations affected by erosion where resistance to water flow is reduced. Their total discharge is roughly 50 m<sup>3</sup> min<sup>-1</sup> (Gaspary 1967). The allocation and use of the waters from some of these springs have been legally established for over a century.

The exploitation of aquifers in the Cariri Basin is intense. The depths of wells studied range from 32 to 240 m, with pumping rates reaching  $300 \text{ m}^3 \text{ h}^{-1}$ ; their water generally derives from contributions from various aquifers. Wells are often arranged in batteries, specially along the alluvial zones of the rivers Rio da Batateira (in Crato), Riacho dos Macacos (in Juazeiro do Norte), Rio Salamanca (in Barbalha) and Riacho dos Porcos (in Milagres). Due to excessive pumping of wells, in some of these rivers during the dry season, the natural runoff has not been replaced by returning waste water from households and factories.



Fig. 1. A) The area of study; B) Geological sketch; C) Stratigraphic of the Araripe Plateau and Cariri Valley.

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## METHODS

<sup>14</sup>C and <sup>18</sup>O measurements were made on groundwater, and <sup>3</sup>H and <sup>18</sup>O were analyzed in water from springs. The discharge from two springs was monitored for *ca*. 3 yr using NaCl dilution after instantaneous injection. <sup>14</sup>C and <sup>3</sup>H were measured in our (Physics Department) laboratory and <sup>18</sup>O was measured by Hydroisotop (Schweitenkirchen, Germany). Electric conductivity (EC) was measured in the field. Geographic coordinates were assigned through GPS.

<sup>14</sup>C values were obtained by gas-proportional counting of acetylene (864 ml at 760 Torr, corresponding to *ca.* 1 g of carbon). The standard used was HOxI. An error of  $\pm 1 \sigma$  applies to the counting statistics of background, samples and modern standard. Some of the <sup>14</sup>C data were published previously (Santiago *et al.* 1994). Tritium was measured in our laboratory with a Packard Tri-Carb<sup>®</sup> 2000 CA/LL liquid scintillation spectrometer after electrolytic enrichment by a factor of 40, starting with a sample volume of 500 ml, and using Insta-Gel<sup>®</sup> as scintillator. The detection limit was 0.3 TU.

## RESULTS

In Table 1, 19 springs are listed together with their locations (township, coordinates), EC,  $\delta^{18}O$  (‰, SMOW) and the approximate discharge (Q). The last column indicates the environment (formation or interface of formations) as assigned by us using a spring's elevation together with the stratigraphic column. Three groups can be distinguished.

- 1. The first set comprises the first 12 springs (from Batateira through Santa Rita), listed in Table 1 in order of decreasing discharge. They originate from the Exu/Arajara contact. Their waters are very weakly mineralized, with EC  $\leq 30 \,\mu$ S cm<sup>-1</sup>.
- The second set is made up of three minor springs from the Arajara Formation (Angélica, Olho d'água and Recanto). Their discharge is much smaller and diffuse and has therefore not been measured. Mineralization is significantly higher (EC from ca. 80 μS cm<sup>-1</sup> to 130 μS cm<sup>-1</sup>) than in the first group.
- 3. The third set comprises the springs Solzinho and Macauba, which we attribute to the contact of the formations Arajara and Santana. Their EC is  $234 \,\mu\text{S cm}^{-1}$  and  $266 \,\mu\text{S cm}^{-1}$ , which is two to three times that of the second set.

We added two springs to Table 1 that do not originate from the Plateau but are located in the Cariri Valley: Rosário and Ciciaca. Rosário is located on the cliff of the horst of Mauriti, and Ciciaca where the Mauriti Formation meets the crystalline basement. We measured  $\delta^{18}$ O for some springs: Pendência in Missão Velha yielded -3.4‰; Boca da Mata and Solzinho in Jardim, -2.9‰ and -3.5‰, respectively. The two springs in the Valley, Rosário and Ciciaca, measured -3.4‰ and -2.9‰.

Results from wells in the Cariri Valley show a wide range of values reflecting the opulence of tectonic features of the region. We group them according to the main aquifers exploited as indicated by the drilling profiles. Tables 2–5 represent, respectively, wells in the aquifers Rio da Batateira, Rio da Batateira in combination with Missão Velha, Missão Velha, and, finally, Mauriti. However, we must emphasize that we deal with production wells where all water entries are utilized, irrespective of their origin. Thus, we cannot expect a clear-cut separation of the different groups. Nevertheless, EC is a fairly good parameter for characterizing the water produced by each group:

 The Rio da Batateira wells have EC below 200 μS cm<sup>-1</sup> if we eliminate a subgroup of five wells (at the end of Table 2) that are located in areas where fluvial deposits along riverbeds are directly connected with the Rio da Batateira aquifer;

- The Rio da Batateira/Missão Velha wells produce waters with EC between 200 μS cm<sup>-1</sup> and 300 μS cm<sup>-1</sup> (excluding P6 and P10, see comments below) (Table 3);
- The Missão Velha wells have from  $300 \,\mu\text{S cm}^{-1}$  to  $580 \,\mu\text{S cm}^{-1}$  (Table 4);
- The Mauriti wells have from  $600 \,\mu\text{S cm}^{-1}$  to  $980 \,\mu\text{S cm}^{-1}$  (Table 5).

	Town-			EC	Q†	δ <sup>18</sup> Ο	
Locality	ship*	Lat. S	Long. W	(µS cm <sup>-1</sup> )	(m <sup>3</sup> h <sup>-1</sup> )	(‰)	Origin
Springs on the S	Slope of the	e Plateau					····
Batateira	Ct	7°15′35″	39°28′17″	28	376		Exu/Arajara
Pendência	MV	7°24′35″	39°12′46″	25	352	-3.4	Exu/Arajara
Bica do Farias	Bb	7°19′50″	39°24′45″	27	348	011	Exu/Arajara
Sítio Cocos	Bb	7°22′36″	39°17′14″	19	182		Exu/Arajara
Sítio Saco	Ро	7°29′38″	39°09′45″	20	182		Exu/Arajara
Bom Jesus	Bb	7°22′39″	39°17'19"	30	180		Exu/Arajara
Sozinho	Ct	7°19′15″	39°24′48″	17	154		Exu/Arajara
Coqueiro	Ct	7°17′02″	39°25′57″	23	140		Exu/Arajara
Boca da Mata	Jd	7°33'20″	39°16'21"	26	132	-29	Exu/Arajara
Camelo	Bb	7°22'23″	39°20'33"	15	120	2.7	Exu/Arajara
Água Grande	Ct	7°17′15″	39°24′58″	25	113		Exu/Arajara
Santa Rita	Bb	7°21′21″	39°18'48"	15	102		Exu/Anajala Exu/Arajara
Angélica	Ct	7°12'46″	39°26'33"	69	102		Arojoro
Olho d'água	BS	7°28'05"	39°04'49"	77			Arajara
Recanto	Ex	7°25'33″	39°50'25"	118			Arajara
Solzinho	Jd	7°34'43"	39°16′17″	234		25	Anajara Anajara/Samtana
Macauba	MV	7°12'46″	39°39'06"	266		-3.5	Arajara/Santana
Springs in the C	ariri Valle	y 12 40	57 57 00	200			Arajara/Santana
Rosário	Mi	7°18'20″	39°58′05″	118		-3.4	Mauriti
Ciciaca	NO	7°06′11″	39°38'22"	339		-2.9	Mauriti/basement

TABLE 1. The Major Springs in the Cariri Basin, their Coordinates, EC and Discharge (O)

\*Bb = Barbalha; BS = Brejo Santo; Ct = Crato; Ex = Exu; Jd = Jardim; Mi = Milagres; MV = Missão Velha; NO = Nova Olinda; Po = Porteiras.

†After Mont'Alverne et al. (1995)

## DISCUSSION

Based on the values given in Tables 1–5 and previous isotope measurements from springs (Santiago *et al.* 1992) and rain (Frischkorn *et al.* 1990) we designed the following conceptual model for groundwaters on the Araripe Plateau and in the Cariri Valley.

Infiltrated rainwater from the top of the Araripe Plateau percolates the Exu sandstone and feeds a group of *ca*. 300 springs. As shown in Table 1, and in a previous publication (Santiago *et al.* 1988), the springs with the highest discharge are situated at an elevation of *ca*. 700 m and have EC <30  $\mu$ S cm<sup>-1</sup>. According to its drilling profile, a well in Fazenda Janaguba (elevation 730 m, depth 50 m) produces water from the contact of the formations Exu and Arajara. Its water has an EC of 32  $\mu$ S cm<sup>-1</sup> and a tritium concentration of 1.3 TU, which is comparable to values for the first group of springs, which lie between 0.3 TU and 1.3 TU (Santiago *et al.* 1990). Thus, these values verify that springs with EC <30  $\mu$ S cm<sup>-1</sup> discharge from the Exu/Arajara contact.

 $\delta^{18}$ O for one of these springs (Batateira in Crato) was monitored for 10 months and was found to follow seasonal rainfall variations of <sup>18</sup>O with a lag of *ca*. 5 months (Frischkorn *et al.* 1990). The discharge from two springs in Barbalha (Studart *et al.* 1992) was monitored for >2 yr. In this case, we

	*	Town-	P			δ <sup>18</sup> Ο	EC	<sup>14</sup> C
No.	Locality	ship†	(m)	Lat. S	Long. W	(‰)	(µS cm <sup>-1</sup> )	(pMC)
P5	Rch. Macacos 2	Jz	150	7°13′37″	39°18'26"	-3.1	168	74.6 ± 0.9
P26	Coni. Mirandão	Ct	102	7°14′11″	39°23′51″	-3.0	198	91.4 ± 0.8
P27	Lagoa Seca 10	Jz	91	7°14′51″	39°19′19″	-3.3	98	$80.8 \pm 0.5$
P28	Lagoa Seca 11	Jz	115	7°14′56″	39°19′22″	-3.3	186	66.6 ± 0.5
P29	Lagoa Seca 16	Jz	129	7°14′34″	39°18′51″	-3.0	182	88.7 ± 0.6
P34	Rch. Macacos 2a	Jz	32	7°13′38″	39°18′22″	-3.2	154	124.8 ± 0.9
P40	Lagoa Seca 17	Jz	180	7°14′23″	39°18'04″	-3.1	162	80.5 ± 0.6
P47	Cafundó 5	Ct	110	7°14′26″	39°24′20″	-3.0	183	98.8 ± 0.8
P54	SENAI	Ct	95	7°13′34″	39°23'49″	-2.9	199	
P63	Lagoa Seca 15	Jz	130	7°15′43″	39°19′20″	-3.1	157	
P67	Floresta	Ct	129	7°14′29″	39°24′59″	-3.1	113	
P69	São Raimundo	Ct	100	7°13′47″	39°25′31″	-2.8	169	
	Novo 2							
P70	Café da Linha	Ab	60	7°17′54″	39°02′05″	-2.9	79	
P77	Rosário	Mi	50	7°18′29″	38°57′54″	-3.5	98	
P79	Sizani	Ct	120	7°13′20″	39°25′20″	-3.1	174	
P80	Recanto 1	Ct	130	7°14′37″	39°24′56″	-3.1	134	
P15	São Raimundo 2	Ct	126	7°13′47″	39°25′31″		219	111.5 ± 1.1
P20	St. Monte Alegre	Ct	60	7°12′25″	39°24′37″		497	$121.4 \pm 1.1$
P22	V. Padre Cícero	Ct	98	7°13′08″	39°21′32″		88	$108.3 \pm 0.8$
P41	Batateira - lav.	Ct	32	7°13′25″	39°25′31″	-3.2	312	115.5 ± 1.1
P55	São Raimundo N1	Ct	130	7°13′49″	39°25′37″	-3.1	144	$104.2 \pm 1.1$

TABLE 2. Wells that Exploit the Rio da Batateira Aquifer\*

\*For Tables 2–5: No.= well number; P (m) = well depth;  $\delta^{18}O = \delta^{18}O$  (SMOW); EC = electric conductivity; pMC = percentage of modern carbon.

†Ab = Abaiara; Ct = Crato; Jz = Juazeiro do Norte; Mi = Milagres.

		Town-	Р			δ <sup>18</sup> Ο	EC	<sup>14</sup> C
No.	Locality	ship*	(m)	Lat. S	Long. W	(‰)	(µS cm <sup>-1</sup> )	(pMC)
P4	Lagoa Seca, 9	Jz	119	7°14'27″	39°19'22″	-3.3	291	64.2 ± 0.7
P6	Rch. Macacos 8	Jz	191	7°12′36″	39°18′08″	-3.9	522	$35.4 \pm 0.6$
P10	Poco 2	MV	86	7°15′32″	39°17′46″	-3.6	375	$30.3 \pm 0.4$
P19	Sítio S. Pedro	Bb	78	7°17′50″	39°17′12″	-3.2	226	99.2 ± 0.5
P33	Barro Branco	Bb	76	7°18′50″	39°15′22″	-3.4	288	$88.0 \pm 0.8$
P35	Rch. Macacos 3	Jz	140	7°13′35″	39°18′12″	-3.2	243	92.0 ± 0.7
P36	Rch. Macacos 4	Jz	140	7°13′25″	39°18′13″	-3.4	291	$78.2 \pm 0.5$
P38	Rch. Macacos 7	Jz	200	7°12′46″	39°18′20″	-3.2	281	75.1 ± 0.7
P39	Lagoa Seca 14	Jz	82	7°14′38″	39°19′03″	-3.5	299	61.0 ± 0.5
P46	Vila Alta 2	Ct	107	7°13′24″	39°24′43″	-3.2	232	93.7 ± 1.1
P58	Rch. Macacos 1	Jz	120	7°13′48″	39°18′24″	-3.4	258	77.5 ± 0.7
P71	Lagoa Seca 12	Jz	126	7°15′08″	39°19′21″	-3.4	253	72.4 ± 1.1
P72	Lagoa Seca 13	Jz	102	7°15′20″	39°19′23″	-3.5	213	$79.1 \pm 0.7$

TABLE 3. Wells that Exploit the Rio da Batateira and Missão Velha Aquifers

\*Bb = Barbalha; Ct = Crato; Jz = Juazeiro do Norte; MV = Missão Velha.

found a delay of 6–8 months with relation to monthly rainfall amounts. These results indicate a surprisingly quick hydraulic response, certainly due to percolation in fractures of the Exu sandstone. As "modern" rainfall has 3.2 TU, the 0.3–1.3 TU for springs are, according to a modified exponential model for the horizontal flow at the Exu/Arajara contact, compatible with times of transit between 4 and 26 yr (Silva *et al.* 1992).

No.	Locality	Town- ship*	<i>P</i> (m)	Lat. S	Long. W	δ <sup>18</sup> Ο (‰)	EC $(\mu S \text{ cm}^{-1})$	<sup>14</sup> C
P9 P31 P37 P48 P50 P53 P59 P68	Alto da Alegria Usina, prof. 200 Rch. Macacos 5 Abaiara - lav. CAGECE 7 Bela Vista Rch. Macacos 6 Sítio S. Paulo	Bb Bb Jz Ab Mi Bb Jz Bb	113 200 160 130 118 88 200 144	7°18'27" 7°18'08" 7°13'14" 7°21'10" 7°18'54" 7°19'40" 7°13'02" 7°13'02"	39°08'12" 39°14'09" 39°18'15" 39°02'47" 38°56'20" 39°17'46" 39°18'17"	-3.2 -3.2 -3.6 -3.8 -5.1 -4.3 -3.6	349 363 380 417 569 456 438	$93.2 \pm 0.7 \\ 84.5 \pm 0.7 \\ 48.9 \pm 0.4 \\ 44.9 \pm 0.6 \\ \\ 28.1 \pm 0.5 \\ 54.2 \pm 1.1 \\ \end{array}$
* A h _ A	hair Di Di t	-		, 100/	39 17 49	-3.0	332	

TABLE 4. Wells that Exploit the Missão Velha Aquifer

\*Ab = Abaiara, Bb = Barbalha, Jz = Juazeiro do Norte, Mi = Milagres.

TABLE 5. Wells that Exploit the Mauriti Aquifer

No.	Locality	Town- shin*	P (m)	Let C	¥	δ <sup>18</sup> Ο	EC	<sup>14</sup> C
D1			(11)	Lat. S	Long. W	(‰)	(µS cm <sup>-1</sup> )	(pMC)
	K. M. Ferreira	NO	80	7°05′21″	39°41′32″	-36	967	
P2	Poço 2	NO	128	7°06′32″	39°41′02″	-3.4	750	
P3	Baixio 3	NO	130	7°06'58"	20%41/10%	-3.4	152	$62.9 \pm 0.6$
P42	SESI	Ct	63	7 00 38	39 41 19	-3.8	976	3.6 ± 0.4
P44	Hn S Vicente	Dh	150	7 13 37	39°23'46"	-3.1	639	99.1 ± 0.7
P73	Abaiara 1	D0	150	/*18'39"	39°18′03″	-2.9	602	$90.8 \pm 0.8$
175 D74	Abalara 1	Ab	130	7°21′20″	39°02′40″	-3.5	776	$50.0 \pm 0.0$
F/4	Jacu	NO	127	7°05′42″	39°41′10″	-35	966	$39.7 \pm 0.7$
P76	Pedras Cariri	NO	50	7°05'43"	30°40'25"	-5.5	000	
P78	Vila Esperanca	Ma	80	7976122"	39 40 33	-3.0	631	
*Ah = AI	haiara: Ph - Dashalla			/ 20 33	38-57/14"	-2.8	670	

\*Ab = Abaiara; Bb = Barbalha; Ct = Crato; Ma = Mauriti; NO = Nova Olinda.

A second group of springs that emerges from the slope of the Plateau, characterized by EC between  $69\,\mu\text{S}$  cm<sup>-1</sup> and  $118\,\mu\text{S}$  cm<sup>-1</sup>, comprises mostly discharges of groundwater from the Arajara Formation that are not well concentrated but resemble seepage areas. Their discharge is difficult to evaluate. A well at the Colégio Agrícola (agricultural school) in Crato, at an elevation of 630 m and a depth of 45 m, produces, following the drilling profile, water from the Arajara Formation. EC of this well is  $71\,\mu\text{S}$  cm<sup>-1</sup> and tritium concentration is 0.9 TU, which confirms the attribution of this second group of springs to the Arajara Formation.

At still lower elevation, on the slope of the Plateau, there is another horizon of springs at the Arajara/ Santana contact with higher mineralization (with EC *ca.* 250  $\mu$ S cm<sup>-1</sup>). Their discharge is much smaller than that of the first group and normally diffuse. Solzinho Spring, which belongs to this group, shows a  $\delta^{18}$ O of -3.5%, which is the lowest value measured for springs. The scattering of  $\delta^{18}$ O between -2.9% and -3.5% demonstrates that, even at an elevation of *ca.* 200 m below the Plateau, the amount effect that marks rainfall is still perceptible without being blurred much by mixing. Even though, due to a very low water table at *ca.* 100 m deep, access to groundwater is difficult on the Plateau, some isolated clayey lenses produce perched aquifers that are being exploited in some places through dug wells. We determined EC for two of them and found 169  $\mu$ S cm<sup>-1</sup> (at Semião) and 526  $\mu$ S cm<sup>-1</sup> (at Romoaldo). The fact that these conductivities are considerably higher than those of the springs far below is still another indication of a vertical flow in fractures in the Exu Aquifer.

Tectonic fracturing is very intense in the Cariri Valley. This causes a varying sequence of formations in the sedimentary basin (Fig. 1C) and produces local mixtures of waters that are difficult to interpret. In our discussion, we adopt the new stratigraphic column proposed by DNPM (Mont'Alverne *et al.* 

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1995) in our assignments of isotopic characteristics to hydrogeologic structures. This column distinguishes, in the area of interest of the Cariri Valley, the formations, from top to bottom, Rio da Batateira (aquifer), Abaiara (aquitard), Missão Velha (aquifer), Brejo Santo (aquitard) and Mauriti (aquifer).

Phreatic waters from the Rio da Batateira Formation (Table 2) are weakly mineralized (EC <200  $\mu$ S cm<sup>-1</sup>). Figures 2A and 3A show pMC between 67 and 125 and  $\delta^{18}$ O from -2.8‰ to -3.5‰, with a mean of -3.1‰, which corresponds roughly to the mean value for rainfall in the area. The scattering of  $\delta^{18}$ O around the mean reflects seasonal and annual fluctuation in rainfall (amount effect). It is noteworthy that wells in zones of fluvial deposits exhibit special features: EC increases with pMC in a very good correlation (EC = 22.7 pMC - 2295) with a correlation coefficient of R = 0.94, different from the "traditional" decrease of EC with increasing pMC as shown by the other groups of wells. This behavior can be explained as a mixing line of very young, yet highly mineralized water from the fluvial deposits with young recharge from the unconfined Rio da Batateira aquifer.

Waters from wells that simultaneously exploit the aquifers Rio Batateira and Missão Velha (Table 3) are characterized by EC from  $200 \,\mu\text{S} \text{ cm}^{-1}$  to  $300 \,\mu\text{S} \text{ cm}^{-1}$  (with the exceptions of wells P6 and P10; see below). They comprise pMC from 30 to 99 and  $\delta^{18}$ O from -3.2% to -3.9%. The following correlations were obtained (Figs. 2B and 3B):

$$EC = -3.21 \text{ pMC} + 524 (R = -0.83)$$
 and (1)

(1)

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$$\delta^{18}O\% = 0.008 \text{ pMC} - 3.96 (R = 0.81)$$
 (2)

These correlations describe mixtures of young and weakly mineralized waters from the Rio da Batateira with a older and more mineralized water from the Missão Velha Formation. Wells P6 and P10 occupy an extreme position in this group. Even though their drilling profiles integrate them into the Rio da Batateira/Missão Velha wells, their  $\delta^{18}$ O and pMC values are the lowest of the group; their conductivities are the highest. As a result, these two wells seem to be "strangers" in an otherwise rather homogeneous group. In fact, mathematical simulation using MODFLOW of the dynamic levels for the battery of wells they are part of, and also the evaluation of pumping tests (Mendonça 1995), confirm that at least P6 is situated exactly upon a leak that, bridging the Abaiara aquitard, directly connects the aquifers Rio da Batateira and Missão Velha. Thus, for wells P6 and P10, pMC of 35.4 and 30.3 (with uncorrected ages of 7840 and 9600 yr, respectively) and  $\delta^{18}$ O levels of -3.9%o and -3.6%o may be taken as characteristic for this old component of the Missão Velha aquifer.

Wells that produce water from the Missão Velha aquifer (Table 4) are represented in Figures 2C and 3C. EC ranges from  $330 \,\mu\text{S cm}^{-1}$  to  $570 \,\mu\text{S cm}^{-1}$ , pMC from 93.2 (corresponding to an uncorrected age of 564 yr) to 28.1 (10,185 yr), and  $\delta^{18}$ O from -3.0% to -5.1%. The correlations obtained are

$$EC = -1.50 \text{ pMC} + 489 (R = -0.87)$$
 and (3)

$$\delta^{18}O(\%) = 0.016 \text{ pMC} - 4.54 (R = 0.94).$$
 (4)

Again, we understand them as mixing of a young component, originating from recharge by rainfall, and an ascending paleo-component. It is noteworthy that P31, P37, P48 and P59 were flowing wells at the time of drilling; now levels are some meters below surface.

The measurement -5.1% for  $\delta^{18}$ O of P50 is the lowest value for all the wells. It clearly indicates a strong contribution of paleo-waters dating back to a colder and more humid climate than at present in the northeast of Brazil. In previous research (Frichkorn *et al.* 1984; Frischkorn and Santiago 1992; Stute *et al.* 1995) on the deep aquifers in Piaui State we could prove that, at *ca.* 10,000 BP, temperature rose by roughly 5°C in the region, causing  $\delta^{18}$ O to rise from *ca.* -6‰ to present day -3‰. Thus, the same paleoclimatic effect should be found in the Cariri. The fact that no values <-5.1‰ have



Fig. 2. Electrical conductivity as a function of pMC for wells in the Cariri Valley. The exploited aquifers are listed below each graph.

been found is an indication of mixing with components younger than *ca.* 10,000 yr (unfortunately, we do not have <sup>14</sup>C for P50 as it is installed with an air-lift pump). Waters from the Mauriti Formation (Table 5) exhibit EC rates from 600 to  $980 \,\mu\text{S cm}^{-1}$ , pMC from 99.1 to 3.6 (corresponding to uncorrected ages from modern to 26,800 yr) and  $\delta^{18}$ O from -2.8% to -3.8% with mixing lines:

$$EC = -3.83 \text{ pMC} + 996 (R = -0.98)$$
 and (5)

$$\delta^{18}\text{O} = 0.009 \text{ pMC} - 3.89 (R = 0.92).$$
(6)

P42, with EC =  $639 \,\mu\text{S cm}^{-1}$ , pMC = 99.1 and  $\delta^{18}\text{O} = -3.1\%$ , can stand for the young contribution, originating from infiltration in the recharge area of the Mauriti aquifer. P3, with EC = 976  $\mu$ S cm<sup>-1</sup>, pMC = 3.6 and  $\delta^{18}\text{O} = -3.8\%$ , which exploits the Mauriti aquifer where confined by the aquitard Brejo Santo, may represent the aged component.



Fig. 3.  $\delta^{18}O$  (‰) as a function of pMC for wells in the Cariri Valley. The exploited aquifers are listed below each graph.

Summing up our results, we conclude that the predominant presence of young waters in all aquifers of the Cariri Valley makes them suitable for sustainable exploitation. However, at the same time, our findings reflect a high vulnerability to contamination of these aquifers due to easy infiltration in the sedimentary basin. This is of special importance because of the intense large-scale agricultural activity, mostly from sugar cane plantations, in the basin.

On the other hand, the Cariri Basin constitutes a sedimentary lens engraved in the crystalline basement, with aquifers outcropping on its border and submerging, in the area of this study from north to south, confined by aquitards. Accordingly, one expects the existence of a regional flow pattern for deep groundwaters leading from the margin to the center of the basin. In fact, we detected the presence of a paleo-component of >10,000 yr, marked by a lower atmospheric temperature than at present (as may be seen from the  $\delta^{18}$ O values of the paleowaters). This component may have a favorable influence on the exploitation of Cariri Valley aquifers as it can smooth down the influence of drought periods that affect the region.

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# *PENGUIN*, A MACINTOSH APPLICATION FOR ENTRY AND PRESENTATION OF RADIOCARBON-DATED SAMPLES

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ABSTRACT. Penguin is a Macintosh computer application that facilitates the use of CALIB 3.03, the <sup>14</sup>C age calibration program by Stuiver and Reimer (1993). Penguin offers an easy user interface based on the well-known Macintosh standard multiwindow environment to create and edit the CALIB 3.03 calibration files and to export data in text format. Penguin and CALIB interact at the file level, *i.e.*, Penguin is capable of reading and writing files in CALIB formats. Files containing the data are created in the Penguin environment and then saved on disk in the Penguin format. Penguin allows multiple editing of the calibration parameters and recalibration of the list of samples without the need to insert any modifications manually throughout the list. Penguin can also be used to read already calibrated files in order to extract the "cal" ages and display them in a spreadsheet-like window.

## INTRODUCTION

*Penguin* is a Macintosh computer application that facilitates the use of CALIB 3.03, the well-known  $^{14}C$  age calibration program by Stuiver and Reimer (1993).<sup>2</sup> The "*Penguin* project" emerged from our need for flexibility in managing data sets of  $^{14}C$ -dated samples (The name was suggested by the prettiest subject we were dealing with). In particular, we frequently need to update and calibrate sets of  $^{14}C$  dates from marine organisms (Baroni and Orombelli 1991) or from organisms that lived or fed in the sea, such as penguins and seals (Baroni *et al.* 1991; Baroni 1994; Baroni and Orombelli 1994). Nevertheless, the program is also useful for managing and editing sets of calibrated dates of other origin.

*Penguin* is at an early stage of development and is currently used at the Earth Science Department of the University of Pisa (Italy). Its currently implemented capabilities reflect the needs of the researchers who deal with <sup>14</sup>C dates. Features are added or modified each time a new need arises from our work. This means that the look and the functionality of *Penguin* may change in future releases, particularly if users assist us by supplying observations and suggestions for adding capabilities and/or modifying existing ones. Furthermore, some tools for graphical processing are currently being studied and could be added shortly.

Penguin is free software and is available from glsun2.gl.rhbnc.ac.uk via anonymous FTP, in the directory /pub/mac/apps.

## **PENGUIN CALIBRATION UTILITIES**

As is well known, the <sup>14</sup>C dates from remains of organisms that lived or fed in the sea are affected by an offset known as the "reservoir effect", induced by the depletion of <sup>14</sup>C in the ocean. This depletion is related to regional variations in oceanic and atmospheric circulation and its magnitude has also varied through time (Broecker and Olson 1961; Broecker, Peng and Engh 1980; Östlund and Stuiver 1980; Stuiver and Östlund 1980; Gordon and Harkness 1992). In the Antarctic Ocean, the reservoir effect is particularly elevated, owing to the dilution of circumantarctic water with glacial meltwater and by the upwelling of deep and old oceanic water (Harkness 1979; Omoto 1983;

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Stuiver, Pearson and Braziunas 1986). Thus, the apparent ages yielded by Antarctic samples are anomalously old as a consequence of the very low level of <sup>14</sup>C concentration in Antarctic water; the error is estimated to be >1000 yr and varies with different organisms and materials (Harkness 1979; Omoto 1983; Stuiver *et al.* 1981; Stuiver, Pearson and Braziunas 1986; Whitehouse, Chinn and Höfle 1988, 1989; Björck *et al.* 1991; Gordon and Harkness 1992; Berkman and Forman 1996). In order to compare the <sup>14</sup>C dates obtained from Antarctic samples with <sup>14</sup>C ages derived elsewhere, the <sup>14</sup>C ages need to be corrected for the reservoir effect and calibrated as accurately as possible.

According to Stuiver, Pearson and Braziunas (1986) and Stuiver and Reimer (1993), the calibration procedure for marine-derived organisms requires the computation of a parameter,  $\Delta R$ , that is the constant difference in reservoir age of a regional part of the ocean and the world ocean.  $\Delta R$  values can be determined if samples of known historical age are available (actually, only samples from organisms that died before the era of nuclear tests are suitable for this purpose). Such samples mainly derive from penguins and seals killed at the beginning of the century during the historical Antarctic expeditions (Stuiver *et al.* 1981; Mabin 1985, 1986; Orombelli 1988; Whitehouse, Chinn and Höfle 1988, 1989; Björk *et al.* 1991). Recently, dates from shells of known age have been supplied as well (Berkman and Forman 1996).

Table 1 lists <sup>14</sup>C dates of known-age samples; it can be observed that the conventional ages span a wide time interval. Therefore, in order to perform the best possible calibration, different  $\Delta R$  values should be applied to different sets of <sup>14</sup>C dates obtained from different organisms. Namely, <sup>14</sup>C dates from penguin remains should be calibrated using a  $\Delta R$  value derived only from penguin samples of known age ( $\Delta R = 688 \pm 55$  is the weighted mean of seven values from penguin remains;  $\Delta R = 656 \pm 55$  is the weighted mean of three values from Adélie penguin remains only).

			Historical	Conv. age	ΔR	
Sample no.	Location	Material	age (AD)	( <sup>14</sup> C yr BP)	( <sup>14</sup> C yr BP)	Reference
Lu31101	Hope Bay	Penguin bones	1903	1280 ± 50	816 ± 50	Bjork et al. 1991
4432	Cape Royds	Adélie penguin flesh	1904	925 ± 75	462 ± 75	Geyh and Wirth in White-
						house, Chinn and Hofle 1988
4433	Cape Adare	Flesh mew of prey	1902	1125 ± 90	660 ± 90	Geyh and Wirth in White-
						house et al. 1988
QL173	Inexpressible Is.	Emperor penguin	1912	$1300 \pm 50$	838 ± 50	Stuiver et al. 1981
QL171	Inexpressible Is.	Weddell seal	1912	1390 ± 40	928 ± 40	Stuiver et al. 1981
NZ6339A	Inexpressible Is.	Emperor penguin bones	1912	1065 ± 50	603 ± 50	Mabin 1985
NZ6327A	Inexpressible Is.	Weddell seal bones	1912	1760 ± 55	1298 ± 55	Mabin 1985
NZ6842A	Inexpressible Is.	Adélie penguin bones	1912	1060 ± 45	598 ± 45	Whitehouse et al. 1988
		and flesh				
NZ6872	Inexpressible Is.	Charcoal from seal	1912	$1240 \pm 45$	778 ± 45	Greenfield in Whitehouse et
		blubber stove				al. 1988
GX-12759	Inexpressible Is.	Seal bones	1912	1175 ± 75	713 ± 75	Orombelli 1988
NZ7079A	Cape Evans	Emperor penguin bone	1916	1105 ± 55	642 ± 55	Mabin 1986
		collagen				
NZ7076A	Cape Evans	Emperor penguin flesh	1916	$1220 \pm 55$	757 ± 55	Mabin 1986
		and feathers				
NZ6851A	Cape Evans	Weddell seal bone col-	1916	1610 ± 80	1147 ± 80	Mabin 1986
		lagen				
GX-18581	68°30′S-67°00′W	Adamussium colbecki	1940	1476 ± 39	1001 ± 39	Berkman and Forman 1996
GX-18582	67°52′S–67°17′W	Adamussium colbecki	1940	1416 ± 40	941 ± 40	Berkman and Forman 1996
AA 14785	68°47′S–90°35′W	Neobuccinum eotoni	1917	1215 ± 57	750 ± 57	Berkman and Forman 1996
GX-19205	78°30'S-164°20'W	Thracia meridionalis	1935	1278 ± 62	798 ± 62	Berkman and Forman 1996

TABLE	1	Radiocarbo	n Dates	from	Samples o	f Known	Age	from	Antarctica
IADLE	1.	Kaulocaloc	m Dates	nom	Samples U		ngu	nom	Amaicuca

With our *Penguin* program it is easy to recalibrate the set of data using both the mean value calculated from all the available samples or different  $\Delta Rs$  obtained from penguin remains only. Furthermore, regional values can be calculated and compared for different sets of data.

Calibration is performed by the computer program CALIB (Stuiver and Reimer 1993); it allows the manual insertion of the data or can take as input a text file that can be created with a word processor. Normally, sample data (code, locality, description) are maintained using common commercial database programs, although the latter are not able either to export data directly in CALIB file format or to import data results (mainly calibrated age ranges) from CALIB output text files; in both cases data must be transferred one at time by retyping or through a tedious copy-and-paste process. Alternatively, sample data can be kept in CALIB input text files and handled with a word processor. However, because such programs are not aware of the format of those files, it is very easy to accidentally modify their structure, making them unreadable by CALIB.

Penguin's main goal is data management, focusing on data exchange with CALIB files in order to speed up the recalibration procedure. Penguin allows easy maintenance of sample data files, much

like a database application. In our view, the program is particularly suited for the calibration procedure when one of the following conditions applies: 1) new <sup>14</sup>C dates are to be added to the set of data; 2) new samples of known age are supplied. In the first case, only the new conventional ages are to be calibrated, using one or more  $\Delta R$  values. In the latter case, the  $\Delta R$  values must be recalculated and the existing sets of <sup>14</sup>C dates must be recalibrated.

The usefulness of *Penguin* can be explained via an example (see the flow chart in Fig. 1). If we assume that new penguin remains (either bone or flesh) of historical known age have been discovered and dated, the existing  $\Delta R$  value based on penguin remains must be updated in order to take into account the new *datum* (actually a weighted mean is computed of all the  $\Delta R$  values derived from each dated sample). Then, this new  $\Delta R$  value is used to recalibrate all the penguin <sup>14</sup>C dates.

If we have our sample data in a *Penguin* file called "MySamples" (Fig. 2), all we have to do is to open this file, assign the new  $\Delta R$  value to all samples contained in the file and save a copy of the file in CALIB format. At this point we can use CALIB to calibrate all the dates. Calibration results are collected by CALIB in one



Fig. 1. Flowchart showing the role of *Penguin* in the calibration process. Data are kept in *Penguin* data files and CALIB input files are generated only when <sup>14</sup>C dates need to be calibrated. *Penguin* then puts into a unique file the calibration results extracted from each output file created by CALIB.



Fig. 2. *Penguin* allows easy editing of sample data files. Sample records are selected through the list on the left side of the window. Text boxes and radio buttons are provided for editing the content of the fields.

file for each sample, so after a calibration session there are as many files as the number of samples contained in the original input file. With *Penguin*, all these files can be parsed in order to extract the calibration results, which can be saved in unique file (Fig. 3). The current release of *Penguin* accepts only the calibrated age ranges from CALIB output files; future releases will allow extraction of all the other information. Furthermore, *Penguin* allows calibration results to be exported in standard text format files (variable-sized records with variable-sized fields separated by a tab character or comma) that can be directly read by database or spreadsheet programs.

Following a basic rule, common sample data should be kept in a *Penguin* file, which can be edited by either adding, deleting or modifying sample records; CALIB input files should be created only

		MySamples.r	es 🔤	
	Lab. number	Conv. Age	1 sigma range (B.P.)	囵
1	Lab-1	2015 ± 75	875 (737)659	]
2	Lab-2	4290 ± 50	3448 (3359) 3283	]
3	Lab-3	4495 ± 135	3806 (3602) 3423	]
4	Lab-4	5385 ± 85	4870 (4812) 4643	]
5	Lab-5	4190 ± 80	3360 (3254) 3119	]
6	Lab-6	4930 ± 85	4343 (4181) 4048	]
7	Lab-7	5770 ± 60	5427 (5295) 5232	]
8	Lab-8	5575 ± 185	5295 (5011) 4810	
9	Lab-9	2900 ± 90	1805 (1681 ) 1533	
10	Lab-10	5360 ± 90	4856 (4801 ) 4600	Ð
6	1		•	

Fig. 3. Calibration results are displayed in a spreadsheet-like window

for calibration purposes and then deleted when they are no longer needed. Of course, for each set of <sup>14</sup>C dates there will be as many *Penguin* calibration result files as the number of times the same set of dates has been calibrated.

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## INGEIS RADIOCARBON LABORATORY DATES III

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### INTRODUCTION

The following list presents results of dating of 55 paleoenvironmental and archaeological samples from Argentina, processed between 1984 and 1986 by M. A. Albero and F. Angiolini. Procedures for sample pretreatment, counting, statistical analysis and age calculation were essentially the same as previously described by Albero and Angiolini (1985). Results are reported as conventional <sup>14</sup>C dates in years before AD 1950. They are corrected for isotopic fractionation.

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### **PALEOENVIRONMENTAL SAMPLES**

### La Poma Series

Peat samples from Río Grande holocenic terraces of La Poma, Humahuaca, Jujuy (23°17'S, 65°42'W), 3600 m asl. Collected and submitted 1983 by J. Fernández.

AC-691	3280 ± 90
Depth 4.30 m	$\delta^{13}C = -25.9 \pm 0.2\%$
AC-692	3460 ± 120
Depth 4.50 m	$\delta^{13}C = -26.0 \pm 0.2\%$

Comment (J.F.): Dates related to leaf-shaped stemless points. The paleoenvironmental interpretation and chronology have been published in Coleman (1973) and Fernández (1973).

## **Guayatayoc Series**

Peat samples from Guayatayoc stream, Guayatayoc, Rinconada Jujuy (22°18'S, 66°02'W), 3500 m asl. Collected and submitted 1983 by J. Fernández.

AC-0733	9170 ± 170
Depth 1.82 m	$\delta^{13}C = -27.4 \pm 0.2\%$
AC-0734	10,390 ± 190
Depth 1.97 m	$\delta^{13}C = -26.5 \pm 0.2\%$

*Comment* (J.F.): Data belonging to this same profile were published in Albero and Angiolini (1985: 333). Human occupation, *ca.* 2800 yr BP, is indicated by quartzitic artifacts.

## Leuto Caballo Series

Peat, marl and other carbonatic sediments from terraces of the Bordo Leuto Caballo stream (37°05'S, 70°15'W),1670 m asl, Chos Malal, Neuquén. Collected and submitted 1983 by J. Fernández. All samples belong to peat layers.

AC-0980 Depth 9.67 m	$12,280 \pm 160 \\ \delta^{13}C = -25.0 \pm 0.2\%$
AC-0981 Depth 8.57 m	$11,940 \pm 160 \\ \delta^{13}C = -27.9 \pm 0.2\%$
AC-0979 Depth 7.67 m	$11,300 \pm 160 \\ \delta^{13}C = -25.0 \pm 0.2\%$
AC-0983 Depth 7.52 m	$11,070 \pm 140 \\ \delta^{13}C = -25.0 \pm 0.2\%$
AC-1048 Depth 7.32 m	$10,180 \pm 130 \\ \delta^{13}C = -7.0 \pm 0.2\%$

Comment (J.F.): Dates are related to a Late Pleistocenic marshy environment with megafaunal remains (Megatherium, Hippidion and Paleolama); see Panarello and Fernández (1992).

## **Barro Negro Series**

Peat, marl and collagen of several profiles from Barro Negro (23°S, 65°37'W), 3.820 m asl, Humahuaca, Jujuy. Collected and submitted 1980–1986 by J. Fernández. All samples belong to peat layers.

A) Quebrada Linda Profile (Site A)

AC-0748 Depth 0.20 m	$510 \pm 70$ $\delta^{13}C = -25.3 \pm 0.2\%$
<b>AC-0740</b> Depth 1.42 m	$940 \pm 80$ $\delta^{13}C = -27.6 \pm 0.2\%$
AC-0738 Depth 2.10 m	$\frac{1270 \pm 80}{\delta^{13}C = -25.4 \pm 0.2\%}$
AC-0739 Depth 2.80 m	$\frac{1690 \pm 80}{\delta^{13}C} = -27.6 \pm 0.2\%$
AC-0735 Depth 8.40 m	$12,530 \pm 160 \\ \delta^{13}C = -26.8 \pm 0.2\%$
B) Quebrada de Las Piedras Profile	
AC-0747 Depth 1.17 m	$1000 \pm 80 \\ \delta^{13}C = -24.0 \pm 0.2\%$
AC-0673 Depth 1.60 m	$\frac{1140 \pm 80}{\delta^{13}C} = -23.7 \pm 0.2\%$
AC-0682 Depth 1.70 m	$\frac{1680 \pm 120}{\delta^{13}C} = -25.0 \pm 0.2\%$
AC-0680 Depth 1.90 m	$\frac{2890 \pm 110}{\delta^{13}C} = -24.3 \pm 0.2\%$
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AC-0746 Depth 2.12 m	$3470 \pm 90$ $\delta^{13}C = -27.5 \pm 0.2\%$
AC-0745 Basal depth 5.82 m	$10,200 \pm 140 \\ \delta^{13}C = -25.0 \pm 0.2\%$
AC-0744 Basal depth 6.77 m	$12,300 \pm 170 \\ \delta^{13}C = -28.7 \pm 0.2\%$
Comment: Marl and collagen samples were found in basal peat layer	
AC-0974 Marl 2. depth 5.90 m	$9870 \pm 220$ $\delta^{13}C = -4.0 \pm 0.2\%$
AC-0975 Marl 4. depth 6.20 m	13,400 ± 400
AC-0976 Marl 5. depth 6.80 m	$11,600 \pm 160 \\ \delta^{I3}C = -3.8 \pm 0.2\%$
AC-0969 I Collagen from <i>Hippidion</i> bone.	$9120 \pm 130$ $\delta^{13}C = -18.0 \pm 0.2\%$
AC-0969 II Collagen from <i>Hippidion</i> bone.	$11,500 \pm 400 \\ \delta^{13}C = -18.0 \pm 0.2\%$
C) Zanjón Largo Profile	
AC-0742 Peaty silt with Lymnaea, depth 4.56 m	$9050 \pm 140$ $\delta^{13}C = -24.8 \pm 0.2\%$
AC-0743	9200 ± 140

Peaty silt with fragmented camel bones and lithic objects, depth 5.08 m  $\delta^{13}C = -24.8 \pm 0.2\%$ 

*Comment* (J.F.): Important superior Pleistocenic and Holocenic chronostratigraphic record with archaeological contents, fossil animal remains and paleoenvironmental indicators. Published in Alberdi *et al.* (1986); Fernández *et al.* (1991); Fernández (1983–1985).

## ARCHAEOLOGICAL SAMPLES

## **Haichol Series**

Charcoal samples from Haichol cave (38°35'S, 70°40'W) 1050 m asl. Pehuenches, Neuquén. Collected and submitted 1979–1981 by J. Fernández.

AC-0896 Grid 13B2. depth 0.40–0.50 m	$2350 \pm 150 \\ \delta^{13}C = -25.0 \pm 0.2\%$
AC-0897 Grid 9B1, depth 0.50–0.60 m	$\frac{1290 \pm 110}{\delta^{13}C = -22.8 \pm 0.2\%}$
AC-0898 Grid 14B1, depth 0.50–0.60 m	$\frac{1440 \pm 90}{\delta^{13}C} = -25.0 \pm 0.2\%$

AC-0899 Grid 16B2, depth 0.50–0.60 m	$2290 \pm 120 \\ \delta^{13}C = -22.6 \pm 0.2\%$
AC-0900 Grid 11B2, depth 0.60–0.70 m	$2130 \pm 110 \\ \delta^{I3}C = -22.9 \pm 0.2\%$
AC-0901 Grid 17B2, 0.80–0.90 m	$2440 \pm 100 \\ \delta^{I3}C = -21.9 \pm 0.2\%$

*Comment* (J.F.): Dates belonging to Late Preceramic occupations (2440–2130 yr BP) and Early Pottery occupations (1830–1250 yr BP). Published in Fernández (1988–1990).

## Vizcayachoc Peat

Peat from lower part of Vizcachayoc peat bog, north of Miyuyoc (22°55'S, 65°20'W) 4000 m asl, Humahuaca, Jujuy. Collected and submitted 1983 by J. Fernández.

AC-0982

 $9540 \pm 170$  $\delta^{13}C = -21.2 \pm 0.2\%$ 

Comment (J.F.): Lower part of peat bed 7 m width, on western slope of Cordillera Oriental.

#### **Pluma De Pato**

Eggshell of *Pterocnemia pennata* (choique) from Pluma del Pato, Añelo, Neuquén (38°10'S,69 5'W) 250 m asl. Collected and submitted 1979 by J. Fernández.

#### AC-1049

 $1700 \pm 90$  $\delta^{13}C = -8.0 \pm 0.2\%$ 

Comment (J.F.): Associated with ceramics and triangular stemless projectile points with concave base, made of obsidian.

#### Ojo De Agua

Interbedded wood at 2.10 m depth in a small fluvial terrace in Ojo de Agua, near Susques (23°23'S, 66°22'W, 3700 m asl. Collected and submitted 1983 by J. Fernández.

F	AC-1053	870 ± 85
7	Wood	$\delta^{13}C = -22.6 \pm 0.2\%$
(	Calcium carbonate biostromal accretion from Pastos Chicos (23°45'S	, 66°23'W) 3900 m asl. Col-
lec	ted and submitted 1983 by J. Fernández.	

AC-1056	$16,250 \pm 290$
	$\delta^{13}C = +8.8 \pm 0.2\%$

Comment (J.F.): Stromatolite (oncolite) from ancient Pastos Chicos stream flood terrace.

## **Anillaco Series**

Charcoal samples from Faldeos del Anillaco, La Rioja (28°46'S, 66°57'W.) 1500 m asl. Collected and submitted 1984 by R. Raffino.

AC-919 Depth 0.60 m  $1160 \pm 120$ 

AC-920 Depth 0.70 m	1200 ± 120
AC-921 Depth 0.80 m	2080 ± 120
AC-922 Depth 0.90 m	1330 ± 120
AC-923 Depth 1.00 m	$1330 \pm 120$ $\delta^{I3}C = -22.9\%$
AC-924 Depth 1.10 m	$1150 \pm 140$ $\delta^{13}C = -21.0\%$
AC-925 Depth 1.20 m	$1440 \pm 120$ $\delta^{13}C = -23.8\%$
AC-926 Depth 1.30 m	1270 ± 120

## La Huerta Series

Bone and charcoal samples from La Huerta, Jujuy, (23°20'S, 65°20'W) 2700 m asl. Collected and submitted 1985 by R. Raffino and published by Raffino *et al.*(1993).

AC-960	480 ± 100
Bone collagen, between 0.22 and 0.32 m	$\delta^{13}C = -16.5\%$
AC-1069	540 ± 90
Charcoal, depth 0.12 m	

## Valle De La Ciénaga Series

Charcoal samples from La Ciénaga valley, Tucumán (26°49'S, 65°39'W) 2700 m asl. Collected and submitted 1984 by B. Cremonte.

AC- 720	1560 ± 80
Depth 1.40 m	$\delta^{i3}C = est. 0$
AC-/21	1240 ± 80
Depth 0.80 m	$\delta^{13}C = -24.80\%$

Dates related to large settlements of early potter and agricultural societies from the northwest of Argentina, especially with those of Tafí valley. Published by Cremonte (1988).

## **Puesto El Rodeo Series**

Vegetal remains and charcoal samples from La Magdalena estancia, Río Pinturas area, Santa Cruz, (46°53'S, 70°27'W) 240 m asl. Collected and submitted 1985 by C. Gradin and A. Aguerre.

AC-943	1380 ± 90
Depth 0.47 m	$\delta^{13}C = -22.1\%$

AC-1075 Depth 0.90 m

*Comment:* Dates correspond to vegetal remains found over a bedded burial place containing three skeletons. The first date is related to the Rio Pinturas IV cultural level and the second one to the Rio Pinturas IIb–III cultural level.

#### **Puesto Giles Series**

Charcoal samples from Puesto Giles Site, Casa de Piedra, La Pampa (38°11'S, 67°13'W) 250 m asl. Collected and submitted 1983 by C. Gradin and A. Aguerre.

AC-728	$700 \pm 100$
Depth 0.53 m	$\delta^{I3}C = -21.3\%$
AC-731	320 ± 120
Depth 0.40 m	$\delta^{13}C = -21.7\%$

*Comment:* Dates related to a late human occupation of hunter-gatherers who used ceramics (under investigation).

#### **El Cuy Series**

Charcoal sample from El Cuy, Cañadón Santa Victoria, Río Negro (40°S, 68°W) 400 asl. Collected and submitted 1986 by C. Gradin and A. Aguerre.

AC-1074	$420 \pm 110$
Depth between 0.05-0.20 m	$\delta^{13}C = -21.2\%$

*Comment:* Date related to tool remains made of bone and a stemmed point, with small patagoniense-type fins. Investigation was stopped at prospecting stage.

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## UNIVERSITY OF GRANADA RADIOCARBON DATES VII

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#### INTRODUCTION

This paper includes determinations of archaeological, geological and palaeobotanical samples from Spain and Portugal, measured at the University of Granada Radiocarbon Dating Laboratory, from 1990 to 1991. Pretreatment of charcoal and wood samples is a standard acid-basic procedure using 8% HCl and 2% NaOH at boiling temperature. The collagen of bone samples is obtained by the Longin (1971) method. The method of dating is benzene synthesis and liquid scintillation counting.

As previously reported (González-Gómez 1992), the sample size and the scintillator were 7 ml low K-40 Packard counting picovials with 5 ml synthesized benzene and 83.5 mg Butyl-PBD directly dissolved in the benzene (16.7 g/liter) as a scintillator; smaller samples were expanded to 5 ml with inactive benzene. <sup>14</sup>C activity was measured in a Packard Tri-Carb<sup>®</sup> Mod 4640 liquid scintillation spectrometer. Efficiency was ~65% using the part of the spectrum above the end point of tritium, with a background of ~7 cpm. At least one modern reference standard and two background vials were measured together with each series of measurements. All results are corrected for fractionation according to the quoted  $\delta^{13}C$  (w.r.t. PDB) values.

In order to prevent any loss of benzene during counting and storage, we sealed the vials hermetically using metallic caps. The joint was made of a sheet composed of 3 mm silicone + 0.05 mm Teflon<sup>TM</sup>. As a second barrier we placed a silicone O-ring, 10 mm in inner diameter and 2 mm thick, around the neck of the vial. The weight of vials, checked one year after filling, remained constant (González-Gómez 1992).

Dates reported here are based on 0.95 of the activity of NBS oxalic acid modern standard, on the Libby <sup>14</sup>C half-life of 5568 yr, and are expressed in radiocarbon years relative to AD 1950, as suggested by Stuiver and Polach (1977). Samples were measured for 100 min repeated 40–45 times, as well as background and standard vials. The standard deviation quoted includes only 1  $\sigma$  of the counting statistics of background, sample and modern standard counts. Calculations and data were processed using a PC running a general program for radiocarbon dating laboratories written by González-Gómez (1995). Calibrated ages for a 2  $\sigma$  interval were obtained by the method of Pearson *et al.* (1986), running the computer programs CALIB (Stuiver and Reimer 1986) and CALI (González-Gómez 1988) and using the bidecadal curve for samples of atmospheric origin. Sample descriptions and comments are essentially based on information provided by submitters.

#### ACKNOWLEDGMENT

We thank the Departamento de Química Analítica, Facultad de Química, Universidad de Barcelona, for the  $\delta^{13}C$  determinations.

#### **ARCHAEOLOGICAL SAMPLES**

Spain

#### Acinipo. Ronda La Vieja Series

Charcoal samples from Acinipo, Ronda la Vieja (36°45'N, 5°10'W), Ronda, Málaga province, were collected from 1986 to 1988 by O. Garrido and M. Carrilero and submitted in 1990 by P. Aguayo, Departamento de Prehistoria y Arqueología, Universidad de Granada, to date the Bronze Age and particularly the building systems of the habitats in the site.

UGRA-360. Ac'88 - 41568	$3720 \pm 70$
Charcoal from depth 6.92 m; 2350–1930 cal BC.	$\delta^{13}C = -26.2\%$
UGRA-374. Ac'86 - 40710	$3640 \pm 70$
Charcoal from depth 6.87 m; 2200–1786 cal BC.	$\delta^{13}C = -24.8\%$
UGRA-375. Ac'86 - 40718	$3640 \pm 50$
Charcoal from depth 6.40 m; 2180–1890 cal BC.	$\delta^{13}C = -24.5\%$

General Comment: Expected ages were  $\sim 3710 \pm 250$  BP.

#### **Cerro De Los Encaños Series**

Charcoal samples from Cerro de los Encaños (40°03'N, 2°27'W), Villar del Horno, Pajaroncillo, Cuenca province, were collected and submitted 1990 by A. Gómez-Ruiz, Granada, to date the violent destruction and abandonment of an Iron Age village.

UGRA-363. VILLAR I	2475 ± 50
Sample from depth 1.6 m; 800-400 cal BC.	$\delta^{13}C = -25.1\%$

Comment: This sample corresponds to a temporary abandonment of the village, in the first Iron Age; expected age was  $\sim 2460 \pm 100$  BP.

UGRA-367. VILLAR II	1940 ± 50
Sample from depth 1.2 m; 60 cal BC-cal AD 140.	$\delta^{13}C = -23.5\%$

*Comment:* This sample corresponds to the definitive abandonment of the village, in the second Iron Age.

#### Los Barruecos Series

Charcoal samples from Los Barruecos (39°25'N, 6°30'W), Malpartida de Cáceres, Cáceres province, were collected and submitted 1987 by M. I. Sauceda-Pizaro, Area de Prehistoria y Arqueología, Facultad de Filosofía y Letras, Universidad de Extremadura, Cáceres, to date not only the site but also the later prehistoric sequence in the Extremadura region.

UGRA-352. № 1	4000 ± 70
Sample from depth 65 cm; 2865–2340 cal BC.	$\delta^{13}C = -24.5\%$
UGRA-354. № 3	3710 ± 80
Sample from depth 1.0 m; 2451–1890 cal BC.	$\delta^{13}C = -24.7\%$

## UGRA-350. Museo de El Puerto de Santa María. IMAGEN. 870 ± 60

 $\delta^{13}C = -25.0\%$ 

Wood sample from the Museum of the town of El Puerto de Santa María, Cádiz province, submitted 1987 by J. de Lucas-Almeida. The sample dated an image of which the original polychromy had disappeared; cal AD 1020–1270.

Comment: Expected age was ~760 BP.

## UGRA-349. El Pla del Riu de les Marcetes PR

## 5040 ± 100

 $\delta^{13}C = -19.7\%$ 

Bones from El Pla del Riu de les Marcetes (41°23'N, 1°50'E), Manresa, Barcelona province. Sample collected and submitted 1986 by J. Castany-Llusa, Instituto de Prehistoria y Arqueología, Universidad de Barcelona, to date a Neolithic grave. Ceramics and flint objects were found in the burial.

Comment: Sample from depth 1.2 m; expected age was ~5210 ± 250 BP; 4038-3640 cal BC.

## **UGRA-351 Puerto Guetaria PG**

 $900 \pm 50$  $\delta^{13}C = -27.6\%$ 

Wood from Puerto Guetaria (43°18'N, 2°12'W), Guetaria, Guipuzcoa province. Sample collected and submitted 1988 by A. M. Benito, Sociedad de Ciencias Aranzadi, Museo San Telmo, San Sebastián. The sample was in a hole in a stone three-cornered anchor submerged at the entrance of the Guetaria port.

Comment: Sample from depth 9 m (under the sea level); cal AD 1020-1250.

#### **Ronda City Area Series**

Samples from Ronda city area (36°45'N, 5°10'W), Ronda, Málaga province, were collected from 1985 to 1989 by B. Padial, J. Castilla and P. Aguayo and submitted from 1990 to 1992 by P. Aguayo, Departamento de Prehistoria y Arqueología, Universidad de Granada, to date several periods in the city as detailed below.

UGRA-376. Ro'89 - 7556	$2400 \pm 50$
765-390 cal BC. Expected age was ~2310 ± 150 BP.	$\delta^{13}C = -25.4\%$

Comment: Charcoal from depth 3.2 m, to date the use of a kiln and its pottery production.

UGRA-423. RO-89-AA-6695	955 ± 40
cal AD 1001–1168.	$\delta^{13}C = -25.0\%$

*Comment:* Charcoal from depth 3.5–3.6 m, to date the Iberic levels of establishment from Prehistory to the present time.

UGRA-427. RO-89-AA-6695	985 ± 40
cal AD 981–1159.	$\delta^{13}C = -25.0\%$

Comment: This is a repetition of the previous sample UGRA-423, in order to confirm the result.

UGRA-424. RO-87-AA-6136	415 ± 45
cal AD 1419–1628.	$\delta^{13}C = -25.0\%$

Comment: Charcoal from depth 3.1-3.2 m, to date the same as UGRA-423.

UGRA-425. RO-89-AA-6671	930 ± 40
cal AD 1015–1213.	$\delta^{13}C = -25.0\%$

Comment: Charcoal from depth 3.4 m, to date the same as UGRA-423 and UGRA-425.

UGRA-426. RO-89-AA-C/7	2495 ± 50
800-410 cal BC. Expected age was ~2700 ± 250 BP.	$\delta^{13}C=-25.0\%$

Comment: Wheat seeds sample from depth 4.5 m, to date Final Bronze Age levels in the site.

UGRA-432. RO-85 GC 3090	2730 ± 70
1040–800 cal BC.	$\delta^{13}C = -25.0\%$

Comment: Charcoal sample, to date levels of superimposed villages.

#### Las Viñas Series

Shell samples from Las Viñas (36°39'N, 6°10'W), El Puerto de Santa María, Cádiz province, were collected and submitted 1987 by F. Giles-Pacheco, Municipal Museum of El Puerto de Santa María.

UGRA-369. Silo nº 50	4800 ± 90
3350–2890 cal BC.	$\delta^{13}C = +0.15\%$

Comment: Shells from depth 1.6 m, to date the geographic area of western Andalusia.

UGRA-370. Silo nº 16	4950 ± 60
cal AD 1001–1168.	$\delta^{13}C = -1.00\%$

*Comment:* Shells from depth 1.2–1.5 m. This is the first dating of the site context; it can be related to nearby Chalcolithic sites with similar pottery.

#### PORTUGAL

UGRA-355 Mamoa 1 de Chã de Carvalhal Amostra nº 2	5860 ± 60
	$\delta^{13}C = -25.7\%$

Charcoal from Mamoa 1 de Chã de Carvalhal (41°08'N, 8°05'W), Serra da Aboboreira, Baião, Distrito Porto, Douro Litoral province. Sample collected 1986 and submitted 1988 by D. de Jesus da Cruz, Instituto de Arqueologia, Faculdade de Letras, Universidade de Coimbra. The sample is from a megalithic necropolis with more than 40 tumulis. To date the Megalithic tomb episode in the region.

Comment: Sample from depth 1.29 m; 4900-4586 cal BC.

#### **GEOLOGICAL SAMPLES**

Spain

#### **Rambla Guzmaina Series**

Wood samples from Rambla Guzmaina (37°24'N, 1°58'W), Huercal Overa, Almería province, collected 1990 by A. Martín-Penela and submitted 1990 by L. García-Rossell, Instituto Andaluz de Geología Mediterránea, Facultad de Ciencias, Universidad de Granada, to study an episode of recent sedimentation in the Almanzora River basin.

UGRA-371. G-A-I Sup.	$200 \pm 60$
Sample from depth 1.3 m; cal AD 1522–1955.	$\delta^{13}C = -25.3\%$

UGRA-372. G-A-I Inf.	> Modern
Sample from depth 2.3 m.	$\delta^{13}C = -24.9\%$
UGRA-373. G-A-II	> Modern
Sample from depth 30 cm.	$\delta^{13}C = -25.0\%$

PALAEOBOTANICAL SAMPLES

#### Spain

#### **Lobeiras Series**

Samples from Lobeiras (43°25'N, 7°31'W), Vivero, Lugo province, collected and submitted 1989 by M. P. Saa-Otero, Colegio Universitario de Orense, Las Lagunas, Orense province, to establish the vegetation existing in this zone during the Quaternary.

UGRA-347. LOB-1	$2080 \pm 60$
Peat sample from depth 51 cm; 354 cal BC-cal AD 52.	$\delta^{13}C = -28.1\%$
UGRA-348. LOB-2 Wood sample from depth 1.2 m.	$9800 \pm 120$ $\delta^{13}C = -26.3\%$
UGRA-324. Pico del Lobo. PLB II C'	$1830 \pm 110$ $\delta^{13}C = -28 \ 1\%$
	0 C = -20.1700

Peat from Pico del Lobo (40°59'N, 3°58'W), La Pinilla, Segovia province; sample was collected and submitted 1989 by M. J. Gil-García, Departamento de Geología, Universidad de Alcalá de Henares, Madrid province, to establish the vegetation history of this zone, cal AD 670–1020.

#### **Puerto De Canencia Series**

Peat samples from Puerto de Canancia (40°52'N, 3°47'W), Canencia de la Sierra, Madrid province, were collected 1988–1990 and submitted 1990 by M. J. Gil-García, Departamento de Geología, Universidad de Alcalá de Henares, Madrid province, to establish the vegetation history of this zone and confirm the pollen data.

UGRA-368. PCO	1170 ± 50
Sample from depth 80–90 cm; cal AD 714–980.	$\delta^{13}C = -27.9\%$
UGRA-411. PCO 2	2580 ± 80
Sample from depth 1.20–1.45 m; 900–421 cal BC.	$\delta^{13}C = -27.9\%$

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## **RESEARCH CENTER OF RADIOISOTOPES AT UNIVERSITY OF OSAKA PREFECTURE RADIOCARBON DATES I**

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## INTRODUCTION

The radiocarbon dating laboratory has been in operation since 1984 at the Radiation Center of Osaka Prefecture (OR), predecessor of the Research Center of Radioisotopes, University of Osaka Prefecture. We use liquid scintillation counting (LSC), following sample conversion to methanol through combustion and LiAlH<sub>4</sub> reduction. This method was developed by Yamada *et al.* (Yamada, Higashimura and Shidei 1966; Yamada and Kobashigawa 1986). In cooperation with Yamada, we somewhat modified their procedure: 1) sample charcoal is burned at 700°C in the presence of CuO needles and Sulfix grains to remove sulfur and halogen compounds produced during the combustion; 2) the combustion is carried out by using  $N_2$ - $O_2$  mixed gas of minimized  $O_2$  content and stopped when a small amount of the charcoal still remains unchanged, because precise investigation of methanol preparation revealed that O<sub>2</sub> gas stimulates byproduct formation during LiAlH<sub>4</sub> reduction (Shibata et al. 1985; Shibata et al. 1993). Usually, methanol is prepared directly from sample charcoal in a reaction apparatus ("direct method"). When the sample quantity is insufficient, generated CO<sub>2</sub> is isolated as CaCO<sub>3</sub> and diluted with inactive commercial CaCO<sub>3</sub> if necessary (< 40 g of CaCO<sub>3</sub> yield). Then CaCO<sub>3</sub> is hydrolyzed with HCl to CO<sub>2</sub> for methanol preparation in the usual way ("separate method"). We use standard oxalic acid SRM 4990C (HOxII) for determination of modern <sup>14</sup>C (Stuiver 1983). The acid is oxidized to  $CO_2$  using the wet method of Valastro, Land and Valera (1977) followed by methanol preparation in the same manner as for unknown samples.

Methanol obtained is purified and checked via gas chromatographic analysis prior to <sup>14</sup>C measurement. Six grams of methanol (7 g, later) are dissolved into 11 g (4 g, later) of homemade xylene cocktail for LSC, using a Packard TRI-CARB<sup>®</sup> 460CD). The counting efficiency is measured using a series of quenched standards close in composition to sample cocktails, while counting conditions are continually checked by measuring methanol prepared from contemporary rice grains as a secondary standard. Sample and background cocktails are alternatively measured for 100 min to attain desired statistical errors. We measure <sup>14</sup>C in HOxII for every batch of samples. The averaged value is 18.209 ± 0.105 dpm/gC (one standard deviation, 1  $\sigma$ ) for all measurements so far (11 times).

Smokeless coal was used for a blank test of methanol preparation and gave a <sup>14</sup>C date of >35,000 yr BP under routine conditions. As a control on our procedures, we <sup>14</sup>C-dated several samples that had been dated by other laboratories. Our results were quite consistent with <sup>14</sup>C dates of other laboratories (Shibata *et al.* 1990).

Samples dated were wood, charcoal and peat. The quoted errors (at the 1  $\sigma$  confidence level) include the standard deviations of count rates for unknown, background and modern samples. We report dates according to the guidelines of Stuiver and Polach (1977), *i.e.*, in yr BP, based on the conventional Libby half-life for <sup>14</sup>C of 5568 yr. Correction for isotopic fractionation was not made. Calendar years shown in parentheses were age ranges (2  $\sigma$ ) calculated by use of Method B (Probabilities) in the CALIB calibration program, Version. 3.3 (Stuiver and Reimer 1993).

#### **ARCHAEOLOGICAL SAMPLES**

JAPAN

#### Kankoji Series

Kankoji site, Kanan-cho, Minamikawachi dist, Osaka pref (34°2'N, 135°37'E), 102 m asl, is a typical upland village of Late Yayoi Age. Site was investigated by Educ Bd, Osaka pref. Collected and submitted 1985 by H. Otani, Educ Bd, Osaka Pref.

#### OR-5. KNJ-No. 1

1680 ± 85

1550 + 05

Charcoal from hearth in rounded square ancient dwelling (sides 3.7 by 3.6 m) No 1, located at the central part of Kankoji site (cal AD 147 to 591).

Comment (H.O.): Estimated as Late Yayoi Age. Result reasonable.

OR-9. KNJ-No. 2 29,260 ± 3860 Wood from dark-gray clay layer, *ca.* 3 m deep, at northwest corner of Kanroji pond located at northeast edge of Kankoji site.

Comment: Separate method with CaCO<sub>3</sub> dilution was used.

OR-6. KNJ-No. 3	32,000 ± 1810
Wood from dark green-gray sandy clay layer, just under portion of OR-9.	
OR-7. KNJ-No. 4	>37800

Wood from black-gray clay layer, ca. 4 m deep, at same place as OR-9.

## Shinpukuji Series

OR.13 Sample No. 1

Shinpukuji site, Mihara-cho, Minamikawachi dist, Osaka pref (34°32'N, 135°33'E) includes remains of temple bell foundry, kilns and structures from Nara to Middle Age. Site was investigated 1984 to 1985 by joint Educ Bd, Osaka pref and Osaka ctr of Cultural Properties. Samples collected at kiln remain, 41 m asl, and submitted 1985 by A. Yamamoto, Osaka ctr of Cultural Properties.

658).
<b>1190 ± 55</b> re (cal AD 706 to 979).
<b>1380 ± 190</b> 1024).
<b>1540 ± 60</b> 2 and No 5 (cal AD 418
<b>1570 ± 80</b> 270 to 649).
<b>1410 ± 65</b> 535 to 778).
1230 ± 55

### OR-12. Sample No. 9

Charcoal from charcoal-kiln No 1 (cal AD 452 to 682).

Comment: Separate method was used for all samples in this series. Samples OR-13, -16 and -17 were diluted with inactive CaCO<sub>3</sub>. Tile kilns No 2 and No 5 est. as Nara Age (AY) and <sup>14</sup>C dates reasonable. Age of charcoal kiln No 1 was either AD 8 or 13 (AY). <sup>14</sup>C dates matched AD 8. Archaeological magnetic date for charcoal kiln No 1 (either AD 730 or 600, H. Shibuya) was quite consistent with our results.

## **Joyama Series**

Joyama site, Hirano Ward, Osaka city, Osaka pref (34°36'N, 135°35'E), 10 m asl, includes many remains from Jomon to Modem Age. Site was investigated 1983 to 1985 by joint Educ Bd, Osaka Pref and Osaka Ctr of Cultural Properties. Samples were in Trench D, at 3 m depth. Collected 1984 to 1985 and submitted 1985 by S. Kanbayashi, Osaka ctr of Cultural Properties.

#### **OR-18. SB0901W-11**

Fragments of burnt wood from rectangular ancient dwelling 5.7 m by 4.3 m, SB0901. Associated with earthenware of Yayoi Age (cal 380 to 68 BC).

#### **OR-19. SB1001W-4**

Fragments of burnt board from ancient dwelling (ca. 7 m in diam), SB1001. Associated with earthenware of Yayoi Age (cal 393 to 50 BC).

#### OR-19'.

Same sample as OR-19. Re-dated at submitter's request to confirm or correct date for OR-19, above (cal 345 BC to AD 71).

OR-20. SB1001W-41 Fragments of burnt wood from same dwelling as OR-19 (cal 66 BC to AD 194).

#### **OR-21. SB1004W-38**

Burnt stick wood from ancient dwelling (8.1–6.0 m in diam), SB1004. Associated with earthenware of Yayoi Age and a spindle wheel (cal 372 BC to 10).

#### **OR-22. SB1004W-14**

Burnt board wood from same dwelling as OR-21 (cal 357 BC to AD 72).

Comment: Burnt materials such as grounds and woods were scattered throughout these ancient dwellings. These were estimated as fire-destroyed dwellings in Late Yayoi Age: dwelling SB0901 est. as middle part and dwellings SB1001 and SB1004 estimated as early part of Late Yayoi Age (SK). Sample for OR-22 was also dated by Kyoto Sangyo Univ (KSU). Result (1970 ± 20 BP, cal 150 BC to AD 205) agreed with our datum at 2  $\sigma$  confidence level.

#### **Tai Series**

Tai site, Mihara-cho, Minamikawachi dist Osaka pref (34°32'3"N, 135°35'42"E) is located at midpoint of terrace, ca. 39 m asl. Investigated 1986 by Osaka ctr of Cultural Properties. Collected and submitted 1986 by K. Kuninori.

OR-24. Driftwood 1	31,110 ± 1640
Driftwood from bluish-green clay layer 12, Trench 1A, at 2 m depth.	
OR-25. Driftwood 2	30,470 ± 1520
Driftwood from just under portion of OR-24.	

#### $1450 \pm 55$

 $2080 \pm 60$ 

 $2190 \pm 60$ 

 $2200 \pm 80$ 

 $1970 \pm 50$ 

 $2150 \pm 65$ 

 $2090 \pm 75$ 

## Kosaka Series

Kosaka site, Hirai, Sakai city, Osaka pref (34°30'55"N, 135°29'33"E), at valley in northern extremity of Senboku Hill, 22–25 m asl, includes many remains from Jomon to Modern Age. Site was investigated 1985 to 1990 by joint Educ Bd, Osaka Pref and Osaka Ctr of Cultural Properties. Samples collected at H, F and I dists.

## H dist group

H dist is located at bed of Hara Reservoir. Collected and submitted 1988 by K. Kuninori.

<b>OR-58. Tree sample A</b> Piece of tree from layer 12, Trench 1H, at 1.5 m depth (cal 919 BC to 530).	2630 ± 65
<b>OR-59. Tree sample B</b> Piece of tree sample same as OR-58 (cal 114 to 802 BC).	2770 ± 75
<b>OR-60. Tree sample C</b> Piece of tree sample same as OR-58 (cal 1113 to 813 BC).	2790 ± 60
<b>OR-61. Tree sample D</b> Piece of tree sample same as OR-58 (cal BC 1113 to 600).	2710 ± 80
<b>OR-62. Peat No. 1</b> Peat from dark brown peat layer 13, Trench 1 H, just under portion of tree samp to 1742 BC).	<b>3600 ± 75</b> le, above (cal 2140
<b>OR-63. Peat No. 2</b> Peat from same portion as OR-62 (cal 2031 to 1613 BC).	3500 ± 80
Comment: Final Jomon Age remains were found at just upper portion of tree samp -60 and -61. Samples were est. as before Final Jomon Age (KK). <sup>14</sup> C dates for sponded to Late Jomon Age.	ple for OR-58, -59, tree sample corre-
F dist group	
Collected and submitted 1988 by K. Akaki, Senboku branch office, Osaka ctr of C	Cultural Properties.
OR-69. Water pipe 1 Fragment of water pipe, 42 cm in diam 5.3 m long and 5–12 cm thick, from drai 1 OF (cal AD 664 to 878).	1280 ± 55 in in layer 4 Trench
<b>OR-67. Driftwood 2</b> Driftwood from same place as OR-69 (cal AD 672 to 883).	1260 ± 50
<b>OR-68. Driftwood 3</b> Driftwood from drain in layer 4 or 5, Trench 12F (cal AD 666 to 883).	1270 ± 55
<i>Comment:</i> Samples for OR-69 and -67 est. as Nara Age (KA) and sample for O before Kofun Age (KA). <sup>14</sup> C dates for these three samples corresponded to I including these samples was classified to old river 4 of Nara Age.	)R-68 estimated as Nara Age. Portion
<b>OR-64. Driftwood 8</b> Driftwood from drain 3 in layer 4, Trench 16F (cal 768 to 405 BC).	2460 ± 65
OR-70. Driftwood 10	2560 ± 60

Driftwood from valley-sand layer in Layer 4, Trench 16F (cal 820 to 424 BC).

OR-73. Decayed plant 11 2490 Decayed plant from valley-humus layer in layer 4, Trench 16F (cal 783 to 415 BC).	± 65
OR-82. Root 12   3630     Root, Trench 16F (cal 2182 to 1775 BC).   3630	± 65
<i>Comment:</i> Sample for OR-64 est. as Kofun Age and samples for OR-70 and -73 est. as before K Age (KA). <sup>14</sup> C dates for these samples corresponded to Late Jomon Age. They were assigned period before Final Jomon Age.	ofun ed to
OR-72. Board 7   1510 ±     Board from base of layer 6, Trench 8F (cal AD 264 to 766).   1510 ±	: 110
OR-65. Root 6   2570     Root from same place as OR-72 (cal 818 to 526 BC).   2570	± 50
OR-71. Root 5 2640 Root from base of layer 6, Trench 9F. Associated with earthenware of Final Jomon Age (cal to 546 BC).	<b>± 60</b> 917
<i>Comment:</i> Samples for OR-72, -65 and -71 est. as Final Jomon Age (KA). <sup>14</sup> C date for OR-72 younger than Yayoi Age. Portion including sample for OR-72 was classified to Kofun Age riv Samples for OR-65 and -71 were assigned to periods before and just Final Jomon Age, respectively.	was ver 6 vely.
OR-66. Root 4   3320     Root from base of layer 6, Trench 11 F (cal 1738 to 1447 BC).   3320	± 65
<b>OR-74. Carbonized root 9</b> 3790Carbonized root from base of layer 6, Trench 12F (cal 2455 to 1984 BC).	± 75
I dist group	
I dist is at flood plain located at left bank of Toki River. Samples collected at Trench 51 and mitted 1988 by K. Akaki.	sub-
OR-75. 1 2350 Peat from the highest peat layer in old river route (cal 534 to 210 BC).	± 45
OR-76. 2 3390 Peat from the second peat layer at same place as OR-75 (cal 1877 to 1517 BC).	± 70
OR-77.3 3800 Peat from the third peat layer at same place as OR-75 (cal 2455 to 2034 BC).	± 70
OR-78. 4 4490 Peat from the fourth peat layer at same place as OR-75 (cal 3356 to 2926 BC).	± 70
OR-79. 6 2500 Peat from peat layer in another portion of the old river route, above (cal 791 to 417 BC).	± 65
<b>OR-80. 7 2600</b> Piece of tree from peat layer in wetland behind the old river above (cal 903 to 425 BC)	± 75
OR-81. 8 2570   Peat from peat layer including tree sample for OR-80 (cal 834 to 418 BC).	± 75

## OR-83.5

8990 ± 230

 $1060 \pm 55$ 

Peat from peat layer in natural levee of the old river route, above (cal 8522 to 7534 BC).

*Comment:* Sample for OR-83 est. as before 500 BC and others est. as between 500 BC and AD 800 (KA). <sup>14</sup>C date for OR-83 corresponded to Initial Jomon Age. <sup>14</sup>C dates for OR-75, -76, -77, -78 and -79 were dispersed during Middle to Final Jomon Age. These samples showed more and more low <sup>14</sup>C activity along with their depth, helping to determine start chronology at the area. The old river was named as Jomon River 28. Samples for OR-80 and -81 were classified as Final Jomon to early part of Yayoi Age.

Syria

OR-36.

Chip of wood from ancient sunken ship found at bottom, 32 m deep, 2 km off coast of Tartous, Syria (34°54'N, 35°51'E). Excavation was made (1985 to 1987) for research into maritime trade history in the Mediterranean jointly by the Directorate of Antiquities and Museums of the Syrian Arab Republic and the Syrian Coastal Archaeological Excavation Operation Committee of Japan. More than 5000 amphorae were found in the sunken ship (cal AD 879 to 1157).

*Comment:* Initially, amphorae est. as Greco-Roman period. <sup>14</sup>C date for OR-36 corresponded to period from 9th to 12th century. Age of the sunken ship was eventually determined to be the first half of 13th century AD on the basis of heat luminescence date and <sup>14</sup>C dates by Nagoya University (Tandetron Accelerator Mass Spectrometry), in addition to our result.

#### **GEOLOGICAL SAMPLES**

JAPAN

#### **Buried Forest of Japan Cedar Series**

Japanese cedars (*Cryptomeria japonica* D. DON) buried under rice field, 50–80 cm deep, Mikatacho, Fukui pref (35°31′50″N, 135°53′22″E), 20 m asl, were investigated and submitted 1986 by H. Takahara, Osaka Agric Res Center (Kyoto pref Univ. For, since 1989).

#### OR-26. KR-W-1

 $3390 \pm 55$ 

Chip of buried Japanese cedar, 35 cm in diam. There was a feeding-trace due to insects.

#### OR-28. KR-W-2

 $3130 \pm 70$ 

Chip of buried Japanese cedar, 30 cm in diam and 6 m long. There were three feeding-traces due to insects.

Comment (HT): Feeding traces were recognized as larval tunnels of sugi bark borers, Semanotus japonicus Lacordaire (Coleoptera: Cerambycidae). Samples yielded first date when sugi bark borers had been inhabitant in Japan (Takahara, Ito and Takeoka 1988).

#### Kigo and Yakumogahara Moors Series

Samples from Kigo moor, Miyazu city, Kyoto pref (35°40'N, 135°11'E), alt 550 m, and from Yakumogahara moor, Shiga district, Shiga pref (35°15'15"N, 135°54'46"E), alt 910 m, were investigated and submitted 1987 by H. Takahara.

## OR-54. KG-46-58

Peat from Kigo moor, at 46–58 cm depth.

910 ± 100

<b>OR-27.</b> Peat from Kigo moor, at 520–530 cm depth.	14,760 ± 220
<b>OR-55. YK-B-47-55</b> Peat from Yakumogahara moor at 47–55 cm depth	1050 ± 90

*Comment:* Separate method with  $CaCO_3$  dilution was used for OR-54 and -55. Pollen analysis revealed that just upper portion of OR-54 or -55 corresponded to increasing period of pine tree forest at each region (HT). These <sup>14</sup>C dates helped determine beginning of natural forest destruction due to human activities, because pine tree forest tends to increase in parallel with destruction of the natural forest (HT). OR-27 est. age from 10,000 to 15,000 years BP and <sup>14</sup>C date was as expected.

#### **Koseinuma Moor Series**

Koseinuma moor, Yabu district, Hyogo pref. (35°21'N, 134°31'09"E), alt 1470 m, is located in Japanese cedar forest near top of Mt. Hyonosen. Collected 1989 to 1991 and submitted by H. Takahara.

OR-107. KS-23-33 Peat from peat layer, 23–33 cm deep.	780 ± 75
<b>OR-108. KS-50-60</b> Peat from same peat layer as OR-107, 50–60 cm deep.	1600 ± 75
OR-109. KS-85-95 Peat from same peat layer as OR-107, 85–95 cm deep.	2380 ± 60
OR-97. KS89-II	3450 ± 90

Peat from clayey peat layer at same place as OR-107, 108–118 cm deep.

*Comment:* Separate method with  $CaCO_3$  dilution was used for OR-107, -108 and -97. Samples were measured to date changes in natural forests of Japanese cedar or pine tree at Mt. Hyonosen. <sup>14</sup>C activity for these peat samples decreased linearly along with their depth, implying constant speed of deposition at the area.

#### OR-110. IH-520-530

#### 8820 ± 360

Peat, 520–530 cm deep, from Ikenotaira moor, Ichishi dist, Mie pref (34°31'27"N, 136°10'43"E), alt 600 m. Collected and submitted 1990 by H. Takahara.

Comment: Akahoya ash (ca. 6400 BP) was found at 264–271 cm depth and sample estimated as deposited during last glacial period (HT).

#### OR-94.

#### $230 \pm 45$

 $6530 \pm 95$ 

Peat, 1170–1200 cm deep, from Ukishimanomori moor, Shingu city, Wakayama pref (33°43'N, 135°59'E). Collected and submitted 1989 by M. Takeoka, Kyoto pref Univ.

*Comment:* Akahoya ash was found at 2000 cm depth and sample est. as 3000 to 4000 BP, whereas <sup>14</sup>C date was nearly modern (MT). Separate method with inactive CaCO<sub>3</sub> dilution was used.

#### OR-95. KR-89-2

Peat, at 134–143 cm depth, from Karakemi moor, Kita-azumi dist, Nagano pref (36°29'10"N, 137°53'35"E), alt 945 m, located in cold temperate zone of Shinshu area. Collected and submitted 1989 by M. Takeoka.

Comment: Sample was measured to date natural forest changes at inland area of Central Japan (TM). Separate method was used.

#### OR-96. OG-89-3

Peat, at 180–190 cm depth, from Ohoawara moor, Ina dist, Nagano pref (35°52'58"N, 138°10'53"E), alt 1815 m. Collected and submitted 1989 by M. Takeoka.

Comment: Sample was measured to date coniferous tree forest changes at subalpine zone in Central Japan (TM). Separate method was used.

#### OR-100. TIK-436.6-446.6

 $2530 \pm 50$ 

 $1670 \pm 150$ 

Peat, at 436.6–446.6 cm depth, from Ikenotaira moor, Higashi-tonami dist, Toyama pref (36°25'N, 136°52'30"E). Collected and submitted 1989 by M. Takeoka.

#### OR-101. NA-350-360

Peat, at 350–360 cm depth, from Nawagaike moor, Tonami dist, Toyama pref (36°28'30"N, 136°56'E), alt 810 m. Collected and submitted 1990 by M. Takeoka.

*Comment:* Methanol yield was unexpectedly low due to bad sample contamination with soil. <sup>14</sup>C date was obtained by using corrected <sup>14</sup>C activities of crude methanol (97% purity).

#### Kaminokodaira Series

Kaminokodaira, Nakashinkawa dist, Toyama pref (36°34′6″N, 137°30′31″E). Collected and submitted 1990 by M. Takeoka.

OR-103. KA-I-35-41	6580 ± 65
Peat, at 35-41 cm depth, alt 1410 m.	
OR-104. KA-II-60-66	$1240 \pm 40$
Peat, at 60-66 cm depth, alt 1425 m.	
OR-102. KA-II-90-94	10,960 ± 300
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Peat, at 90–94 cm depth, from same place as OR-104.

*Comment:* <sup>14</sup>C date for OR-102 was determined by using corrected <sup>14</sup>C activity of crude methanol (98% purity) because of unexpectedly low yield of methanol due to bad sample contamination with soil. Separate method was used for OR-103 and -104.

#### BORNEO

#### South Kalimantan Series

Peat from swamp in South Kalimantan, Bomeo (3°7'S, 114°30'E). Collected and submitted 1986 by H. Furukawa, Center for Southeast Asian Studies, Kyoto University.

OR-39. BM41 Peat, 25–75 cm deep.	1420 ± 70	
<b>OR-40. BM41</b> Peat, 150–194 cm deep.	2000 ± 50	

#### Sumatra

#### **Pulan Kijang Series**

Samples from swamp in Jambi (0°45'S,103°15'E). Collected and submitted 1986 by H. Furukawa.

**OR-41. Pulan Kijang 4** Peat, 30–50 cm deep.  $760 \pm 65$ 

<b>OR-42. Pulan Kijang 4</b> Peat, 100–120 cm deep.	$1040 \pm 50$
<b>OR-43. Pulan Kijang 4</b> Peat, 180–200 cm deep.	1550 ± 60
Dendang Series	
Samples from swamp in Jambi (1°10'S, 103°50'E). Collected	d and submitted 1986 by H. Furukawa.
<b>OR-44. Dendang Unit II</b> Peat, 30–50 cm deep.	1120 ± 55
<b>OR-45. Dendang Unit ll</b> Peat, 100–120 cm deep.	1440 ± 55

**OR-46. SK19 (Pelita)** 220 ± 40 Peat, 30–50 cm deep, from swamp in Jambi (1°06'S, 104°04'E). Collected and submitted 1986 by

H. Furukawa.

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## KSU RADIOCARBON DATES II

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## INTRODUCTION

This date list mainly reports on samples processed from 1982 to 1991. Each sample is converted to methanol. Radiocarbon is measured in a Teflon<sup>™</sup> vial containing a mixture of 40 g of methanol and 50 cc of xylene, 1% Butyl PBD and 0.1% PBBO using low-background scintillation counters Aloka LSC-800, LB-I and LB-III. The background of LB-III is ca. 8 cpm and counting efficiency is ca. 70%. The recent <sup>14</sup>C standard used is 95% of NBS oxalic acid; 5568 yr is used as the half-life of <sup>14</sup>C.

For further detail on our measurement methods, see Yamada and Kobashigawa (1986).

## **GEOLOGIC SAMPLES**

JAPAN

Hokkaido

## Hokkaido University Environmental Science (HE) Series

Samples were submitted 1984 to 1986 by H. Yamamoto of the Laboratory of Fundamental Research, the Graduate School of Environmental Science, Hokkaido University.

KSU-905. HE-1 Mizukami 1  $12,300 \pm 430$ Charcoal from Mizukami, Koshimizu-cho, Abashirigun (43°47'34"N, 144°29'52. 8"E). Collected 1984 by T. Sone.

Comment (T.S.): Expected age: ca. 10,000 BP.

KSU-1248. HE-37 Mizukami 2 26,100 + 2000/-1600Charcoal, 1100 cm depth, in Higashi-Kayano scoria, Koshimizu-cho (43°47'34.1"N, 144°29' 52.8"E, 70 m asl). Collected 1984 by T. Sone.

Comment (T.S.): Expected age: ca. 20,000 BP.

#### KSU-906. HE-2 Kayano

28,200 +1700/-1400 Charcoal, 210 cm depth, from Kayano, Koshimizu-cho (43°50'44.5"N, 144°30'16.8"E). Collected 1984 by T. Sone.

Comment (T.S.): Expected age: ca. 40,000 BP.

## KSU-907. HE-3 Kamishari

30,300 +1400/-1200

Charcoal in Kamishari pumice flow-I, 260 cm depth, from Kamishari, Kiyosato-cho (43°50' 44.6"N, 144°34'6.8"E). Collected 1984 by T. Sone.

Comment (T.S.): Expected age: ca. 32,000 BP.

## KSU-908. HE-4 Nakafurano

 $5220 \pm 90$ 

Peat, 230 cm depth, from Nakafurano-cho, Sorachi-gun (43°22'5.4"N, 142°26'42.4"E). Collected 1984 by H. Daimaru.

Comment (H.D.): Expected age: 4000-5000 BP.

 $2140 \pm 70$ KSU-909. HE-5 Shintoku 1 Peat from Shintoku-cho, Kamikawa-gun (43°5'52"N, 142°44'10"E, 420 m asl). Collected 1984 by K. Yamamoto. Comment (K.Y.): Expected age: 2000-5000 BP.  $1480 \pm 110$ KSU-910. HE-6 Shintoku 2 Peat from Shintoku-cho (43°9'49"N, 142°48'33"E, 405 m asl). Collected 1984 by K. Yamamoto. Comment (K.Y.): Expected age: 2000-5000 BP.  $2140 \pm 35$ KSU-911. HE-7 Shintoku 3 Peat from Shintoku-cho (43°6'2"N, 142°46'30"E, 440 m asl). Collected 1984 by K. Yamamoto. Comment (K.Y.): Expected age: 5000-6000 BP.  $4620 \pm 40$ KSU-913. HE-9 Shintoku 4 Peat from Shintoku-cho (43°31'26.8"N, 142°49'6.8"E, 1635 m asl). Collected 1984 by N. Takahashi.  $140 \pm 50$ KSU-912. HE-8 Kamikawa Peat from Kamikawa-cho, Kamikawa-gun (43°32'14.8"N, 142°57'32.8"E, 1424 m asl). Collected 1983 by N. Takahashi.  $3800 \pm 40$ KSU-914. HE-10 Biei 1 Peat, 130 cm depth, from Biei-cho, Kamikawa-gun (43°36'3.2"N, 142°53'17"E, 1735 m asl). Collected 1984 by N. Takahashi.  $7540 \pm 70$ KSU-915. HE-11 Biei 2 Peat from same site as KSU-914. Collected 1984 by N. Takahashi. KSU-1036. HE-12 Konan 12,400 ± 80 Peat, 190 cm depth, from Konan, Kiyosato-cho, Shari-gun (43°48'7.7"N, 144°38'1.3"E, 147 m asl). Collected 1984 by T. Sone. Comment (T.S.): Expected age: ca. 12,000 BP. KSU-1090. HE-21 Sapporo 1  $3370 \pm 25$ Wood, 820 cm depth, from Sapporo City (43°4′27.6″N, 141°21′34.8″E, 14 m asl). Collected 1985 by H. Daimaru.  $5930 \pm 60$ KSU-1091. HE-22 Sapporo 2 Wood from same site as KSU-1090. KSU-1092. HE-23 Saporo 3 9840 ± 60 Wood, 140 cm depth, from same site as KSU-1090. KSU-1105. HE-25 Shintokugawa  $5910 \pm 60$ Peat from Shintokugawa-cho, Kabato-gun (43°34'58.3"N, 141°44'3.7"E, 100 m asl). Collected 1985 by K. Yamamoto. Comment (K.Y.): Expected age: 1500–12,000 BP. KSU-1106. HE-26 Hokuryu  $21,100 \pm 600$ Peat from Hokuryu-cho, Uryu-gun (43°42'46.6"N, 141°52'39.1"E, 70 m asl). Collected 1985 by K. Yamamoto.

Comment (K.Y.): Expected age: 20,000-30,000 BP.

KSU-1107. HE-27 Kamoi $11,800 \pm 800$ Ash under Toyozumi pumice fall, 295 cm depth, from Kamoi, Kiyosato-cho, Abashiri-gun $(43°48'27.2"N, 144°33'37.6"E, 42 m asl).$ Collected 1985 by T. Sone.
Comment (T.S.): Expected age: ca. 12,000 BP. Comment: gained ca. 1 g carbon from 2 kg ash.
KSU-1108. HE-28Izumikawa 14120 ± 40Humus from between Masyudake-e and Masyudake-f ash fall, 125 cm depth, at Izumikawa, Bek-kai-cho (43°24'37.2"N, 144°41'15.6"E, 140 m asl). Collected 1985 by T. Sone.
Comment (T.S.): dated to 4150 BP by hydrated layer of obsidian.
KSU-1109. HE-29Izumikawa 23100 ± 30Humus from between Komagatake-lava flow and Masyudake-e ash fall, 105 cm depth. Same siteas KSU-1108.
Comment (T.S.): Expected age: 3000 BP.
KSU-1110. HE-30 Kitaoka   25,200 +1300/-1000     Charcoal under Yoteizan. Ps1 pumice fall, 250 cm depth, from Kitaoka, Kyogoku (42°53'   27.6"N, 140°51'42.6"E, 300 m asl). Collected 1985 by T. Sone.
Comment (T.S.): Expected age: 12,000–15,000 BP.
KSU-1242. HE-31Uryunuma 14050 ± 60Peat from Uryunuma marshland, 225–240 cm depth, Uryu-cho (43°41'21"N, 141°36'34"E). Collected 1985 by T. Miyagi, Tohoku Gakuin University.
KSU-1243. HE-32   Uryunuma 2   6220 ± 50     Peat, 65–70 cm depth, from same site as KSU-1242.   6220 ± 50
KSU-1244. HE-33   Uryunuma 3   9580 ± 80     Peat, 105–110 cm depth, from same site as KSU-1242.   9580 ± 80
KSU-1245. HE-34   Uryunuma 4   9910 ± 70     Peat, 112–118 cm depth, from same site as KSU-1242.   9910 ± 70
KSU-1246. HE-35   Horokayantou-numa 1   760 ± 90     Peat, 80 cm depth, from Horokayantou-numa, Taiki-cho, Hiroo-gun (42°31'42.9"N, 143°27'   16.7"E, 8 m asl). Collected 1985 by K. Kanzawa.
KSU-1247. HE-36 Horokayantou-numa 22560 ± 60Peat, 170 cm depth, from same site as KSU-1246.
KSU-1249. HE-38 Kami-nishishunbetsu10,800 ± 230Charcoal underlying Masyu-1 pumice fall, 200 cm depth, from Kami-nishishunbetsu, Bekkai-cho,Notsuke-gun (43°25'24.5"N, 144°47'15.6"E). Collected 1985 by T. Sone.
Comment (T.S.): Expected age: 10,000–13,000 BP.
KSU-916. Uryunuma9500 ± 140Peat, 310–325 cm depth, from Uryunuma marshland, Uryu-cho (43°41'21"N, 141°36'34"E). Collected and submitted 1984 by T. Miyagi.

Comment (T.M.): Result agrees with expected age based on pollen analysis.

#### **Kitahara Series**

Peat from Kitahara basin, Wassamu-cho (44°2'1"N, 142°22'2"E). Collected and submitted 1984 by T. Miyagi.

KSU-917. 375–390 cm depth	6800 ± 80
KSU-918. 600 cm depth	31,500 +810/-740

Comment (T.M.): Result agrees with expected age based on pollen analysis.

KSU-1051. Hassamu 60,300 +15,000/-4900 Peat, 27.0-27.1 m depth, from Shinkotoni-cho, Kita-ku, Sapporo City (43°5'N, 141°18'E, 3.5 m asl). Collected and submitted 1985 by Y. Igarashi, Hokkaido University.

Comment (Y.I.): Expected age: 20,000-30,000 BP.

#### **Nakahurano Series**

Peat from Nakahurano-cho, Sorachi-gun (43°20'12"N, 142°25'30"E, 99.5 m above sea level). Collected and submitted by Y. Igarashi.

Comment (Y. I.): Expected period: from last Ice age to early Holocene.

KSU-1417. 5.0–5.2 m depth	8120 ± 140
KSU-1418. 7.05–7.08 m depth	10,200 + 470
KSU-1078. 8.05–8.10 m depth	$12,400 \pm 120$
KSU-1419. 12.05–12.20 m depth	28,500 +810/-740
KSU-1079. 16.28–16.35 m depth	32,400 +1400/-1200
KSU-1080. 20.0–20.05 m depth	47,100

#### **Karumai Series**

Peat upper Akan ash fall-1, from Karumai, Atsuma-cho, Yufutsu-gun (42°39'47"N, 141°54'0"E). Collected 1985 by N. Wada, Geological Survey of Hokkaido, submitted 1985 by Y. Igarashi.

KSU-1081. Karumai-1	26,800 +1900/-1500
KSU-1082. Karumai-2	38,800 +1200/-900
KSU-1083. Hongo	58,700
Wood, upper Akan ash fall-2, Hongo, Atsuma-cho (	(42°44'0"N, 141°51'51"E). Collected 1985 by
N. Wada, submitted by Y. Igarashi.	

#### KSU-1084. Abira

56,100 +9000/-4100

Peat in Shikotsu pumice fall-6 and Shikotsu pumice fall-7, from Nakaabira, Oiwake-cho, Yufutsugun (42°50'6"N, 141°50'0"E). Collected 1985 N. Wada and submitted 1985 Y. Igarashi.

Comment (Y.I.): Samples from upper Shikotu pumice fall-1 were dated to 32,200 + 4700/-3100, Gak-519, and  $32,200 \pm 2000$ , Gak-714 (Kigoshi 1967: 48).

## **Nemuro Series**

Peat from Habomai-cho, Nemuro City (43°21'20"N, 145°45'5"E, 30 m asl). Collected and submitted 1985 by Y. Igarashi.

KSU-1189.	68–73 cm depth	2030 ± 40
KSU-1190.	155–160 cm depth	10,000 ± 140

## **Fujino Series**

Samples from Fujino, Minami-ku, Sapporo City (42°57.8'N, 141°19'E, 170 cm asl). Collected and submitted 1984 by J. Ishii, Tokai University.

KSU-1000. Fujino A Charcoal in Shikotsu pumice flow deposit.	40,700 +1400/-1200
KSU-1001. Fujino B Carbonized root in same layer as KSU-1000.	40,800 +1400/-1200
KSU-1002. Fujino C Charcoal in peat layer.	36,500 +2000/-1600
KSU-1003. Fujino D Peat from same horizon as KSU-1002.	39,500 +1400/-1200

#### **Komoro Series**

Radiocarbon dates of these samples show the history of activity in Asama Volcano and landslides in Nagano prefecture. The traces of an old large landslide were found by A. Nakagawa and K. Higuchi, 1986 on the landslides in Komoro district (Annuals of the Disaster Prevention Research Institute, Kyoto University, No. 29B-1, April 1986.) Samples were collected in these tracks. Collected and submitted 1984-1991 by K. Higuchi, Komoro High School. Some pieces of Early Jomon pottery were found under the layer (KSU-1260, KSU-1262, KSU-1264, KSU-1302, KSU-1303, KSU-1493, KSU-1496, KSU-1497, KSU-1498, KSU-1516) (T. Hayata 1992). This pottery is the oldest yet found in Japan (N. Kondou, H. Kobayashi, 1990).

KSU-839. Komoro 1 Wood ( <i>Picea</i> ) from Oshidashi, Komoro city, Nagano prefecture (36°19'4	<b>52,100</b> 10"N, 138°23'27"E).
<b>KSU-840. Komoro 2</b> Charcoal ( <i>Picea, Larix</i> ) from Oshidashi, (36°19'48"N, 138°23'33"E).	56,900
<b>KSU-852. Komoro 3</b> Wood from Oshidashi (36°19′54″N, 138°23′42″E).	49,300
KSU-853. Komoro 4 Charcoal from same site as KSU-852.	49,300 +18,000/-5100
KSU-854. Komoro 5 Charcoal from same site as KSU-852.	54,300 +24,000/-5300
<b>KSU-874. Komoro 6</b> Wood from Kubo, Komoro city (36°18'36"N, 138°25'21"E).	49,100 +6300/-3500
KSU-875. Komoro 7 Charcoal from Hakeyama, Kitamimaki village, Kitasaku-gun, Nagano 138°23'42"E).	<b>55,400</b> prefecture (36°19'54"N,
KSU-877. Komoro 8 Wood from Oshidashi (36°20'N, 138°23'E).	51,900 +20,000/-5000
<b>KSU-1121. Komoro 9</b> Wood from Okubo, Komoro city (36°19'31"N, 138°24'29"E).	44,600 +3000/-2200

KSU-1122. Komoro 10 Charcoal from Hakeyama (36°20'32"N, 138°20'35"E).	40,800 +970/-870
KSU-1123. Komoro 11 Wood from Goushi-gawara, Kitamimaki village (36°20'39"N, 138°19'39"H	<b>54,300</b> E).
KSU-1124. Komoro 12 Charcoal from Okui, Komoro city (36°17'55"N, 138°25'28"E).	46,100 +3700/-2500
KSU-1125. Komoro 13 Charcoal from same place as KSU-1124.	54,000 +5900/-3400
KSU-1126. Komoro 14 Wood from Okubo (36°19'40"N, 138°23'58"E).	44,600 +1500/-1300
KSU-1260. Komoro 15 Charcoal in first pumice flow deposit of Asama Volcano, from Namezu, S fecture (36°14'21"N, 138°28'46"E).	<b>13,600 ± 60</b> Saku city, Nagano pre-
Comment (K.H.): Asama first pumice flow was dated to 13,500 ± 500, Ju JGS-36; 13,600 ± 400, JGS-37; 13,600 ± 400, JGS-40 (Togashi 1984: 207-20	GS-16; 13,700 ± 400, 08).
KSU-1261. Komoro 16 Wood from Shionada, Asashina village, Kitasaku-gun (36°15'38″N, 138°2.	<b>38,300 +1000/-900</b> 5′02″E).
Comment (K.H.): Collected in presumed Tsukahara mud-flow deposit.	
KSU-1262. Komoro 17 Wood from Nenei, Saku City (36°15'30"N, 138°27'12"E).	13,700 ± 60
KSU-1263. Komoro 18 Wood in presumed Tsukahara mud-flow deposit, 5 m under KSU-1262.	21,250 ± 140
KSU-1264. Komoro 19 Wood from Mimitori, Komoro City (36°17′18″N, 138°25′38″E).	13,600 ± 70
KSU-1297. Komoro 20 Charcoal from a cliff beside Chikuma River, Mimitori, Komoro City (36°16	<b>54,200</b> 5′44″N, 138°25′22″E).
KSU-1298. Komoro 21 Charcoal from near KSU-1297, Mimitori, Komoro City (36°16'44"N, 138°	<b>57,000</b> 25′25″E).
<b>KSU-1299. Komoro 22</b> Charcoal from Kubo, Komoro City (36°18'17"N, 138°25'14"E).	54,000
KSU-1300. Komoro 23 Charcoal from same place as KSU-1299.	52,800
KSU-1301. Komoro 24 Charcoal from same place as KSU-1299.	51,100 +8100/-4000
<b>KSU-1302. Komoro 25</b> Wood from same layer of KSU-1262, Komoro City (36°19'08"N, 138°25'1	<b>13,700 ± 60</b> .6″E).
KSU-1303. Komoro 26 Charcoal from Chikuma riverside, Mimitori, Komoro City (36°17'16"N, 13	<b>14,000 ± 60</b> 38°25′01″E).

x

KSU-1328. Komoro 27 Charcoal from Chikuma riverside, Okui, Komoro City (36°17'46"N, 138°	<b>57,300</b> 24'58"E).
KSU-1329. Komoro 28 Charcoal from same place as KSU-1328.	57,900
<b>KSU-1379. Komoro 30</b> Charcoal from Kubo, Komoro City (36°18′34″N, 138°25′16″E).	53,000 +8700/-4100
<b>KSU-1380. Komoro 31</b> Charcoal from Kubo, Komoro City (36°18′34″N, 138°25′34″E).	48,300 +3400/-2600
<b>KSU-1381. Komoro 32</b> Charcoal from Kubo, Komoro City (36°18′02″N, 138°25′00″E).	60,000
KSU-1493. Komoro 33 Charcoal from Amaike, Komoro City (36°21′16″N, 138°27′23″E).	13,400 ± 70
KSU-1494. Komoro 34	800 + 20
Wood on a mud-flow deposit, Ohata, Komoro City (36°19'42"N, 138°26 same age as an eruption of Asama Volcano in AD 1108.	6'54"E). Datum shows
KSU-1495. Komoro 35 Charcoal from same site as KSU-1263.	23,700 ± 290
KSU-1496. Komoro 36 Charcoal from same site as KSU-1303.	13,500 ± 60
KSU-1497. Komoro 37 Charcoal from 400 m depth, Mimitori, Komoro City (36°17'20"N, 138°25	<b>13,300 ± 50</b> ''19"E).
KSU-1498. Komoro 38 Wood from Hishino, Komoro City (36°21′06″N, 138°26′17″E).	13,600 ± 100
KSU-1499. Komoro 39 Charcoal from Hebihori riverside, Komoro City (36°19'47"N, 138°26'34"] age as an eruption of Asama Volcano in AD 1108.	<b>860 ± 20</b> E). Datum shows same
KSU-1516. Komoro 40 Charcoal from upper part of a bed rock, Yugawa riverside, Saku City (36°1'	<b>13,800 ± 60</b> 7'26"N, 138°30'47"E).
KSU-1517. Komoro 41 Charcoal from Mitsuishi, Karuizawa village (36°20'38"N, 138°31'40"E). I as an eruption of Asama Volcano in AD 1281.	700 ± 20 Datum shows same age
KSU-1518. Komoro 42 Charcoal from 120 cm depth, 1225 m asl, Karuizawa village (36°20'38"N, shows same age as an eruption of Asama Volcano in AD 1281.	<b>700 ± 20</b> 138°31'40″E). Datum
KSU-1519. Komoro 43 Charcoal from Namezu riverside, Saku City (36°14'43"N, 138°27'50"E).	$22,500 \pm 40$
KSU-1583. Komoro 44 Charcoal in the pumice flow 2 of Asama Volcano, 100 cm depth, K (36°18'48"N, 138°25'32"E).	<b>11,400 ± 130</b> Kohara, Komoro City

KSU-1584. Komoro 45 Charcoal under a pumice flow, Kohara, Komoro City (36°18'51"N, 138°25'3'	<b>11,800 ± 400</b> 7″E).
KSU-1585. Komoro 46 39,1 Wood in volcanic gravel layer in a deep well, 141 m depth, Amaike, Komoro 138°28'03"E).	100 +14,000/-4800 City (36°21'30"N,
KSU-1586. Komoro 47 Charcoal from same site as KSU-1517.	850 ± 40
KSU-1587. Komoro 48 Charcoal under the Oiwake pyroclastic flow, 14 m depth in a deep well, (36°21'08"N, 138°32'01"E).	1010 ± 210 Karuizawa village
41 Charcoal from Saikouji riverside, Oshidashi, Komoro City (36°21'30"N, 138	<b>l,800 +2000/–1600</b> °28′03″E).
KSU-1735. Komoro 51 Charcoal from Karuizawa village (36°20'51"N, 138°31'45"E).	1760 ± 80
KSU-1736. Komoro 53 Charcoal in volcanic ash at the foot of Sekison Mountain, 200 cm depth, (36°22'07"N, 138°31'46"E).	<b>1180 ± 12</b> Karuizawa village
KSU-1762. Komoro 54 Charcoal from Karuizawa village (36°29'40"N, 138°31'18"E).	780 ± 15
KSU-1763. Komoro 55 Charcoal from Karuizawa village (36°27'58"N, 138°30'53"E). Datum shows s tion of Asama Volcano in AD 1281.	710 ± 20 ame age as an erup-
KSU-1764. Komoro 56 Charcoal from Karuizawa village (36°28'05"N, 138°34'22"E).	100 ± 10
KSU-1765. Komoro 57 Charcoal from Karuizawa village (36°21′03″N, 138°27′02″E).	13,190 ± 40
KSU-2061. Komoro 58 Charcoal from Shiozawa, Karuizawa village (36°19'09"N, 138°34'45"E).	13,480 ± 50
KSU-2127. Komoro 59 Charcoal under the Aira volcanic ash, Nakasato elementary school, Saku 138°27'02"E).	<b>23,400 ± 300</b> city (36°16′00″N,
KSU-2128. Komoro 60 Wood (Susuki) from 350 cm depth, Asamadai, Karuizawa village (36°20'49")	<b>0 ± 140</b> N, 138° 33′26″E).
Comment (K.H.): It seems that there is a possibility of an eruption of Asama V	/olcano in AD 1800.
Myoko Series	

Samples were collected at various places around the Myoko Volcano. Submitted 1982 by S. Nohda, Kyoto Sangyo University.

KSU-518. MK-797-3 Suginosawa 14080 ± 4Wood in Suginosawa formation, 2.5 m depth, from Suginosawa, Myoko-kogen-cho, Niigata profecture (36°51'N, 138°10'E). Collected 1980 by K. Hayatsu.
KSU-1103. MK-797-4 Suginosawa 2460 ± 8Charcoal in peat layer upper Suginosawa formation, 0.7 m depth, from Suginosawa (36°51'N138°10'E). Collected 1980 by K. Hayatsu.
KSU-519. MK-801-1 Taguchi 7880 ± 3 Wood upper Taguchi formation, 5 m depth, from Shin-akakura, Myoko-kogen-cho (36°53'N 138°12'E). Collected 1980 by K. Hayatsu.
KSU-1098. MK-802-3 Fukazawa17,200 ± 7Charcoal in Fukazawa formation from same place as KSU-519. Collected 1980 by S. Nohda.
KSU-538. MK-802-1 Matsugamine 0 ± 5 Wood in Matsugamine volcanic conglomerate, from Matsugamine, Nakagou village, Niigata pro fecture (36°58'N, 138°14'E). Collected 1980 by S. Nohda.
KSU-1099. MK-802-10 Furuma 139,100 +1200/-100Peat between Kan-noki and Sekiyama scoria, 4 m depth, from Furuma Shinano-cho, Nagano prefecture (36°48'N, 138°12'E). Collected 1979 by K. Hayatsu.
KSU-1100. MK-802-14 Furuma 229,700 ± 60Peat above Ohira scoria, 6 m depth, from Furuma (36°47'N, 138°12'E). Collected 1979 by FHayatsu.
KSU-1101. MK-802-17 Kannoki23,800 +2700/-200Sand and charcoal in Mutsuki volcanic conglomerate, from Kan-noki, Shinano-cho (36°49'N138°12'E). Collected 1980 by S. Nohda.
KSU-1102. MK-601-2 Sekiyama4110 ± 4Peat in Otagirigawa formation, 1 m depth, from Sekiyama, Myoko village, Niigata prefectur(36°56'N, 138°12'E). Collected 1972 by K. Hayatsu.
KSU-536. YK-50-1 Sasakura 890 ± 3 Wood from bottom of Hayakawa formation, 2 m depth, from Sasakura-onsen, Itoigawa city, Ni gata prefecture (36°59'N, 138°1'E). Collected 1971 by K. Hayatsu.
Okamura Fault Series
Samples were collected by the trench excavation survey across the Okamura fault belonging to the Median Tectonic Line active fault system of Southwest Japan. Samples from Ioka, Saijo city, Ehim prefecture (33°55'N, 133°13'E). Collected 1983 and submitted 1984 by M. Andou, Kyoto University, and A. Okada, Aichi Prefectural University.
KSU-791. NW 30a 2050 ± 6

KSU-791. NW 30a	$2050 \pm 60$
Peat. x (level distance) = $8.0-8.5$ m and $z = 2$ m in northwest wall.	
KSU-789. NW 30b	$2030 \pm 50$
Peat from under part of peat layer, $x = 8.5$ m and $z = 2$ m.	
KSU-786. NW 31	$11,100 \pm 260$
Peat from peat and fine sand layer, $x = 8.5$ m and $z = 4.6$ m.	

<b>KSU-790.</b> NW 32 Peat from upper part of thick peaty silt-sand layer, $x = 6.5$ m and $z = 5.3$ m.	11,200 ± 200
<b>KSU-787.</b> NW 33 Peat from under part of thick peaty silt layer, $x = 6.5$ m and $z = 5.3$ m.	12,000 ± 80
<b>KSU-782.</b> NW 34 Soil from peaty sand layer, $x = 6.5$ m and $z = 5.7$ m.	13,100 ± 120
<b>KSU-784.</b> NW 35 Soil from under peaty silt layer, $x = 6.5$ m, and $z = 6.7$ m.	17,800 ± 370
<b>KSU-785.</b> SW 39 Peat, $x = 9.2$ m and $z = 3.2-3.3$ m in southwest wall.	3870 ± 90
<b>KSU-788.</b> SW 40 Wood and soil from conglomerate layer, $x = 12.2$ m and $z = 3.9$ m.	2900 ± 60
<b>KSU-783. SW 42</b> Soil from fine sand layer, $x = 10.2$ m and $z = 6.5-6.6$ m.	13,200 ± 380
<b>KSU-793. SW 43</b> Soil from upper thick beach silt layer, $x = 10.2$ m and $z = 7.0-7.05$ m.	9990 ± 80
<b>KSU-792. SW 44</b> Soil from under thick beach silt layer, $x = 10.2$ m and $z = 7.8$ m.	10,300 ± 120

#### **Kochi Bore Samples**

Peat from Kochi plain, Itachino, Kochi prefecture (33°33'30"N, 133°37'50"E). Collected and submitted by M. Ando.

KSU-1067. 105–110 cm depth	1740 ± 70
KSU-1068. 240–245 cm depth	4850 ± 90
KSU-1069. 460–465 cm depth	8040 ± 60
KSU-1070. 670–675 cm depth	$29,100 \pm 1000$
KSU-1071. 910–915 cm depth	30,200 +1500/-1200
KSU-1072. 1140–1145 cm depth	46,500 +38,000/-5500
KSU-1073. 1795–1800 cm depth	26,600 +1300/-1100

General comment: KSU-1073 was the same layer KSU-1070 and KSU-1071. KSU-1073 fell by a fault.

## Hamana Lake Series

Peat samples were collected by boring, at the shore of Hamana Lake, Shizuoka prefecture (34°44'N, 137°38'E). Collected and submitted 1984 by H. Wada, Shizuoka University.

KSU-845. H7 240–245 cm depth	5390 ± 420
KSU-846. H7 245–250 cm depth	7480 ± 900
KSU-847. H9 230–235 cm depth	4850 ± 370
KSU-848. H9 235–240 cm depth	5290 ± 640
KSU-849. H10 250–255 cm depth	4490 ± 340
KSU-850. H10 255–260 cm depth	6700 ± 1400

## Jindai-sugi Series

Samples in Kawagodaira pumice, from Ikadaba, Nakaizu-cho, Shizuoka prefecture (34°54'N, 138°57'E). Collected 1982 by E. Okawa, submitted 1984 by H. Wada.

Comment (H.W.): Kawagodaira pumice was believed to have erupted ca. 3000 BP.

KSU-952. Ikadaba 0-5 Wood (Cryptmeria japonica). 5 tree rings from outer side.	2820 ± 50
KSU-953. Ikadaba S	2860 ± 50
Bark of same wood as KSU-952.	

#### **Kagiana Series**

Wood samples were embedded in sandy and clayish sediments, from Kagiana, Shizuoka City (35°2'N, 138°15'E). Collected and submitted 1985 by H. Wada.

KSU-1032. KG-1 Bark and 20 tree rings from outer side.	2490 ± 25
KSU-1033. KG-2 15 tree rings from outer side.	2500 ± 30
KSU-1034. KG-3a Bark of tree.	$2670 \pm 30$
KSU-1035. KG-3b	$2430 \pm 50$

15 tree rings from outer side.

#### KSU-954. Odanoike

## $4740 \pm 230$

 $4200 \pm 170$ 

 $890 \pm 70$ 

Peat, 290–300 cm depth, from Odanoike pond, Kuju-cho, Kusu-gun, Ohita prefecture (33°12'N, 131°18'E). Collected and submitted 1984 by M. Takeoka, Kyoto Prefectural University.

## KSU-955. Nonbara

Peat, 400–410 cm depth, from Ohike pond, Nonbara, Takeno-gun, Kyoto prefecture (35°45'N, 135°8'E). Collected and submitted 1984 by M. Takeoka.

#### KSU-956. Chojidani

Peat, 120–130. cm depth, from Chojidani valey, Miyama-cho, Kitakuwata-gun, Kyoto prefecture (35°17'N, 135°46'E). Collected and submitted 1984 by M. Takeoka.

## KSU-957. Sugiyaike

Peat, 140–150 cm depth, from Sugiyaike pond, Ohtsu city, Shiga prefecture (35°11'N, 135°53'E). Collected and submitted 1984 by M. Takeoka.

## KSU-958. Kojorogaike

#### $4600 \pm 110$

 $4120 \pm 120$ 

Peat, 200–210 cm depth, from Kojorogaike pond, Katsuragawa, Otsu City, Shiga prefecture (35°14'N, 135°53'E). Collected and submitted 1984 by M. Takeoka.

## KSU-959. Meiji

Peat, 240–250 cm depth, from Meiji, Miyama, Tango-cho, Takeno-gun, Kyoto prefecture (35°44'N, 135°10'E). Collected and submitted 1984 by M. Takeoka.

## 60 ± 90

#### KSU-1041. Habikino

 $12,770 \pm 130$ Peat, 120-130 cm depth, from Konda, Habikino City, Osaka prefecture (34°33'N, 135°15'E). Collected and submitted 1985 by M. Takeoka.

## KSU-1042. Kashiwara

Peat, 80-85 cm depth, from Kashiwara, Toyono-gun, Osaka prefecture (34°58'N, 135°25'E). Collected and submitted 1985 by M. Takeoka.

### KSU-985. Kamifukada

Wood, 60-90 cm depth, from Kamifukada, Sanda City, Hyogo prefecture (35°53'N, 135°12'E). Collected and submitted 1984 by K. Mino, Ritsumeikan University.

Comment (K.M.): Collected in Yamasaki fault. Dated speciment was presumed to be buried by earthquake of AD 868.

#### KSU-1149. Natadera

Charcoal in Natadera fault, Komatsu City, Ishikawa prefecture (36°18'N, 136°25'E). Collected and submitted 1985 by K. Mino.

#### **Yachidaira** Series

Peat from Yachidaira, Onoda-cho, Kami-gun, Miyagi prefecture (38°29'N, 140°37'30"E). Collected 1984 and submitted 1985 by K. Hibino, Miyagi Agricultural College.

KSU-1074. Yachidaira 1 Peat, 120–130 cm depth.	830 ± 40
KSU-1075. Yachidaira 2 Peat, 85–100 cm depth.	640 ± 30

KSU-1085. Miyatoko-Ohyachi

Peat, 275-300 cm depth, from Miyatoku-Oyachi, Nagano, Minami-aizu-gun, Fukushima prefecture (37°15'N, 139°34'E). Collected 1984 and submitted 1985 by K. Hibino.

Comment (K.H.): This <sup>14</sup>C date age almost agrees with a palynological estimate.

KSU-1086. Amou marshland 9680 ± 70 Peat, 275-295 cm depth, from Amou, Kawai, Kichijo-gun, Gifu prefecture (36°15'N, 136°58'E). Collected 1984 and submitted 1985 by K. Hibino.

#### **Yadegawa Series**

Samples from Yadegawa, Nobeyama, Minamisaku-gun, Nagano-prefecture (36°51'N, 138°29'E). Collected by Y. Yasuda, Hiroshima University. Submitted 1981 by M. Tozawa, Meiji University.

Comment (Y.Y.): KSU-1160 and KSU-443 were presumably from the last Ice Age (ca. 20,000 to 11,000 BP). KSU-1161 and KSU-1162 were expected to be of Jomon Age.

KSU-1163. Yadegawa 1 Peat.	22,500 ± 160
KSU-443. Yadegawa 3 Wood.	29,750 ± 600
KSU-1161. Yadegawa 4 Peat.	2940 ± 20

## $2330 \pm 30$

 $3340 \pm 40$ 

 $7000 \pm 50$ 

54,600 +12,000/-4600

## **KSU-1162. Yadegawa 6** Peat.

#### KSU-649. Hananoego

Peat under Akahoya volcanic ash, Hananoego, Yaku Island, Kumage-gun, Kagoshima prefecture (30°18'30"N, 130°30'45"E). Collected and submitted 1983 by Y. Yasuda. *Comment* (Y.Y.): Expected age: *ca.* 7000 BP.

#### CHINA

# KSU-1163. Saiko 200–250 cm depth1540 $\pm$ 60Peat from Lake Sha, Hong-chou (30°15'N, 120°10'E). Collected 1984 and submitted 1985 by Y.Yasuda.

KSU-1166. Mongol 140–160 cm depth $3340 \pm 140$ Peat from Shioziki, Inner Mongolia (40°25'N, 111°10'E). Collected 1984 and submitted 1985 byY. Yasuda.

#### GREECE

#### **Hotousa Moor Series**

Peat from Peloponissos (37°48'N, 22°30'W, 900 m asl). Collected 1984 and submitted 1985 by Y. Yasuda.

KSU-1111.	100–200 cm depth	$2280 \pm 40$
KSU-1112.	180–200 cm depth	3700 ± 70
KSU-1113.	260–280 cm depth	5180 ± 70

## **Korone Moor Series**

Peat from Korone moor, northwest Greece (39°20'N, 20°11'W, 10 m asl). Collected 1984 and submitted 1985 by Y. Yasuda.

KSU-1252.	150–160 cm depth	$2740 \pm 40$
KSU-1114.	380–390 cm depth	$4500 \pm 40$
KSU-1253.	1400–1420 cm depth	<b>3940 ± 80</b>
KSU-1115.	1610–1630 cm depth	$6360 \pm 40$

#### **Katouna Series**

Peat from Katouna moor, northwest Greece (38°50'N, 21°5'W). Collected 1984 and submitted 1985 by Y. Yasuda.

KSU-1170. 170–200 cm depth	710 ± 50
KSU-1171. 370-400 cm depth	1770 ± 50
KSU-1172. 910–940 cm depth	3970 ± 80

#### TURKEY

#### **Civiril Series**

Peat from Civiril moor, western Anatolia (38°20'N, 29°40'W). Collected 1984 and submitted 1985 by Y. Yasuda.

#### 4730 ± 25

 $6100 \pm 25$ 

KSU-1164.	100–175 cm depth	$2700 \pm 70$
KSU-1165.	305–330 cm depth	4140 ± 80

#### **Abant Gol Series**

Peat from the lake Abant, northern Turkey (40°50'N, 31°30'W). Collected 1984 and submitted 1985 by Y. Yasuda.

KSU-1167. 100–120 cm depth	350 ± 80
KSU-1168. 320-340 cm depth	1410 ± 180
KSU-1169. 620–650 cm depth	2400 ± 70

NEPAL

KSU-648. Kathmandu basin 9950  $\pm$  70 Wood from Kathmandu (27°40'N, 85°20'E, 1400 m asl). Collected 1982 and submitted 1983 by Y. Yasuda.

Comment (Y.Y.): Wood was embedded in lake deposit. Expected time range: Last Ice Age.

KSU-657. Lake Rara

7720 ± 200

Peat, 480–450 cm depth, from Lake Rara, western Nepal (29°27'N, 82°0'E, 3000 m asl). Collected 1982 and submitted 1983 by Y. Yasuda.

Comment (Y.Y.): deposit of 7 m depth was dated to ca. 10,000 BP by pollen analysis. Therefore this deposit was presumed ca. 7000 BP.

#### **Partical Series**

Samples from mud-flow deposits in Indrawati River, northeast of Kathmandu basin (27°44'N, 85°40'E). Collected 1985 by M. Yoshida, Trivhuvan University, and submitted 1985 by Y. Igarashi, Hokkaido University.

Comment (M.Y.): Expected period: Last Ice Age.

<b>KSU-1087. 84Y 18-1</b> Peat.	55,600 +29,000/-5400
KSU-1088. 84Y 18-2 Wood.	56,200
KSU-1120. Thimi-1	45,300 +6000/-3400

Wood in Thimi Formation, from Kathmandu basin (27°44'N, 85°23'30"E). Collected 1985 by M. Yoshida, submitted 1985 by Y. Igarashi.

#### INDONESIA

## KSU-502. Krakatau Volcano

Wood in the outcrop of ash flow at Danan Islands, west Java (6°20'S, 105°30'E). Collected and submitted 1982 by T. Yokohama, Doshisha University.

Comment (T.Y.): Presumed to be related to eruption of Krakatau Volcano in AD 1883.

#### KSU-768. Ug. Kulon

## Shell (*Tridacnidae*) in coral reef, 20 m above sea level, from Ug. Kulom, Java Island (6°44N, 105°13'E). Collected 1983 by P. Hebanssa, Bandung Giological Laboratory, and submitted 1984 by J. Nishida, Ohtani University.

#### $4810 \pm 40$

 $190 \pm 25$ 

#### **ARCHAEOLOGICAL SAMPLES**

JAPAN

#### Hokkaido Mamachi Site Series

Charcoal from Mamachi, Chitose City (42°48'58"N, 141°38'54"E). Collected 1982 by H. Taguchi and submitted by R. Asai, Hokkaido Archaeological Research Center.

Comment (H.T.): Associated with Satsumon pottery.

KSU-560. Mamachi 1 Charcoal from B point, on floor of Dwelling Pit H-1 of Satsumon period.	1930 ± 40
KSU-561. Mamachi 2 Charcoal from fireplace of same pit as KSU-560.	$1210 \pm 40$

#### **Horikapp Site Series**

Charcoal from Horikapp, Furuu-gun (43°0'N, 140°30'E). Collected 1982 by N. Kimura, submitted by R. Asai.

Comment (N.K.): Samples found in layer, included relics of later Middle to early Late Jomon Age.

KSU-574. Horikapp 1 Charcoal under layer 4, F-8.	4020 ± 50
KSU-588. Horikapp 2 Charcoal upper layer 2, D-9-30, P-8.	2330 ± 170
KSU-589. Horikapp 3 Charcoal from fireplace 2, E point.	2380 ± 500
KSU-590. Horikapp 4 Charcoal from F-12-87, fireplace 14.	3800 ± 120
KSU-591. Horikapp 5 Charcoal from H-10, upper black sand layer.	2550 ± 450

KSU-967. Bibi 2 site 2430 ± 90 Charcoal on floor of Dwelling Pit H-1, upper Tarumae-c scoria. Collected 1984 by B. Aoyanagi and submitted 1984 by R. Nakamura, Hokkaido Archaeological Research Center. Charcoal from Bibi, Chitose City (42°46'N, 141°4'E).

Comment (B.A.) Expected age: later Jomon to Zoku-Jomon Age.

#### **Bibi 4 Site Series**

This site is near place of Bibi 2 site. Submitted 1983 to 1985 by R. Nakamura.

KSU-676. Bibi 4-1 2370 ± 30 Charcoal in buried soil of grave, excavated from upper Tarumae-c volcanic ash layer, P-59. Collected 1983 by K. Nonaka.

Comment (K.N.): Associated with Daido-A pottery.

KSU-677. Bibi 4-2 Charcoal on floor of Dwelling Pit H-1 in peat layer between Tarumae-d and Tarum 1983 by K. Satou.	<b>3420 ± 30</b> ae-c. Collected
Comment (K.S.): Associated with Teine-shiki pottery of middle Late Jomon Age.	
KSU-747. Bibi 4-3 Human bone in grave X-208. Collected 1983 by K. Nonaka.	2640 ± 400
Comment (K.N.): Grave was presumed later Jomon Age.	
KSU-969. Bibi 4-4 Charcoal from Pit-149, on grave. Collected 1984 by K. Endo.	3240 ± 130
Comment (K.E.): Expected age: middle Late Jomon Age.	
KSU-970. Bibi 4-5 Charcoal from Pit-123. Collected 1984 by H. Mori.	3460 ± 40
Comment (H.M.): Presumed same age as KSU-969.	
KSU-1194. Bibi 4-6 Charcoal from Pit-904 under Tarumae-c. Collected 1985 by K. Nonaka.	3180 ± 80
Yunosato 2 Site Series	
Samples were collected 1983 by T. Takahashi, and submitted 1983 by R. Nakamus Shiriuchi-cho, Kamiiso-gun (41°35'N, 140°20'E).	ra at Yunosato,
KSU-748. Yunosato 2-1 Charcoal from Y2-H-1, Dwelling Pit-1, later Early Jomon Age.	4530 ± 120
KSU-749. Yunosato 2-2 Charcoal from Y2-H-2, Dwelling Pit-2, later Early Jomon Age.	4380 ± 90
Yunosato 5 Site Series	
Samples were collected 1983 by H. Taguchi and submitted 1983 by R. Nakamura.	
KSU-678. Yunosato 5-1 Charcoal in Layer 3 on Stone Circle of early Late Jomon Age.	3020 ± 80
KSU-679. Yunosato 5-2 Charcoal in covered soil of Stone Circle.	3590 ± 70
<b>KSU-680.</b> Yunosato 5-3 Charcoal in pit, presumed same age as Stone Circle.	3970 ± 90
Yunosato 6 Site Series	
Samples were collected 1983 by Y. Nakata and submitted 1983 by R. Nakamura.	
KSU-681. Yunosato 6-1 Charcoal from Y-6, G-64-b, Layer 3, contained Final Jomon Relics.	2510 ± 180
KSU-682. Yunosato 6-2 Charcoal from Y-6, I-63-d, Pit 5, presumed grave of Final Jomon Age.	2780 ± 180

KSU-683. Yunosato 6-3 Charcoal from Y-6, E-65-a, S-1, Layer 4, fireplace of Late Jomon Age.	3380 ± 640
Kusunoki Site Series	
Charcoal from Kusunoki, Bifune-cho, Nakagawa-gun (44°28'N, 142°15'E). Colle Oniyanagi and submitted 1983 by R. Nakamura.	cted 1983 by A.
Comment (A.O.): Expected age: Satsumon Age.	
KSU-684. Kusunoki 1 Charcoal in kitchen range of Dwelling Pit H-6.	920 ± 70
KSU-685. Kusunoki 2 Charcoal in kitchen range of Dwelling Pit H-7.	760 ± 50
KSU-686. Kusunoki 3 Charcoal on floor of Dwelling Pit H-37.	820 ± 60
Pirika 1 Site Series	
Charcoal from Pirika, Imakane-cho, Setana-gun (42°28'N, 140°13'E). Collec Naganuma and submitted 1983 by R. Nakamura.	ted 1983 by T.
KSU-687. Pirika 1-1 Charcoal from M-44-c, bottom of Layer 3.	19,800 ± 380
Comment (T.N.): Expected age was comparable to microlith of ran-etsu type of Age.	later Early Stone
KSU-688. Pirika 1-2 Charcoal from L-52-d, middle of Layer 3.	17,500 ± 200
Comment (T.N.): Expected age was comparable to Yuzetsu point of later Early S	tone Age.
KSU-689. Pirika 1-3 Charcoal from M-53-b, bottom of Layer 3.	20,900 ± 260
<i>Comment</i> (T.N.): Expected age was comparable to grave of Araya type and micro ita type of later Early Stone Age.	olith of Tougesh-
KSU-750. Nishinopporo 12-17 site Charcoal from Pit 88, Nishinopporo, Ebetsu city (43°4'N, 141°32'E). Collected and submitted by R. Nakamura.	<b>1070 ± 20</b> 1983 by T. Kasai
Comment (T.K.): Expected period: late Zoku-Jomon Age.	
Nakahama E Site Series	
Charcoal from Nakahama, Shirikishinai-cho, Kameda-gun (41°44'N, 141°4'E). C H. Taguchi and submitted by R. Nakamura.	ollected 1984 by
Comment (H.T.): Expected age: Initial to Late Jomon Age.	
KSU-961. Nakahama E-1	4390 ± 40

Charcoal from P-1.

KSU-962. Nakahama E-2 Charcoal from TP-5.	1920 ± 190
KSU-963. Nakahama E-3 Charcoal on floor of Dwelling Pit H-2.	6820 ± 180

#### **Chitose 5 Site Series**

Charcoal from Chitose-cho, Noboribetsu City (42°24'N, 141°11'E). Collected 1984 by S. Nishida and submitted by R. Nakamura.

Comment (S.N.): Expected age: Middle to Late Jomon Age.

KSU-964. Chitose 5-1	$4210 \pm 60$
Charcoal on floor of Dwelling Pit, H-23-B.	
KSU-965. Chitose 5-2 Charcoal from bottom of Layer 3, E-100-d.	4110 ± 70
KSU-966. Chitose 5-3 Charcoal from Layer 2, E-1-c.	4200 ± 50

## **Osaka Prefecture Sadoh Site Series**

Wood from Sadoh-cho, Yao city (34°37′56″N, 135°35′32″E). Collected and submitted 1982, 1983 by Y. Nishiguchi, Osaka Archaeological Research Center.

KSU-521. Sadoh 1 Wood.	1390 ± 20
KSU-526. Sadoh 2 Wood.	1500 ± 20
KSU-575. Sadoh 3 Wooden stake from bank of Nagase River.	1360 ± 20
KSU-577. Sadoh 4 Wooden stake from bank of Nagase River.	1410 ± 20
KSU-716. Sadoh 5 Wooden stake from Tr. F.	250 ± 40

#### Yamaga Site Series

Sample from Yamaga-cho, Yao City (34°38'36"N, 135°35'58"E). Collected and submitted 1982 by Y. Nishiguchi, Osaka Archaeological Research Center.

KSU-524. Yamaga 1	2950 ± 20
Wood from rough sand layer, Tr C-6, 0 m, associated with Final Jomon potte	ery.
KSU-529. Yamaga 2	3750 ± 20
Wood from gray-brown gravel layer, Tr B-2, -1.5 m below sea level, asso	ciated with Middle
Jomon pottery.	
KSU-527. Yamaga 3	4130 ± 20

Wood from dark gray sand layer, Tr B-2, -3 m below sea level. Early Jomon Age.
	KSU-533. Yamaga 4 Shell from same layer as KSU-527.	4480 ± 20
	KSU-528. Yamaga 5 Wood from dark grayish-green clay layer, Tr B-2, -4 m below sea level. Early Jomon	<b>4490 ± 20</b> Age.
	KSU-534. Yamaga 6 Shell from same locality as KSU-528.	4470 ± 30
	KSU-605. Yamaga 7 Peat from second black clay layer, Tr B-2, 0 m above sea level. Final Jomon Age.	2520 ± 30
	KSU-604. Yamaga 8 Peat from second black clay layer, Tr B-2, -0.6 m below sea level. Late Jomon Age.	3140 ± 40
N	lishiurabashi Site Series	
V ir	Vood from Hikisho, Sakai City (34°31'31"N, 135°28'24"E). Collected 1983 by S. Anzat noto, K. Ohno and submitted by Y. Nakanishi, Osaka Archaeological Research Center.	o, T. Hash-
	KSU-704 Nishiurabashi 1 Wood from base of bluish gray sand layer, Sec B.	4270 ± 25
	Comment (Y.N.): Expected period: later Middle Jomon Age.	
	KSU-705 Nishiurabashi 2 Wood from same locality as KSU-704.	4360 ± 25
	KSU-706 Nishiurabashi 3 Wood from base of bluish gray sand layer, Section A.	4450 ± 40
	Comment (Y.N.): Expected period: later Middle Jomon Age.	
	KSU-707 Nishiurabashi 4 Wood in 67b line.	3640 ± 25
	Comment (Y.N.): Expected period: early Late Jomon Age.	
	KSU-708 Nishiurabashi 5 Wood in Section A.	3970 ± 30
	Comment (Y.N.): Expected period: early Late Jomon Age.	
	KSU-709 Nishiurabashi 6 Wood from a river of Jomon Age, Section D.	1630 ± 20
	Comment (Y.N.): Expected period: middle Final Jomon Age.	
	KSU-710. Nishiurabashi 7 Wood from a river of Jomon Age, Section C.	2590 ± 20
	Comment (Y.N.): Expected period: middle Final Jomon Age.	
	KSU-711. Nishiurabashi 8 Wood from a river of Yayoi Age, Section C.	2400 ± 20

Comment (Y.N.): Expected period: Early Middle Yayoi Age.

<b>KSU-712. Nishiurabashi 9</b> Wood in Section C and Section D.	2160 ± 25
Comment (Y.N.): Unknown Period.	
KSU-713. Nishiurabashi 10 Wood in Section A and Section B.	$2620 \pm 20$
Comment (Y.N.): Unknown Period.	
Misono Site Series	
Samples from Misono, Yao City (34°38'10"N, 135°35'45"E). Collected 1983 by imoto, K. Ohno and submitted by Y. Nakanishi, Osaka Archaeological Research	y S. Anzato, T. Hash- ch Center.
KSU-714. Misono 1 Charcoal and soil from Section 7B.	2690 ± 30
Comment (Y.N.): Expected period: later Early to early Middle Yayoi Age.	
KSU-715. Misono 2 Natural wood.	2120 ± 50
Comment (Y.N.): Expected period: early Kofun Age.	
Jogoji Site Series	
Samples from Jogoji, Kumatori-cho, Sennan-gun (34°23'N, 135°22'E). Coll 1984 by S. Anzato and K. Ohno, Osaka Archaeological Research Center.	ected and submitted
KSU-995. Jogoji 1 Soil and charcoal in Point 321, Section B-8.	940 ± 50
KSU-996. Jogoji 2 Soil and charcoal in Point 355, Section B-4.	$880 \pm 40$
KSU-997. Jogoji 3 Soil and charcoal in Point 355, Section B-4. Comment (Y.N.): Expected age:	<b>850 ± 50</b> AD 1300 to AD 1400.
Kyuhoji Site Series	
Samples from Nishi-Kyuhoji, Yao City (34°37'N, 135°35'E). Collected and su Imamura, 1985 by K. Ichinose, Osaka Archaeological Research Center.	ıbmitted 1983 by M.
KSU-1004. Kyuhoji 1 Peat from Layer 4a, SX303, Section H3.	1760 ± 30
Comment (K.I.): Expected period: Early Kofun Age.	
KSU-1005. Kyuhoji 2 Peat from same locality as KSU-1004.	$1700 \pm 40$
<b>KSU-1006. Kyuhoji 3</b> Peat from same locality as KSU-1004.	1680 ± 30

KSU-1007. Kyuhoji 4 Wood, Shigarami 205, w-167.	2330 ± 30
Comment (K.I.): Expected period: early Middle Yayoi Age.	
KSU-1008. Kyuhoji 5 Wood, Shigarami 103, w-1.	2550 ± 50
Comment (K.I.): Expected period: middle Middle Yayoi Age.	
KSU-1009. Kyuhoji 6 Wood, Shigarami 402, w-39. Comment (K.I.): Expected period: later Middle Yayoi A	<b>2140 ± 25</b> ge.
KSU-1061. Kyuhoji 7 Soil. Associated with Jomon pottery.	5030 ± 60
Comment (M.I.): Expected age: ca. 3000 BP.	
KSU-1062. Kyuhoji 8 Soil. Same as KSU-1061.	4050 ± 30

# **Joyama Site Series**

Wood from Joyama, Nagayoshi-Nagahara-cho, Hirano-ku, Osaka City (34°36'N, 135°34'E). Relics of Incipient Jomon (*ca.* 10,000 BP) were found *ca.* 2.5 m, and samples were found *ca.* 5 m, below present surface. Collected 1983 and submitted 1985 by K. Abe, Osaka Archaeological Research Center.

Comment (K.A.): Expected age: ca. 30,000 BP.

KSU-1063. Joyama 1 Wood.	43,800
KSU-1064. Joyama 2 Wood.	43,300
KSU-1065. Joyama 3 Wood.	40,000
KSU-1066. Joyama 4 Wood.	39,400 +7300/-3700

# Kyoto Prefecture Kitakanage Site Series

Samples from Kitakanage, Ohi-cho, Kameoka City, Kyoto prefecture (35°01'48"N, 130°32'38"E). Collected 1984 and submitted by K. Tsutsumi, Kyoto Prefecture Archaeological Research Center.

KSU-797. Kitakanage 1 Wooden stake in SD01.	1715 ± 15
KSU-798. Kitakanage 2 Wooden plate in SD01.	2495 ± 15
KSU-799. Kitakanage 3 Wooden plate in SD01.	2040 ± 15

<b>KSU-800. Kitakanage 4</b> Wooden stake in SD01.	1930 ± 70
KSU-801. Kitakanage 5 Natural wood in SD01.	1715 ± 15
KSU-802. Kitakanage 6 Charcoal from dwelling pit, SB03.	1820 ± 15

Comment (K.T.): expected period: KSI-797 to KSU-801, early Kofun Age, KSU-802, Late Yayoi Age.

### Hyogo Prefecture Iwaya-kanre Site Series

Samples from Iwaya-Kanre, Itami City, Hyogo prefecture (34°46′ 33.6″N, 135°26′36″E). Collected and submitted 1984 by T. Asaoka, Board of Education, Itami city.

KSU-841. Iwaya-Kanre 1	3170 ± 25
Wood in Section 3.	
KSU-842. Iwaya-Kanre 2 Nuts in Section 3.	3950 ± 260
KSU-843. Iwaya-Kanre 3 Wood in Section 4.	3190 ± 60

KSU-951. Arioka Castle  $3550 \pm 150$ Charcoal from Arioka Castle, Itami City (34°46′28″N, 135°26′12″E). Collected and submitted 1984 by T. Asaoka.

*Comment* (T.A.): Associated with Upper Kitashirakawa-shiki pottery of middle Late Jomon Age. Result as expected.

#### KSU-1044. Aramaki 25,800 ± 180

Peat from Aramaki, Itami City (34°48'33"N, 135°23'9"E). Collected 1984 and submitted 1985 by T. Asaoka.

Comment (T.A.): Date indicates the formative period of Itami plateau. Result as expected.

#### **Morimoto-tsuruta Site Series**

Sample from Morimoto, Itami City (34°46'36"N, 135°26'25"E). Collected and submitted 1985 by T. Asaoka.

KSU-1045. Morimoto 1 Peat from bluish-gray clay layer, Point 3 180 cm depth, Final Jomon Age.	3090 ± 20
<b>KSU-1046. Morimoto 2</b> Peat from same locality as KSU-1045.	3940 ± 120
KSU-1047. Morimoto 3 Wood from black-gray peat layer, Point 3, 270 cm depth, before Final Jomon.	3700 ± 20
KSU-1048. Morimoto 4 Peat from black-gray peat layer, Point 2, 180 cm depth.	2640 ± 20

KSU-1128. Morimoto 5 Wood from grayish-yellow peat layer, Point 4, 230 cm depth, Jomon Age.	2730 ± 25	
KSU-1129. Morimoto 6 Peat from gray clay sand layer, Point 10, 160 cm depth, Final Jomon.	2490 ± 40	
KSU-1130. Morimoto 7 Wood from gray sand layer, Point 14, 330 cm depth, Jomon Age.	3090 ± 25	
KSU-1131. Morimoto 8 Wood from gray clay layer, Point 17.	2740 ± 25	
KSU-1132. Morimoto 9 Wood from grayish-blue sand layer, Point 30-1, after Final Jomon.	2330 ± 25	
KSU-1133. Morimoto 10 Peat from black-brown clay layer, Point 30-2, after Final Jomon.	2450 ± 25	
KSU-1134. Morimoto 11 Wood from brown peat layer, Point 30-3, Final Jomon.	3630 ± 25	
Comment (T.A.): Results of KSU-1045, 1046, 1134 seemed to be older, other results as	expected.	
Kuchisakai Site Series		
Peat from Kuchisakai, Itami city (34°46'20"N, 135°26'33"E). Submitted 1985 by T. Asa	oka.	
KSU-1049. Kuchisakai 1 Peat from gray clay layer, Point 7, Early Yayoi Age. Collected 1985 by T. Asaoka.	2570 ± 40	
KSU-1050. Kuchisakai 2 Peat from grayish-blue sand layer, Point 16, Final Jomon Age.	2530 ± 20	
KSU-1127. Kuchisakai 3 Peat from black sand layer, AM 15, Layer 15, Final Jomon Age. Collected 1985 by T. I University.	<b>2690 ± 35</b> zumi, Nara	
Comment (T.A.): Result of KSU-1049 is older. Other results as expected.		
Taiyohno-oka Site Series		
Charcoal from Taiyohno-oka, Hachioji city, Tokyo prefecture (139°20'N, 36°41'E). Collected and submitted 1984 by M. Koshida, Souka University.		
<b>KSU-919. Taiyohno-oka 1</b> Charcoal from kiln N-25. Heian era.	1210 ± 30	
KSU-920. Taiyohno-oka 2 Charcoal from kiln 1. Heian era.	1420 ± 200	
KSU-921. Taiyohno-oka 3 Charcoal, same locality as KSU-920.	1160 ± 50	
KSU-922. Taiyohno-oka 4 Charcoal from O-18. Jomon Age.	860 ± 120	
KSU-923. Taiyohno-oka 5 Charcoal from Dwelling Pit P-22. Late Kofun Age.	1390 ± 80	

KSU-924. Taiyohno-oka 6 Charcoal on floor of Dwelling Pit U-27. Heian era.	1100 ± 110
KSU-925. Taiyohno-oka 7 Charcoal from Dwelling Pit S-23. Late Kofun Age.	1630 ± 30
KSU-926. Taiyohno-oka 8 Charcoal from Dwelling Pit N-25b. Late Kofun Age.	1610 ± 50
KSU-927. Taiyohno-oka 9 Charcoal from Dwelling Pit Q-24. Late Kofun Age.	1570 ± 70
KSU-928. Taiyohno-oka 10 Charcoal from Dwelling Pit N-22. Middle Kofun Age.	1390 ± 60
KSU-929. Taiyohno-oka 11 Charcoal from Dwelling Pit S-21. Late Kohun Age.	1200 ± 100

#### **Itai Site Series**

Samples from Itai site, Sasaki-cho, Taki-gun, Hyogo prefecture (35°6'N, 135°11'E). Collected and submitted 1984 by T. Mizuguchi, Board of Education, Hyogo prefecture.

*Comment* (T.M.): Associated with Knife Blade and Point of Late Palaeolithic Age (layer 9 and 44). These results are arranged in order of depth. Charcoal samples are all from fireplaces.

KSU-931. Itai 1 Peat from Point 11, Layer 40, <i>ca.</i> 100 cm depth.	19,700 ± 230
KSU-932. Itai 2 Peat from Point 11, Layer 4, 110 cm depth.	19,600 ± 200
KSU-933. Itai 3 Peat from Point 11, Layer 41, 120 cm depth.	20,400 ± 260
KSU-934. Itai 4 Wood (numerous twigs) from Point 11, Layer 9A, 130 cm. These twigs an	<b>25,000 ± 260</b> re similar to KSU-939.
General Comment: It is presumed that the twigs are of same age as KSU higher place to lower.	J-939 piled again from
KSU-935. Itai 5 Wood and peat from Point 11, Layer 9B, 140 cm depth.	22,700 ± 330
KSU-936. Itai 6 Wood and peat from Point 11, Layer 9C, 150 cm depth.	21,500 ± 230
KSU-937. Itai 7 Ash of Aira Volcano, from Point 11, Layer Iic, 170 cm depth.	17,000 ± 330

General Comment: Ash of Aira Volcano is presumed ca. 23,000 BP between KSU-936 and KSU-938. The ash includes very little organic matter. The result is affected with permeated substance afterward.

KSU-938. Itai 8	$23,600 \pm 200$
Peat from Point 11, upper part of Layer 44A, 190 cm depth.	

KSU-939. Itai 9 Wood (numerous twigs) from Point 11, lower part of Layer 44Q, 210 cm of	<b>25,900 ± 340</b> lepth.
KSU-940. Itai 10 Peat from Point 11, upper part of Layer 44B, 230 cm depth.	24,900 ± 320
KSU-941. Itai 11 Peat from Point 11, upper part of Layer 44B, 250 cm depth.	26,000 ± 340
KSU-942. Itai 12 Wood and peat from Point 11, Layer 50, 270 cm depth.	25,800 ± 440
KSU-943. Itai 13 Clay from Point 11, Layer 45,300 cm depth.	21,400 +1400 / -1200
KSU-1139. Itai 14 Charcoal from Point F-10, Layer Peat-3.	25,100 ± 360
KSU-1140. Itai 15 Charcoal from Point I-15, Layer Peat-3.	26,300 ± 360
KSU-1141. Itai 16 Charcoal from Point M-5 and N-5, Layer Peat-3.	25,000 ± 1100
KSU-1142. Itai 17 Charcoal and peat from Point F-10, Layer Peat-3.	24,700 ± 250

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# **BOOK REVIEWS**

H. E. Gove. *Relic, Icon or Hoax? Carbon Dating the Turin Shroud.* Bristol and Philadelphia, Institute of Physics Publishing, 1996: 336 p. ISBN 0-7503039-8-0.

The 16 February 1989 issue of *Nature* carried an article reporting a suite of  $^{14}$ C measurements carried out by three AMS laboratories—Arizona, Oxford, and Zürich—on four textile samples. Three were known-age control samples; the fourth was the Shroud of Turin, a stained, fire-damaged and patched rectangular-shaped piece of linen cloth showing, on closer inspection, the pale image of a man exhibiting various wounds. Since the 14th century, this cloth has been regarded by many as the burial shroud of Jesus. The <sup>14</sup>C ages obtained indicated that it was a medieval artifact.

*Relic, Icon or Hoax?* recounts what the author, Harry E. Gove, emeritus Professor of Physics at the University of Rochester, calls an "adventure"—one that "lasted too long and was filled with too much acrimony" (p. 309)—his effort to date the Shroud of Turin by the <sup>14</sup>C method. Written in a direct and bold style, the story told is no dry recitation of events by a disinterested individual. Rather, as Allen Bromley in the forward notes, this is a "personal memoir" that recites a record of "deception, outright lies, low cunning, misrepresentation, and a pathological hunger for publicity as well as solid science and technology" (p. x–xi). Professor Gove quotes a newspaper article talking about "intrigue, political maneuvering, broken promises, questioned motives and bruised egos" (p. 233) as part of the process of obtaining access to samples of the Shroud to carry out the <sup>14</sup>C measurements.

As told by Professor Gove, the story has both heroes and villains. In both camps are scientific colleagues and ecclesiastical authorities. There are colorful personalities, mysterious organizations (e.g., STURP (Shroud of Turin Research Project) and the "Catholic Counter Reformation in the XXth Century"), confrontations and machinations that would have been appreciated by Machiavelli. The author indicates (p. xiv) that the published text represents a version that was tempered on advice of his attorney. One assumes that libel laws were considered.

While one topic predominates—the author's understanding of events and his perspective on the actions and motivations of individuals involved in dating the Shroud—the narrative considers two other themes: Gove's views concerning the invention of accelerator mass spectrometry (AMS) and what the author sees as the "exasperating interaction between science and religion" (p. 10). The mass of detail in the volume suggests that the author kept a thorough diary and an archive of documents from the very beginning of the events he recites and interprets. Examples of such a record are the minutes of the many phone calls placed or received by Gove in conjunction with his Shroud adventure.

Gove reports that his direct involvement began in June 1977 with a letter from an Anglican clergyman inquiring about the possibility of dating the Shroud using the recently announced AMS method. A series of informal meetings, formal conferences, and negotiations followed. In February 1979, a request sent to the Archbishop of Turin on behalf of a group of labs, of which Professor Gove was the spokesperson, offered to date the Turin Shroud using milligram amounts of fabric. In August 1985, a protocol setting out the approach to radiocarbon-dating the Shroud was developed. One of the provisions of the protocol was that six laboratories—five AMS labs including the University of Rochester and one microcounter decay-counting laboratory—were to take part. Following a series of further discussions, the Archbishop determined in October 1987 that only three AMS laboratories—not including Rochester—had been selected to date the Turin Shroud. Despite protests over this reduction initiated by Professor Gove, including an appeal to the Pope and a request that the

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senior Senator from New York ask the United States representative to the Vatican to make inquiries, samples were removed in April 1988 and the three AMS laboratories undertook their measurements.

In telling the tale of <sup>14</sup>C-dating the Turin Shroud, the author offers a commentary on the origins of AMS technology. In his view, AMS was "invented in 1977 at the University of Rochester's Nuclear Structure Research Laboratory. . . . It was also conceived independently at the Lawrence Berkeley National Laboratory in California" (p. 7). In several other places, "invented" (pp. xiii, 233, 320) or "invention" (p. 10) is used to describe the role of the Rochester group. Future historians of science interested in the reconstruction of the discovery process in the development of AMS technology might consult Professor Gove's "Cast of Characters" entry in the book under D. E. Nelson (Simon Fraser University) where Gove states that Nelson "carried out the first measurements of carbon-14 in natural material virtually simultaneously with the group at the University of Rochester" (p. 316); also, the entry under R. A. Muller (University of California at Berkeley), which notes that Muller "invented the idea of radiocarbon dating by accelerator mass spectrometry independently of and virtually simultaneously with the University of Rochester group" (p. 316). There is also the information contained in and the dates of submission of the earliest papers appearing in the open scientific literature (Nelson et al. 1977; Bennett et al. 1977). It might also be helpful to compare and contrast statements in *Relic, Icon or Hoax?* with material included in a previous chapter-length treatment by Professor Gove on the development of AMS technology (Gove 1992).

With respect to the relationship between science and religion in the context of dating the Shroud, Gove quotes a newspaper article to the effect that Pope Paul VI called the Shroud "the most important relic in the history of Christianity" (p. 233). That statement surely does not do justice to the complexity of the actual situation with respect to relics in the western Christian tradition as a whole—and even in the Pope's own church. Despite cults and cult-like groups having adopted the Shroud, most Roman Catholic authorities have persistently refused to declare it authentic. Such a stance began in the decade of the first public display of the Shroud in the 1350s, when a local bishop is reported to have declared that the image of a crucified man appearing on the cloth had been recently painted by a local artist. The alleged corrupting nature of relics in the late Medieval church was one issue that split western Christendom at the time of the 16th century Reformation and this tradition has been passed down in all mainline Protestant groups. Only a small minority within the theologically fundamentalist segments of contemporary Christianity-Catholic and Protestant alike-support attempts to provide "scientific" data to buttress traditional religious convictions concerning such objects as the Shroud. It is from these groups that most of the continuing objections to the validity of the <sup>14</sup>C results on the Shroud ultimately appear to derive. Their common approach is to obtain scientific data in an attempt to "objectivize" religiously based positions.

It is interesting to reflect that few samples of actual or purported direct religious import have been subjected to <sup>14</sup>C dating. The dating of "Noah's Ark" wood by six <sup>14</sup>C labs in the 1970s (Taylor and Berger 1980) represents an exception. Obviously, <sup>14</sup>C values can provide definitive results only in the negative, since, even if a 1st century AD date for the Shroud had been obtained, that result alone would have only circumstantial bearing on the alleged provenience. The tradition of avoiding the dating of religiously oriented samples can be traced back to the beginning of <sup>14</sup>C studies in the Chicago laboratory—apparently due more to James Arnold's concerns than to those of Libby (J. R. Arnold, personal communication). The series of events associated with <sup>14</sup>C dating the Shroud showed the wisdom of the pioneering researchers on this point. As Professor Gove's book vividly illustrates, such high-profile samples bring out into the open less-than-ideal behavior on the part of some members of the scientific community as well as those who have a strong ideological interest in an object being dated.

This is a stimulating book that, most of the time, carries a reader along with fascinating detail. Unfortunately for the general reader, there is no discussion of the scientific basis of radiocarbon dating, and it would have been helpful to have a bibliography listing some of the earlier literature on the Shroud. This volume is not the definitive work on this topic. However, it contains a wealth of information on what, to the general public, is probably the most famous sample dated by the <sup>14</sup>C method.

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#### 118 Book Reviews

Martin Hewlett. Sangre de Cristo: A Novel of Science and Faith. Tucson, Arizona, Spirit Rider Press, 1994: 251 p. ISBN 0-9639790-0-0.<sup>1</sup>

When Martin Hewlett's novel Sangre de Cristo appeared in 1994, we considered reviewing it in *RADIOCARBON*; in the end we balked at setting ourselves up as critics of fiction. But as an appendix to R. E. Taylor's notice of Dr. Gove's history of the Shroud of Turin dating, we thought it would be appropriate to take notice of another scientist's imaginative rendering of a similar event.

Hewlett, Associate Professor of Molecular and Cellular Biology at the University of Arizona, was not directly involved in analysis of the Shroud of Turin, but has based his first novel, *Sangre de Cristo*, in part on firsthand stories about the dating from colleagues who were. As a university faculty member, he understands the complex interplay of idealistic curiosity, political self-interest and financial pressure that surrounds the research scientist; as a practicing Roman Catholic, and past participant in the Arizona Newman Center's St. Albert the Great Forum on Theology and the Sciences, he understands the symbolic importance of an artifact like the Shroud to both individual believer and the Church hierarchy. Hence in *Sangre de Cristo*, as in Gove's history, neither scientists nor believers fit neatly into the categories "hero" and "villain".

The novel is set in the science-fictional near future of the year 1999. During church repairs, Father Carriere, the Jesuit curator of medieval artifacts at the Abbey of Hautecombe in the French Alps, discovers a reliquary containing an apparently ancient burial shroud. He assumes that it, like the Shroud of Turin, is an "elaborate construction" of medieval origin. (Hewlett takes some artistic license with actual history by making the Shroud of Turin date to the 12th century instead of the 14th, so that it can be connected with a subplot involving the expulsion of the Knights Templar from Jerusalem in 1172.) Despite Carriere's skepticism, he hopes to advance his scholarly career by heading up analysis and dating of the textile. He receives authorization from the Vatican to take the shroud to Tucson for dating at the "Laboratory of Atomic Chronometry".

The dating of the shroud *per se* is of secondary importance to the major plot involving Joshua Francis, an assistant professor of molecular biology at Arizona. Like Carriere, he sees the shroud as a career opportunity, but his interest is in the traces of blood preserved on the cloth: if he can recover and clone DNA contributed by the presumably medieval body that was wrapped in the shroud, he can make a name for himself as a pioneer in the emerging field of "molecular archaeology"<sup>2</sup> (and, he hopes, improve his rather dim chances of earning tenure). Obtaining 200  $\mu$ l of the blood extract from a friend on the research team, Francis succeeds in producing a recombinant DNA clone.

But to his dismay, the AMS dating of the cloth yields a firm result of *ca*. AD 30. Word of Francis' experiment having already leaked out, a reporter at the research team's press conference underlines its obvious implications: "So, you have a linen cloth, potentially used as a burial sheet, that dates to the time and geographical location of Christ and that contains coloration produced by human blood. In addition, you have produced recombinant DNA molecules that include the DNA of the person whose blood is on the cloth. Is that correct?" (p. 95–96). What's more, by this point in the novel the reader knows that the Shroud of Hautecombe *is* the actual burial cloth of Jesus, thanks to a subplot involving a fictional Society of Arimathea, formed in the first century, whose members have tended,

<sup>&</sup>lt;sup>1</sup>Available from the publisher at 2920 E. Mabel St., Tucson, Arizona 85716-3848 USA; or see the entry for the book at <a href="http://www.amazon.com/exec/obidos/ISBN=0963979000/2742-1505152-209568">http://www.amazon.com/exec/obidos/ISBN=0963979000/2742-1505152-209568</a>, which includes an interview with the author. <sup>2</sup>In fact, molecular archaeology is already a well-established subdiscipline of molecular biology. The term was popularized at least as early as Benditt (1989); for a useful introduction to the technique, accomplishments and prospects of archaeological DNA analysis, see Brown and Brown (1992).

lost, and now positively identified the sacred shroud. So Hewlett's story quickly evolves into tangled fabric of conflicts over scientific ethics, university and Church politics, theology and financial self-interest, as researchers, journalists, fundamentalist Protestants, secret societies, the National Science Foundation, the United States Congress and the Vatican all seek to appropriate the Shroud for their own (mostly self-serving) ends. Not surprisingly, the novel's climax involves Francis's final decision about what to do with the cloth and the cloned DNA.

Apart from its similarity in plot—and even in personalities—to Gove's account of the actual shroud dating, Sangre de Cristo touches on radiocarbon dating only incidentally. (And probably nobody except readers of this journal will note the minor error in Hewlett's use of uncalibrated <sup>14</sup>C measurements: the date he offers as equating to "about 30 A.D." is cited (p. 94) as  $1922 \pm 33$  yr BP, which is supposed to signify a calendar year of *ca*. AD 28; but after calibration, the probability curve for this date actually centers around AD 78 (CALIB 3.0.3c, decadal treering data set).) Nevertheless, the moral issues that Hewlett insists on are relevant to almost anybody who draws inferences based on radioisotope dating.

If I can risk a sweeping generalization: there is no such thing as a value-neutral radiocarbon date. To put it another way, a conventional <sup>14</sup>C age, or a  $\Delta^{14}$ C ratio, almost never has any importance as a bare number. A <sup>14</sup>C date has meaning because carbon isotope content is a *proxy*, a sign that stands for something else: usually for an estimated chronological age, but also for the movements of ocean waters and atmospheric layers, the cycling of CO<sub>2</sub> between air and soil, or even (as in AMS biomedical analysis) the dose-response levels of DNA adduction for a potential carcinogen. And proxies are meaningful because they point to phenomena that humans are interested in: the age of a Paleoindian culture, the forcing effect of CO<sub>2</sub> on climate change, the contributions of various aquifers (and perhaps of the pollutants they contain) to groundwater. Ironically, this status as proxy evidence is something <sup>14</sup>C dates share with religious artifacts, which are likewise valuable not in themselves but for what they point to—as, for instance, the Shroud of Turin may be thought by believers to be evidence supporting the New Testament story of crucifixion and resurrection.

In short, the Shroud of Turin is a remarkable case, but not a special case, of the intricate relations between objective measurement or experimentation and human values. Among users of <sup>14</sup>C dates, archaeologists and anthropologists are the most keenly aware of the potential political-ethical hazards of their discipline, if only because their projects so often explode into political confrontation—witness the recent dispute over dating of the rock art in Côa Valley, Portugal, which halted dam construction (Bahn 1995), or the fierce arguments over whether the bones of "Kennewick Man" should be reburied in accordance with the Native American Graves Protection and Repatriation Act (Preston 1997; *Tri-City Herald* 1997). But the immunity from controversy of researchers in other fields is only relative. For all of us, *Sangre de Cristo* is a useful cautionary tale about the dangers of divorcing one's scientific work from the larger web of which it is invariably a part.

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Half-life of <sup>14</sup>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 <sup>14</sup>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  $\pm$  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.

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Measuring <sup>14</sup>C. In Volume 3, 1961, we endorsed the notation  $\Delta$ , (Lamont VIII, 1961), for geochemical measurements of <sup>14</sup>C activity, corrected for isotopic fractionation in samples and in the NBS oxalic-acid standard. The value of  $\delta^{14}$ C that entered the calculation of  $\Delta$  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  $\delta^{14}$ C as the observed deviation from the standard. At the New Zealand Radiocarbon Dating Conference it was recommended to use  $\delta^{14}$ 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  $\delta^{13}$ C = -19‰.

In several fields, however, age corrections are not possible,  $\delta^{14}$ C and  $\Delta$ , 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  $\Delta$  notations for samples not corrected for age.

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