^{14}C-AMS: PROS AND CONS FOR ARCHAEOLOGY

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ABSTRACT: This paper discusses AMS radiocarbon dating in terms of problems and uncertainties with reference to archaeology. Difficulties may arise from contamination and processes that disturb the age relationship between sample and event to be dated, especially for small samples used in AMS. Recommendations for sample collection and handling are given.

KEYWORDS: Isotope physics, radiocarbon, accelerator mass spectroscopy, β-decay counting, archaeology, palynology.

1. INTRODUCTION

Three isotopes of carbon occur in nature: the stable isotopes ^{12}C and ^{13}C and the radioactive isotope ^{14}C (or radiocarbon). The relative abundances are 99%, 1% and 10^{-10}%, respectively. ^{14}C is continuously formed by the interaction of cosmic ray produced neutrons with atmospheric nitrogen. This radioactive carbon isotope enters the global carbon cycle and finally decays. A stationary state of production, distribution and decay results in a constant ^{14}C concentration in atmospheric CO_2 (apart from variations yielding problems with respect to calibration).

Living plants and animals incorporating this carbon have the same ^{14}C concentration (apart from isotope fractionation). Once carbon has become fixed in the vegetable or animal tissue, ^{14}C can only disappear by radioactive decay which occurs at a known rate. Thus, the age of the carbon-containing matter can be determined by measuring the amount of ^{14}C left in the sample. A general introduction of the ^{14}C method can be found in many reviews, e.g. Mook & Waterbolk (1985), Mook & Streurman (1983), Olsson (1989).

The conventional ^{14}C age reported by all laboratories is defined by using the following assumptions and internationally agreed conventions:

1. During geological times, the ^{14}C activity of carbon-containing material during its formation has always been the same;

2. This ^{14}C activity is defined by standard oxalic acid distributed by the US National Institute of Standards and Technology (NIST, formerly NBS);

3. ^{14}C activities of the carbon content of dated samples are to be corrected for isotope fractionation, based on the measured ^{13}C/^{12}C ratio;

4. The ^{14}C half life used is 5568 years; this is known to be in error by 3%. The better value of 5730 years should not be used in order to avoid confusion with earlier reported dates. The difference can be taken into account with the larger discrepancies caused by the natural ^{14}C variations by means of calibration (i.e. converting BP to cal BC or cal AD);

5. ^{14}C ages are given in years BP (Before Present), i.e. before AD 1950.

The radiocarbon content of samples can be measured by two methods based on different principles: 1) measuring the ^{14}C radioactivity (the conventional dating method) and 2) measuring the ^{14}C concentration (by means of AMS, a form of mass spectrometry).

Conventional ^{14}C dating is based on measuring the specific ^{14}C activity in the carbon sample. This is carried out by proportional gas counting of CO_2, C_2H_2, CH_4 or C_2H_6, or by liquid scintillation counting of C_6H_6. The fractional decay rate of ^{14}C is 10^{-3} per 8 years or 6×10^{-7} per 2 days. The specific ^{14}C activity of natural carbon is (by definition) 13.56 disintegrations per minute per gram of carbon. One can then calculate that, in order to achieve a precision of around 5‰ several grams of organic matter with a carbon content of 50% are needed for an activity measurement during the usual period of two days (Mook, 1984).

The new technique of Accelerator Mass Spectrometry (AMS) is based on measuring the ^{14}C/^{12}C abundance ratio. Determining the ^{14}C concentration rather than ^{14}C decay comes to measuring a system, which is 1.5×10^{6} times as large. This implies that much less carbon is required for obtaining the same precision, i.e. a few milligrams instead of grams. In addition the measuring time is much shorter: less than one hour to a few hours, depending on the sample size and age.

The AMS technique using mg size samples, offers specific advantages and possibilities. One can distinguish

between samples which are intrinsically small, and samples from which we deliberately choose to date a selected, small part or fraction. On the other hand, mg size sampling may cause unexpected problems.

After describing the AMS technique, this paper described mg size sample aspects with emphasis on archaeological applications.

2. AMS: PRINCIPLE OF THE METHOD

As stated above, AMS is a technique which makes it possible to date small samples by means of radiocarbon. As with the conventional technique, one has to prepare pure CO_2 from the sample material. For organic materials this is achieved by means of combustion after thorough pretreatment (Mook & Streurman, 1983). AMS implies an extra step: the CO_2 has to be reduced to C (graphite) by means of a reaction with hydrogen gas (Hut et al., 1986). The use of carbon in the form of solid graphite is dictated by the ion source technology; recently, however, the Oxford group has demonstrated the feasibility of a gaseous CO_2 source (Bronk & Hedges, 1990).

Direct atom counting is possible by means of mass spectrometry (MS). Any mass spectrometer consists of 3 basic elements: an ion source, a flight tube and analyser (White & Wood, 1986). There are many forms of MS developed around these elements. For instance, IRMS (Isotope Ratio Mass Spectrometry) was developed for the measurement of isotopic abundance ratios such as $^{13}C/^{12}C$. In this case, CO_2 gas is ionized in the ion source to CO_2^+ molecules. These molecules are accelerated in vacuum through a flight tube by means of an electric field, typically a few kV (thousand volts). Next, the beam of molecules is led through an analyser, usually a magnet. The magnet separates charged molecules according to mass, yielding beams of $^{12}C^{16}O_2$ (mass 44), $^{13}C^{16}O_2$ (mass 45), $^{12}C^{16}O^{18}O$ (mass 46), etc. These beam currents can be measured very accurately, yielding isotopic abundance ratios such as $^{13}C/^{12}C$, reported in $^{13}\delta$ with respect to a standard such as PDB.

It is this technique that is used in archaeology for fractionation correction of radiocarbon measurements: a 1‰ deviation in $^{13}C/^{12}C$ means an error in $^{14}C/^{12}C$ corresponding to 16 years. In conventional counting, the same CO_2 gas prepared from the sample to be dated is used for the proportional counters (typically 1 litre for ^{14}C) as for the mass spectrometer (1 ml for ^{13}C).

Measuring isotopic abundance ratios by IRMS was developed about 4 decades ago (Craig, 1957) and works very well for isotopes such as ^{2}H, ^{13}C, ^{15}N, ^{18}O. These isotopes have natural abundances ranging from 0.01 to 1.1%. The abundance of cosmogonic isotopes (^{14}C, but also ^{10}Be, ^{26}Al, ^{41}Ca) is less by a very large factor; for instance the abundance ratio for modern ^{14}C is 10^{-10}%, and for 50,000 year old samples about 10^{-13}%.

At these low levels, 'standard' IRMS is not possible because of the isobar problem: the occurrence of $^{12}CH_2$, ^{13}CH and ^{14}N (air!) is much higher using even the best vacuum technology.

This problem can be overcome by a special form of MS: accelerate C ions (rather than CO_2 molecules) over several MV (million Volts, ratherthan kV) – Accelerator Mass Spectrometry, or AMS. An AMS system consists of the following components:

Ion source. The graphite from the sample is pressed into small holes (typically 2 mm) in sample holders. These holders are loaded into a carrousel and can be individually selected by rotating the carrousel to load the desired sample into the ion source. The source is a so-called sputter source containing a cesium gun; a beam of Cs ions is focused on the loaded target. Negatively charged carbon ions (C^-) are sputtered from the target. This way, a beam of C^- ions is extracted from the source into the AMS system;

Injector. The injector consists of one or more magnets, designed to inject the C^- ions into the accelerator. These magnets also act as a mass discriminator so that only the ions with desired masses will be injected into the accelerator;

Accelerator. The accelerator is a flight tube for the carbon ions with a length of several meters. In the middle (the so-called terminal), a positive high voltage is applied; in the Groningen machine this is 2.5 MV. The negative carbon ions are accelerated to the terminal. At the terminal, the C^- ions are led through argon gas; in this process, they are 'stripped', i.e. they loose electrons – usually four so that C^- becomes C^{3+}. The positively charged ions are now repelled from the terminal; they leave the accelerator tube with an effective acceleration voltage of 2.5+3x2.5 = 10 MV. Because of the two acceleration steps, this type of accelerator is called a Tandem Accelerator;

Analyser. The analysing part is a combination of magnetic and electrostatic ion optical elements, designed to separate the ^{12}C, ^{13}C and ^{14}C beams. The ^{12}C and ^{13}C beams (needed for fractionation correction) are measured as currents, the ^{14}C particles are counted in a so-called ion chamber.

Using this elaborate technique enables mass spectrometric analysis of ^{14}C by solving the isobar problem:

1. ^{14}N can not form negative ions, and is therefore essentially not part of the ion source/accelerator system;

2. The stripping process at the terminal destroys molecules ($^{12}CH_2$, ^{13}CH) and other molecular fragments present in the accelerator system.

AMS has been developed since 1977 (for a review see Elmore & Phillips, 1987). There are now three generations of AMS systems:

Large Tandem Van de Graaff accelerators. These were originally built for nuclear physics research in the 1950's and 60's. Several machines have been rebuilt for applications such as AMS. This work was pioneered by the groups in Rochester and Zurich. These accelerators are large. Depending on their original mission, some

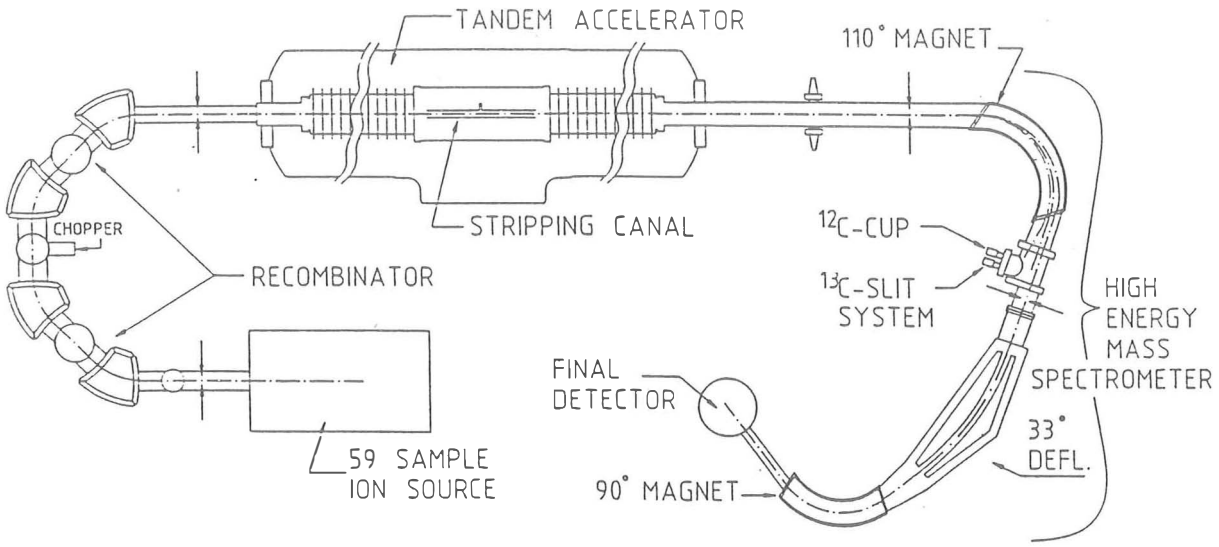

Fig. 1. Lay-out of the Groningen AMS, showing the principal components: ion source, injection system (recombinator), accelerator and analyser (high energy mass spectrometer).

can obtain terminal voltages above 10 MV;

Tandetron – the first generation (1980's). This is a small accelerator (terminal voltages around 2 MV) especially designed for AMS (in particular ^{14}C). Systems of this generation are operating in Oxford, Toronto and Tucson;

Tandetron – the second generation (1990's). This is an improved version of the older Tandetron-based systems. The major upgrade is that the three isotopes ^{12}C, ^{13}C and ^{14}C are accelerated simultaneously through the tandem – in the older systems this took place sequentially. The new generation is designed as a high-throughput, fully automated mass spectrometer (Purser, 1988). These systems are operating in Woods Hole and Groningen. The ion source carrousel can contain up to 59 samples. A lay-out of the Groningen AMS, showing the principal components is shown in figure 1. One can easily recognize the components described above: ion source (with carousel for 59 samples), injection system (so-called recombinator; Purser 1988), accelerator, and high-energy mass spectrometer (analyser for ^{12}C, ^{13}C and ^{14}C).

Compared to conventional radiocarbon dating, AMS has the following advantages:

1. The amount of material needed can be a factor of 1000 less (mg rather than g of carbon);

2. The time needed for measurements is much less (hours rather than days).

In addition, the precision of the 3rd generation accelerators is higher than that of most conventional dating laboratories, and comparable to that of high-precision conventional laboratories, i.e. better than 5‰.

With conventional dating, the age limit is about 50,000 years; for older samples the count rate is so small as to be comparable to the cosmic ray background which can never be fully eliminated. In principle, AMS is insensitive to background radiation; the 'machine background' is at least 100,000 years. In practice, dates as old as this have not yet been achieved, probably due to the contamination of samples with younger carbon which cannot be completely removed at this level. On the other hand, enrichment procedures for AMS samples may make dating very old samples (50-75 ka) feasible (Grootes et al., 1980)

3. AMS: EXPERIENCE IN ARCHAEOLOGY

More and more AMS laboratories are producing dates for archaeology and related fields of study on a regular basis. Despite this, it is not yet possible to get an overview of the results. An exception to this is the Oxford laboratory, thanks to its datelists which appear regularly in the journal *Archaeometry*. These give a clear picture of both the possibilities and the problems of AMS dating. The following survey is illustrated chiefly with examples from the Oxford datelists.

3.1. Possibilities

The great advantage of AMS is, of course, that very small quantities of carbon containing material can be used for dating. This is illustrated in the following examples:

OxA-3289	Altar Wedge Tomb. Single unburnt human premolar (datelist 17)	3670± 80 BP
OxA-931	Stepleton. Single grape pip from pit 52 (datelist 7)	4660± 80 BP

OxA-3199	Hogebeintum. Small shell bead Not corrected for reservoir effect (datelist 15)	1820± 75 BP
OxA-942	Rekem. Resin attached to Tjonger point (datelist 5)	11,350±150 BP

This means that artefacts of organic material which could not previously be sampled because this would have necessitated the destruction of the object itself or of a large part of it, can now be dated. The very small amount of material needed for AMS allows the artefact to be sampled without visible damage.

An additional advantage is that there is less chance of a mixture of carbon from different sources being present. AMS samples may consist of a single piece of charcoal, a fragment of bone, etc. Conventional radiocarbon samples frequently consist of a large number of fragments of charcoal, several pieces of bone, etc. which may have come from several trees or animals. As a result, there is a very real chance that material of different ages will be present.

Despite the advantages of AMS, conventional radiocarbon dating should not be abandoned. Conventional radiocarbon dating and AMS should both be used, each having their own merit.

3.2. Problems

The Oxford datelists show that some of the advantages that AMS offers are not without their own problems.

3.2.1. *Charcoal fragments/seeds*

Charcoal, in whatever form, is a very resistant material. Contamination with 'older' charcoal, i.e. charcoal which had been already in the ground for a period of time, was already recognized as a problem with conventional radiocarbon dating. With AMS, an additional problem arises from the mobility of very small pieces of charcoal, as a result of which younger material may be present in older contexts. Examples of this are:

Hengistbury Head. Late Palaeohistoric settlement with an estimated date of 12,000 BP. Four small charcoal samples from an area in which hearth stones were found, were dated (datelist 2):

OxA-398	8590±120 BP
OxA-399	4770±180 BP
OxA-411	7690±100 BP
OxA-412	8140±120 BP

Rekem. Several concentrations of Federmesser artefacts were excavated. Concentration 7 is dated on resin on a Tjonger point (OxA-942, 11,350±150 BP). Four small charcoal samples have been dated (datelists 5 and 9):

OxA-943	RE-5, hearth	2230± 70 BP
OxA-944	RE-5, hearth	6390±100 BP
OxA-945	RE-10	9900±110 BP
OxA-1375	RE-10	5220±100 BP

Barmose 1. Very early Mesolithic settlement from the late Preboreal. The flint assemblage is similar to Duvensee 8 and Star Carr, on the basis of which a date of around 9500 BP may be assumed. Five small charcoal samples were dated (datelist 15):

OxA-2248	9370±90 BP
OxA-2249	8930±90 BP
OxA-2250	9160±90 BP
OxA-2251	9250±90 BP
OxA-2252	9130±90 BP

With the possible exception of OxA-2248, all these dates are too young. Peat immediately above the level of the finds was dated to 4470±100 BP (K-1773). Apparently, in the course of the centuries after settlement had ceased, the site was contaminated with charcoal, of either anthromorphic or natural origin.

Letterkeen. Charcoal particle found below the paved floor of a stone cist with Food Vessel of the Early Bronze Age (datelist 17):

OxA-2656	1320±70 BP

Long Down flint mines. Charcoal from dumped chalk rubble 1m below present ground surface, mixed with early Neolithic pottery sherds, antler and bone. Antler and bone were dated to 4900±100 BP, OxA-1151, resp. 5050±100 BP, OxA-1152, as expected (datelist 7).

Two charcoal samples (*Corylus*) appear to be considerably younger:

OxA-1063	3110±80 BP
OxA-1088	3130±60 BP

Flagstones. Two grape pips found in the well-sealed Bronze Age fill of an enclosure ditch, 1 m below the present ground surface (datelist 13):

OxA-2380	205±60 BP
OxA-2381	160±60 BP

Slough House Farm. Two samples of grain from the filling of prehistoric features, in both cases however, shallow, unsealed contexts (datelist 14):

OxA-3036	*Tr. dicoccum* & *Tr. aestivum* from Neolithic enclosure ditch	1570±100 BP
OxA-3037	*Avena* from Late Bronze Age enclosure ditch	530± 90 BP

This does not mean that small fragments of charcoal and seeds should be considered to be unsuitable. On the contrary, the Oxford datelists give many examples of useful results from this type of material. Much depends on the context and the degree of association with the subject of the dating exercise. But serious consideration must be given to the vertical movement of charcoal particles and seeds, above all when dealing with lighter soils subject to much worm activity, burrowing animals etc.

3.2.2. *Organic tempered pottery*

At first sight, pottery with organic tempering would

appear to be very suitable for AMS dating. In practice, however, the opposite is the case, because it is not possible to isolate the charred temper particles. The burning of the entire sherd usually results in dates which are too old, because clay naturally includes carbon.

Oxford datelist 5 gives a number of examples of dates from early Neolithic pottery with expected ages of c. 7000 years (Banja), resp. c. 6500 BP (Grivac and Vinca):

OxA-558	orange semi fineware, Banja	9310±150 BP
OxA-559	dark monochrome fineware, Grivac	8760±150 BP
OxA-368	coarse barbotine ware, Vinca	7230±170 BP

Where it was apparently possible to isolate the organic temper, the dates were as expected. This was the case with the early Neolithic pottery from China which was tempered with rice straw (datelist 14):

Pengdoushan

OxA-2210	sherd 1	7550± 90 BP
OxA-2214	sherd 2	7040±140 BP

Hujiawochan

OxA-2731	sherd 1	6580± 90 BP
OxA-2218	sherd 2	6210± 90 BP
OxA-2222	sherd 3	6310± 90 BP
OxA-2733	sherd 4	6350±170 BP

It is strongly recommended that pottery with organic temper should only be dated if the temper can be separated from the clay matrix, which is a very labour-intensive procedure, however (see Hedges, Chen Tiemei & Housley, 1992). This is quite apart from the possibility of dating pottery with charred food residues.

3.2.3. *Bone with a low collagen content*

Initially Oxford encountered problems in dating bone with a low collagen content (that is, with less than 5% of the original quantity of c. 200 mg collagen per gramme of bone), if the standard pre-treatment was applied (isolation of the crude collagen and the extraction of crude gelatine from this). The difficulty appears to lie in the incomplete separation of the collagen from the specific soil-derived materials such as humic acids, and the fact that bone with such a low collagen content often contains insufficient extractable gelatin. Due to chemical changes a large portion can no longer be dissolved. Examples of this are:

Aveley. Two dates on bone from *E. antiquus* with an expected age of at least 100,000 years. The bone contained only 0.3 mg of collagen per gramme (datelist 5):

OxA-370	collagen	20,640±630 BP
OxA-823	collagen, separate preparation	21,100±400 BP

Thatcham. Two dates on bone with a low collagen content from the early Mesolithic settlement. Expected age, c. 9500 BP:

OxA-940	collagen, humerus of pig	6500±130 BP
OxA-1201	collagen, femur of beaver	5100±350 BP

An extensive pre-treatment, in the form of ion-exchange of the gelatin, or even hydrolysis of the gelatin and ion-exchange of the amino acids, usually yields acceptable dates. This is extremely labour-intensive, and can not be applied routinely.

3.2.4. *Conserved bone*

The conditions as outlined for bone with a low collagen content are also applicable to conserved bone. The standard pre-treatment is not sufficient to separate collagen from the preservative. An intensive pre-treatment as described above can give good results. Some examples:

Olenii Ostrov (Karelia). Grave 57 of a large Mesolithic cemetery. The bone was treated with preservative (datelist 11):

OxA-1664	crude collagen	5700±80 BP
OxA-1665	gelatin	7280±80 BP
OxA-2266	ion-exchanged gelatin	7350±90 BP

Cuello (Belize). Burial 62 from a 'Preclassic' Maya cemetery. The bone was treated with preservative (datelist 12):

OxA-1648	collagen	3750± 70 BP
OxA-1649	gelatine	3000± 60 BP
OxA-2103	aminoacids	2840±100 BP
OxA-2166	aminoacids	2830± 70 BP
OxA-2112	'contaminant'	4540±100 BP

It is also true here that extensive pretreatment yields acceptable results, but that this labour intensive procedure cannot be carried out on a routine basis.

3.2.5. *Charred/burnt bone*

There is a sliding transition between charred and burnt bone, that is, between bone which has been exposed briefly to heat so that the organic part has been charred and bone that has been exposed to heat for a long period so that practically all the organic content has disappeared. In general, charred bone yields reliable results; with burnt bone it is often not clear what the source of the carbon containing material is, which remains after the inorganic part has been dissolved. But charred bone can also yield strange results, as a result of absorbing carbon containing material from the surrounds. The Oxford datelists contain various examples of this:

Barrow Hills	cremations associated with Collared Urns	datelist 11
Hayonim Terrace Natufian		datelist 15

On the other hand, an example from datelist 14 can be cited, which demonstrates that charred bone can yield good results:

OxA-3025	insoluble fraction of charred bone fragments belonging to St. Adalbert (early 8th century)	1300±70 BP

4. AMS: RECOMMENDATIONS

In view of the cost of AMS dating and the problems associated with certain categories of material, AMS should be used chiefly in circumstances where the specific possibilities of the methodcan best be exploited. The Oxford datelists clearly demonstrate which applications are most appropriate.

4.1. Dating of artefacts of organic material

AMS allows for the dating of artefacts of organic material without causing unnecessary damage to the artefact. The amount required is so small that sufficient material can usually be collected from one or more small cores. Conventional dating is ruled out if the complete artefact has to be sacrificed. The advantage is clear: there is absolute certainty that the date relates to the artefact.

There are many examples of this application:

Antler and bone points	datelist 10
Antler mattocks	datelists 7 & 10
Wooden bowls	datelists 12 & 14
Wooden figurines	datelist 9
Shell-bead necklace	datelist 15
Turin shroud	datelist 11

4.2. Dating of organic remains on artefacts of non-organic material

This concerns organic remains with which the artefact came in contact during its use, or which as a result of use were deposited on it. In this case, complete certainty of association is involved. The following examples have been selected from the Oxford datelists:

Rekem. Resin on Tjonger point, Concentration RE-7 (datelist 5):
OxA-942 11,350±150 BP

Bronneger. Burnt food residues on sherds of a pot of the Swifterbant culture, dredged up from the Buinen-Schoonoord canal (datelist 15):
OxA-2908 5890±90 BP

Seuso. Soot from the surface of the large cauldron which contained the Seuso hoard. The date is accurate enough to make clear that this hoard belongs to the late 3rd – early 5th century, rather than the 6th/7th century (datelist 13; see also Bender, 1992):
OxA-2175 1740±55 BP

4.3. Dating of human remains

This category is concerned with material which depending on its size may vary from an isolated fragment, to a complete skull, a complete skeleton, a bogbody or a mummy. This material is frequently of value to physical-anthropologists and is sometimes of exhibition value. In most cases, dating by conventional means is not possible. Practically every Oxford datelist includes examples of datings of this type of material. A few typical examples are:

Ofnet skulls	datelist 9
Lindow man	datelist 7
Drente bogbodies	datelists 9 & 14
Ötztal 'ice man'	datelist 15
'River Valley People'	datelist 5

An interesting application is the dating of the period of use of collective burials. In collective burials the remains of a large number of individuals are often found, although only exceptionally a virtually complete or anatomically intact skeleton is found. Usually the burial consists of a mixture of bones and fragments of bones, in which only a single bone or some small fragments of each individual may be present. Physical anthropologists are in any case able to define a minimum number of individuals on the basis of the number of specific bones. By using AMS it is possible to date each of these bones even when these are small bones such as heel bones. But even if these were long bones and conventional radiocarbon dating could be applied, AMS still would have the advantage that these bones with their intrinsic physical-anthropological value need not be completely sacrificed. By this means is it possible to get an impression of the duration of use of the grave (provided, of course, that cremation was not also practised). Examples:

Hazleton Long Cairn	datelists 7 & 8
Poulnabrone Portal Tomb	datelist 10
Lough Gur Wedge Tomb	datelist 17
Rössberga Passage Grave	datelist 14

4.4. Bone from Palaeolithic cave deposits

Many dates produced by the conventional method for Palaeolithic deposits in caves are on collections of bone fragments, usually originating from more than one individual, and frequently without clear traces of human modification. The availability of this material in a specific layer with a flint industry certainly does not have to mean that it originated in a human settlement. It could also be connected with animal activity during periods without human occupation. Dating 'humanly modified' bone fragments by means of AMS avoids this type of association problem.

The Oxford datelists include many examples of this type of dating, chiefly from English caves.

4.5. Refining typochronologies of prehistoric pottery

The existing ^{14}C chronologies of prehistoric pottery are as a rule based on conventional radiocarbon dates on charcoal from graves and settlements. The use of charcoal involves some real disadvantages, which can be brought together under the umbrella term 'old wood effect'. Unknown number of year rings in the sample, no information concerning the number of rings missing between the youngest piece of charcoal and the outer-

most part of the tree, the possibility of a long period of use before the wood was burnt. 'Old wood effect' can in extreme cases be in the order of centuries, but there are also charcoal samples with a negligible 'old wood effect'. As a result ^{14}C dates based on charcoal are, in general, too old and furthermore give a diffuse picture with large overlaps of successive phases. By using samples with a negligible own age, where re-use can be ruled out, a refined typochronology can be obtained.

Furthermore, by using small samples, such as a small fragment of bone, a burnt grain or cooking residues on pottery, the danger of mixing older and younger material is avoided. AMS offers these possibilities.

The Oxford datelists include some interesting examples:

Linear Pottery Culture (LBK). On the basis of conventional radiocarbon dates it appeared that the oldest phase of the LBK, phase Ia, could be dated to before 6400/6500 BP, and perhaps even to around 7000 BP. Datelist 9 includes a series of dates on bone and burnt grain from various sites in Austria and Germany, which made it clear that the earliest LBK should be placed in fact between 6300 and 6100 BP. Datelist 17, includes a number of dates around 6200 BP on charred grain from LBK-settlements on the Aldenhovener Platte, from contexts corresponding to late LBK phases II b and c. Wiggle matching on a decadel calibration curve shows that phase Ia can be placed between 5260-5210 cal BC, phase IIb/c between 5080-5040 cal BC. It is clear that LBK lasted much shorter than the traditional charcoal dates suggested (see Appendix 1).

Tankardstown I and the chronology of western Neolithic pottery. A similar difference between charcoal and grain dates is also present at House 1 in Tankardstown Co. Limerick (datelist 9). Four samples of oak charcoal, originating from charred planks in the wall trench, produced conventional dates of 5085±25, 5105±45, 5005±25 and 4880±110 BP (GrN-16643, -14713, -15386, -15387). Two samples of charred grain from a posthole within the house gave dates of 4890±80 and 4840±80 BP (OxA-1476, -1477). Wiggle matching provides a precise date for House 1, and for the associated early western Neolithic pottery (see Appendix 2).

4.6. More accurate dating of pollen diagrams

Pollen diagrams have up to now been dated using slices of sediment (peat, gyttja), the thickness of which depended on the quantity of material available, which thus could go to 5-6 cm in cores of boreholes. A sample of this thickness could include the sediment of many decades. Samples of peat and gyttja often have problems caused by younger root penetration, infiltration by younger humic material etc.

It is possible to reduce these problems by selecting macrofossils such as seeds and stem fragments embedded in the sediment for dating by AMS. This material has a negligible own-age and there is a much greater certainty that it was contemporary with the formation of the sediment. In principle, using similar samples, it should even be possible to reconstruct the 'wiggles' in the curve of the atmospheric ^{14}C content, which is particularly important for the Late Glacial period.

An early application was the dating of terrestrial macrofossils from calcareous sediment in the Swiss lakes. At Usselo, two seed samples from a thin organic band beneath the Bølling peat have been dated by AMS.

4.7. 'Wiggle-matching'

Finally, AMS can also be used for the absolute dating of large wooden items. In principle, by using multiple samples each with only a few year rings and from a known position relative to each other, a tolerably accurate dating for the youngest ^{14}C dated ring can be obtained by wiggle-matching. However, the success of this application depends on the shape of the of the ^{14}C curve at the time the wood was growing. This method is not possible in flat parts of the curve. This type of dating has so far been rarely used due to the costs involved. One of the few examples is the dating of St. Cuthbert's coffin, with only two samples, each with one year ring (datelist 11).

5. CONCLUSIONS

The specific possibilities of AMS are clearly demonstrated. But in the future also, the most desirable samples will not always be available. And then quickly the tendency will be present to submit samples of lower quality for dating. In part this will concern small amounts of the large samples of wood, charcoal, bone or peat which up till now make up the majority of the material submitted to conventional ^{14}C laboratories. But now that AMS is available, the great temptation will be to collect and submit isolated flecks of charcoal. The dating of this type of sample by AMS should be discouraged. If sufficient material is available, the samples can be more cheaply and as accurately dated by conventional means. The possibility of disappointment in the form of an unexpected date is great when isolated small fragments of charcoal or seeds are used, and certainly if these have not been found in a clearly defined feature or in a close association. Moreover, in large parts of Europe the ^{14}C chronology of the pre- and protohistoric period is broadly known. This chronology is to a large extent based on charcoal samples – with all their associated problems. The archaeological community is not concerned with reaffirming this chronology, only with its refining on the basis of dates with small standard deviation, performed on material with a negligible own-age and with a high degree of association with artefacts or features.

In addition, there is a need for dating of artefacts of organic material which are either not found in graves or

in settlements or do not survive in these contexts, but which are usually found unassociated in bogs and rivers.

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APPENDIX 1: The duration of the Linear Pottery Culture, by J.N. Lanting

Since ^{14}C-dates first became available for the Linear Pottery Culture (LBK, after its German name *Linienbandkeramische Kultur*) in the early 1950's, archaeologists have speculated on the duration of this culture.

According to Modderman (1970: Ch. VIII) and Stehli (1973: pp. 98-100) the LBK in the southern part of the province of Limburg and on the adjacent Aldenhovener Platte in Germany can be divided in seven phases, numbered I^b-I^d (earlier LBK) and II^a-II^d (later LBK). The earliest LBK, phase I^a, does not occur in these areas. In the Middle Rhine area – roughly the area between Cologne and Mainz – a younger phase of LBK occurs, called III^a by Dohrn-Ihmig (1973), which seems to be the last LBK anywhere (see also Coudart, 1989: fig. 2).

Later, Stehli (1989a) developed a system of 15 *Siedlungsphasen* for the LBK settlements in the Merzbach-valley on the Aldenhovener Platte. A correlation of the two systems can be extracted from literature, with some difficulty:

Merzbach I-V	=	Modderman I^b
Merzbach VI	=	Modderman I^c
Merzbach VII	=	Modderman $I^{c/d}$
Merzbach VIII	=	Modderman I^d
Merzbach IX	=	Modderman II^a
Merzbach X	=	Modderman $II^{a/b}$?
Merzbach XI	=	Modderman II^b
Merzbach XII	=	Modderman $II^{b/c}$?
Merzbach XIII	=	Modderman II^c
Merzbach XIV	=	Modderman II^d
Merzbach XV	=	Modderman II^d

Based on then available ^{14}C-dates Stehli (1989a) placed Merzbach I around 5330 cal BC, and Merzbach XV around 4980 cal BC. Each phase lasted about 25 years. For the earliest LBK, Lüning (1980) considered that phase I^a started around 5700-5800 cal BC or 6750-6850 BP. Whittle (in: Hedges et al., 1989: p. 225) also was in favour of phase I^a beginning between 7000 and 6500 BP.

Recently, a series of grain and bone samples from a group of earliest LBK settlements, and also from settlements of the later LBK in the Merzbach valley (phases XI-XIII) were dated by AMS in Oxford. The results, which were all more-or-less the same for both groups and in the order of 6300-6100 BP, caused a certain amount of disbelief amongst the submitters of the phase I^a samples (see comments in: Hedges et al., 1989: pp. 224-226; Hedges et al., 1993: pp. 317-318; see also: Whittle, 1990). But it will be shown here that these dates actually allow reliable estimates of the duration of LBK in general, and of the 15 Merzbach phases in particular.

As a starting point, it is assumed that the typochronology of LBK pottery in southern Limburg and on the Aldenhovener Platte, as worked out by Modderman (1970) and Stehli (1973), is correct. As this system has proved itself repeatedly when applied to new find groups, this seems to be a valid assumption. Stehli (1989a) has even demonstrated that it was possible to refine this typochronology when he developed his 15 Merzbach phases. It is also safe to assume that an even earlier phase of LBK exists, that does not occur in southern Limburg and on the Aldenhovener Platte, which can be called phase I^a. The possibility that this I^a material could be contemporary with part of phases I^b-II^d can be excluded.

As there are no obvious reasons to doubt the reliability and accuracy of the Oxford AMS dates, it seems logical to try to divide the two sets of dates in such a way over the calibration curve that the phase I^a dates fall into one range of calendar years and the Merzbach dates into another, separated from the earlier one by an 'empty' period, wide enough to accommodate Merzbach phases I-X. This involves a kind of wiggle-matching, for which the decadel calibration curve of Stuiver & Becker (1993) can be used. The Oxford dates were after all produced on materials with negligible own age.

The wiggle-matching can be carried out in only one way. The dates for earliest LBK have to be placed between 5300 and 5200 cal

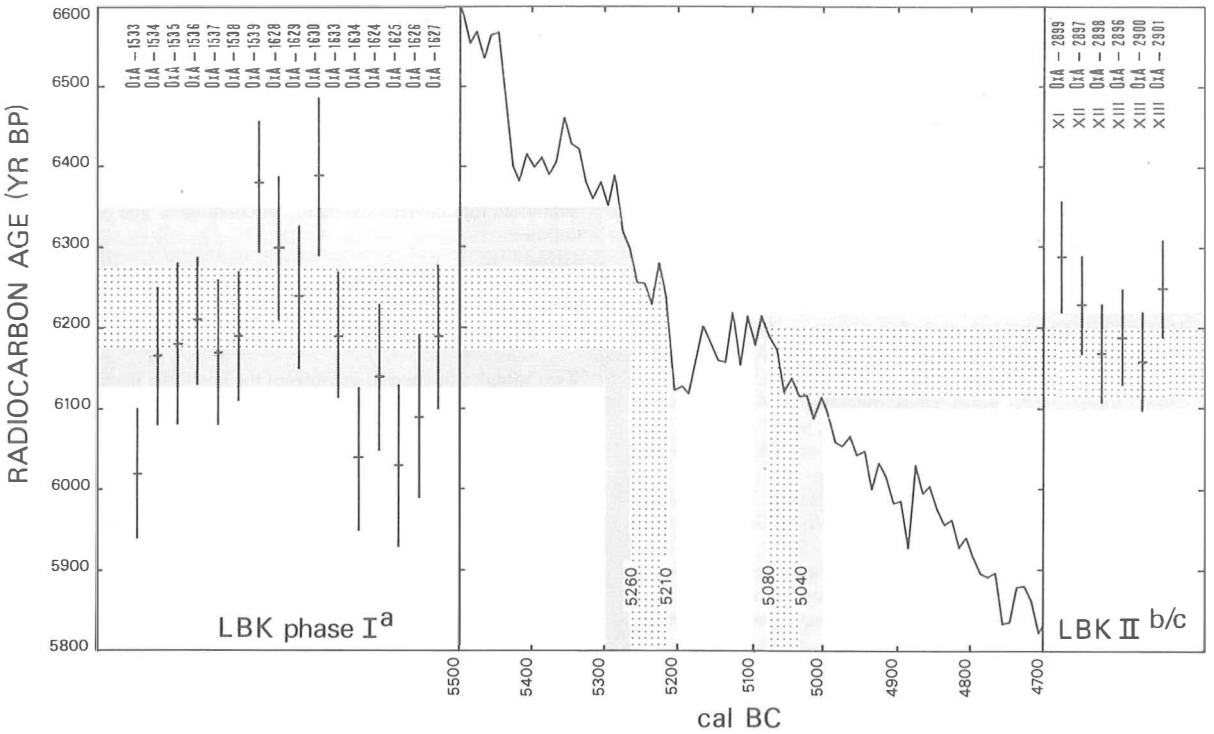

Fig. 2. Wiggle matching of a series of bone and grain dates from earliest LBK contexts (phase Ia), and a series of grain dates from later LBK contexts (phases II b-c, Merzbach *Siedlungsphasen* XI-XIII). The shaded bands represent the maximum timespans, both in terms of radiocarbon and calendar ages, resulting from this exercise. The lightly shaded areas within these bands represent the most likely timespans (see text). The decadel curve of Stuiver & Becker (1993) was used. Drawing B.A.I., J.H. Zwier.

BC, those for Merzbach phases XI-XIII between 5100 and 5000 cal BC (see fig. 2). Within these two ranges some movement is still possible, however. The beginning of phase I^a can be placed later, around 5250 cal BC, but in that case it must be assumed that with two of the Oxford dates the real ^{14}C-ages fall outside the one-sigma ranges. The final date of phase I^a may be 10 years earlier at the most, in which case one must assume that with 3 ^{14}C-dates the real age falls outside the one-sigma range. Given that 16 dates are involved, the number if 5 exceptions is acceptable. In a large series of dates nearly 1/3 of the results should lie outside the one-sigma ranges. The date for the beginning of the Merzbach phases XI-XIII cannot be earlier than indicated, because otherwise the timespan for phases I-X is too short. It is more likely that phase XI started somewhat later, around 5080 cal BC. In that case it must be assumed that the real age of the phase XI sample lies outside the one sigma range. The end date of this small series could in fact be placed around 5050 cal BC without any difficulty.

The calibrated dates for Merzbach phases XI-XIII are confirmed by the dendrodates of the three construction periods of the wooden waterwell in the LBK settlement near Kückhoven, of 5090, 5067 and 5055 BC. According to Lehmann (1993) the ceramics in the refuse layer in the oldest well belong to 'later LBK'. On the other hand, according to Weiner (1993) habitation in Kückhoven continued after the third well had gone out of use. This is sufficient to demonstrated that the three wells must be placed somewhere during Modderman's periods II$^{a, b}$ and ^{c}. Given the maximum timespan available for Merzbach phases I-X, namely 5210-5080 BC, the length of a *Siedlungsphase* must be much shorter than the 25 years Stehli (1989b) allowed. In fact each phase cannot have lasted more than 13 years. If Merzbach phase XI started earlier than 5080 BC, the length of each phase must have been even shorter. Merzbach phases XI-XIII can be parallelled with Modderman's periods II^b and ^{c} (see above), and cannot have lasted more than 40 years. A duration of 5080-5040 BC seems likely. That means that the first well in Kückhoven must have been constructed during Merzbach phase X (= II$^{c/b}$?) and the third well during the transition of phases XII and XIII (= II^c). If phase XI had already started by 5100 BC the first well should have been constructed near the end of this phase, for a *Siedlungsphase* would then have lasted only 11 years. The third well should have been constructed by the end of phase XIV (= II^d) in that case. This contradicts the published evidence, however.

During the earliest LBK the duration of each settlement phase will not have been much longer or shorter than during later periods. If Lüning (1988, p. 38) is correct in stating that the settlements of phase I^a lasted only three house generations (read: *Siedlungsphasen*) phase I^a cannot have lasted much longer than half a century, and consequently must have started around 5260 BC. This is not contradicted by the results of the wiggle-matching. In fact it fits these results quite nicely. On the other hand, it seems likely that even Dohrn-Ihmig's phase III^a will have come to an end before 5000 BC. This means that the whole of LBK may have lasted only some 250 years.

It must be clear that this precise chronology of LBK could only be reconstructed because dates on grain and bone were available. This reconstruction would not have been possible with dates on charcoal, because charcoal samples contain a number of chronological uncertainties. It is hardly ever clear how many rings a charcoal sample contains, how many rings are missing (because heart- and sapwood have been removed) and how much time has passed between the moment of cutting the wood and the moment of charring. With this type of sample wiggle-matching should not be carried out.

An impression of the influence of the chronological uncertainties inherent to charcoal samples can be obtained by comparing the charcoal dates of LBK in the Lower Rhine area (Modderman, 1970; Stehli, 1989b; *Liste* 1) with the ages that can be expected based on the wiggle-matching described above. It is clear that by using charcoal

age differences of 100 years occur regularly. In a few cases age differences of several centuries are visible.

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APPENDIX 2: Tankardstown 1, Co. Limerick, Ireland, by A.L. Brindley

The first of two Neolithic houses in Tankardstown South, Co. Limerick was discovered and excavated in 1986 and 1987 and published shortly afterwards (Gowen, 1988). A second house was found nearby and excavated in 1987 and 1988; a preliminary report of this has been published (Gowen & Tarbet, 1988).

The remains of House 1 consisted of a wall trench containing packing boulders and charred wood remains. This trench had apparently supported a wall of split oak timbers, of a rectangular house of 7.4x6.4 m. Two post-holes, probably for roof supports, were found in the interior. One of these contained quite a lot of charred wheat remains. The fill of the wall trench contained locally large quantities of charred remains of dried wild apples.

Regarding the charred wood remains in the wall trench, the excavator remarks that "discrete concentrations of charcoal with a vertical grain were noted. They were never very thick and it was not possible to determine the timber sizes from them. Rather, it appeared that the charcoal represented a burnt skin on the exterior of the timbers which had been removed" (Gowen, 1988: p. 34).

The large amounts of charred grain and crabapples suggest that the house burnt down. The grain and apples are almost certainly food stores which could not be retrieved. The occurrence of grain and apples in post-holes and wall trench must be due to later subsidence when the remaining unburnt timber rotted away.

Four charcoal samples from the wall trench, all *Quercus*, were submitted for conventional dating in Groningen. The results are as follows:

GrN-14713	5105± 45 BP
GrN-15386	5005± 25 BP
GrN-15387	4880±110 BP
GrN-16643	5085± 25 BP

Two samples of charred grain from the post-hole inside the house were dated in Oxford by AMS:

OxA-1476	4890±80 BP
OxA-1477	4840±80 BP

As these two samples are identical in terms of material, own age and context, the mean date has been calculated and used in the following discussion. The mean is 4865±57 BP.

These dates were calibrated with the CAL15 program (van der Plicht, 1993), resulting in the following ranges:

GrN-14713	1σ	3966-3930 and 3874-3808 cal BC
	2σ	3978-3892 and 3890-3796 cal BC
GrN-15386	1σ	3902-3882, 3802-3766 and 3732-3720 cal BC
	2σ	3926-3916, 3914-3876 and 3808-3710 cal BC
GrN-16643	1σ	3948-3932 and 3874-3810 cal BC
	2σ	3956-3902 and 3882-3800 cal BC

GrN-15387 has not been calibrated because of its large standard deviation.

OxA-1476/1477	1σ	3706-3628 cal BC
	2σ	3780-3614 and 3596-3516 cal BC

The dating of the construction of Tankardstown 1

Experiments have shown that 'green' oak can easily be split into 5 cm thick planks (Pleyer, 1991). It is likely that planks of this type were used for the construction of the wall of Tankardstown 1. The excavation showed clearly that these planks had been charred on the outside but whether this was done by the builders prior to construction to preserve the wood, or whether it was the result of the fire which destroyed it, is irrelevant to this discussion. In either case, the charcoal from each plank is made up of a mixture of older and younger material, its composition depending on the position of the plank in the tree trunk. For building purposes, trees of 150-200 years were probably used, from which the sapwood (about 30 years) would have been removed. The 'old wood effect' of the planks is about 60-110 years (without the missing sapwood), depending on the way in which the planks were actually made. With older trees, the 'old wood effect' is more pronounced. Calibration suggests that the wood samples of the wall should be dated between 3880 and 3800 cal BC. Taking the 1σ ranges into account, a date around 3800 cal BC is more likely. Visual examination of the curve also suggests this (fig. 3). Taking the 'old wood effect' and the removal of sapwood into account, the actual construction of the house must lie after this date, between c. 3710 and c. 3660 BC.

The date of the grain cache and the destruction of Tankardstown 1

The charred grain from the post-hole inside the building probably dates from the last year of the house's use. Grain has an own-age of one year, and can therefore be considered as free of 'old wood effect'. The calibrated mean age ranges are 3780-3614 and 3596-3516 cal BC at the 2σ level. The highest probabilities are in the 3706-3628 cal BC range.

The construction date of the house can be placed between c. 3710 and c. 3660 cal BC. If the house had a maximum life of about 30 years, the fire which destroyed the building must have occurred between c.

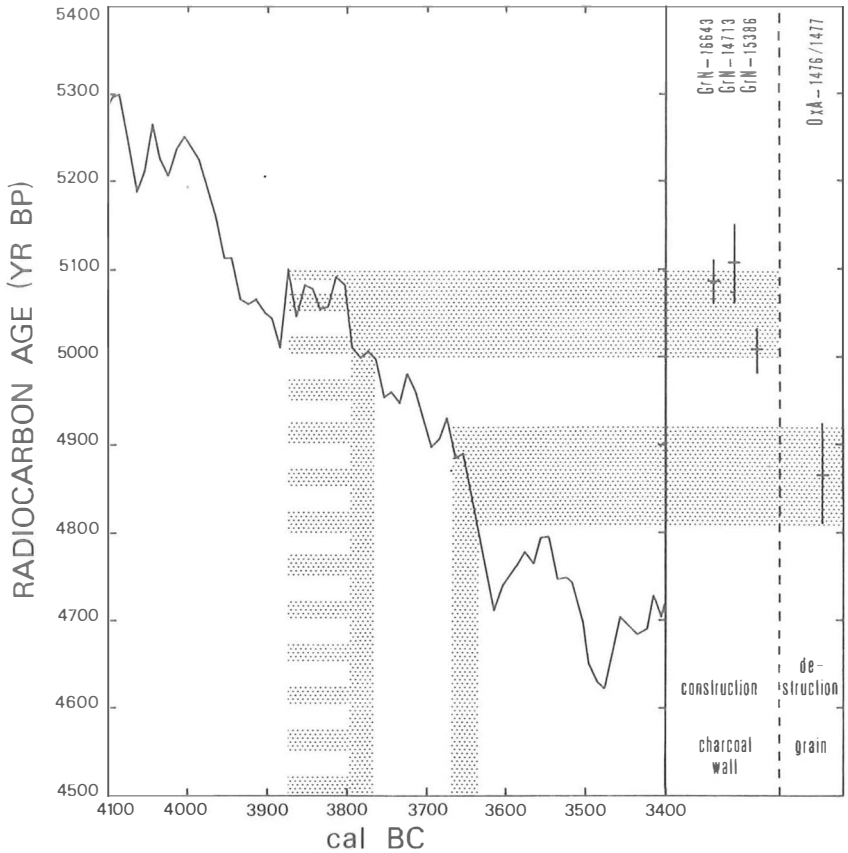

Fig. 3. Wiggle matching of charcoal and grain dates for house 1 at Tankardstown. The difference in age between charcoal and grain dates can be explained by the own age of the wood, missing sapwood and the period of use of the house. The charcoal samples represent the moment of construction, the grain samples the moment of destruction of the house. Drawing B.A.I., J.H. Zwier.

3680 and c. 3630 cal BC. This coincides with the 1σ range of the grain date.

Implications for the dating of earlier Neolithic pottery in Ireland

The pottery associated with House 1 at Tankardstown is undeveloped Western Neolithic pottery, undecorated with unaccentuated rims and carinations and open necks. The consistent style and its limited quantity indicate a short period of use. The age brackets of c. 3710-3660 and c. 3680-3630 cal BC receive some confirmation from other dates for Neolithic activity in the neighbourhood. Three forms of burial rite are known to have been practised during the early Neolithic, deposition in Portal tombs (e.g. Poulnabrone, Co. Clare), unprotected crouched inhumations accompanied by Western Neolithic pots of simple and partially developed type and/or fragments of polished stone axes (Rathjordan, Lough Gur Site C and Lough Gur 1989, Co. Limerick, Kilgreany A and B, Co. Waterford), and the more complex Linkardstown type burials accompanied by highly decorated pottery derived from the Western Neolithic group (Brindley & Lanting, 1989/1990). Although the latter two traditions overlapped to some extent, the typology of the accompanying pottery indicates that the unprotected burials appeared earlier than the Linkardstown type burials. Dates of about 3600 cal BC or earlier for the Kilgreany and Lough Gur burials have been suggested, and dates of 3550 cal BC and later for Linkardstown type burials. The pottery which occurs at Tankardstown 1 belongs to a similar stage of development as the earlier group and very probably to a primary stage within that group. It is therefore extremely unlikely that the actual date of Tankardstown 1 falls after 3600 cal BC when the ornate developed form of Western Neolithic pottery had developed.

Three other early Neolithic structures have been discovered and radiocarbon dated. The dates and calibrated ranges (2σ) are given below.

The well-preserved foundations of a second rectangular building (Tankardstown 2) were found c. 20 metres northwest of Tankardstown 1 (Gowen & Tarbet, 1988). Grain samples will be dated by AMS in Groningen in the near future.

4995±20 BP GrN-16557 E372:332 Charcoal from wall slot
3898-3885 or 3798-3710 cal BC

5070±20 BP GrN-16558 E372:334 Charcoal from wall slot
3948-3899 or 3884-3799 cal BC

Portion of the slot trench of another structure was also discovered at Pepperhill, Co. Limerick in advance of pipe laying (Gowen, 1988: pp. 44-50).

4860±70 BP GrN-15476 Charcoal from wall slot
3790-3506 or 3410-3382 cal BC

Both structures were associated with early and undeveloped forms of Western Neolithic pottery.

A fourth rectangular house of the same type was discovered at Newtown, Co. Meath (Gowen & Halpin, 1992).

5033±42 UB-3521 Charcoal from wall slot
3948-3760 or 3744-3712 cal BC

4978±32 UB-3522 Charcoal from wall slot
3906-3878 or 3804-3696 cal BC

4996±39 UB-3568 Charcoal from pit
3940-3862 or 3816-3698 cal BC

5059±31 UB-3569 Charcoal from pit
3948-3790 cal BC

All these buildings have produced samples from similar contexts i.e. charcoal from oak planks found in wall slots. In each case an own age for the wood of c. 90-140 years can be assumed, placing the date of construction sometime after the calibrated ranges given above. All three structures are associated with undeveloped Western Neolithic pottery. It is therefore unlikely that the buildings were constructed after 3600 cal BC or even after 3650 cal BC. This means that the younger range of the Pepperhill date can also be eliminated. Both Tankardstown 2 dates can be reconciled with the section of the calibration curve giving dates of about 3800 cal BC and suggesting a construction date of about 3700 cal BC. The Pepperhill structure has only one date with what appears initially to be a wide calibrated range. However, once the own-age of the sample and the date after which the associated pottery is unlikely to have been made are taken into account, a construction date of around 3700 cal BC also seems most probable. Both the Newtown dates can also be reconciled on the part of the curve which represents dates of about 3800 cal BC, thus suggesting a construction date around 3700 cal BC. The two samples from the pit within the Newtown house are unfortunately on charcoal of unknown age and therefore, unlike the similarly contexted samples from Tankardstown 1 cannot be used to date the use of the structure.

CONCLUSIONS

The Tankardstown 1 radiocarbon dates provide fairly closely established dates for

- The construction of the house between 3710 and 3660 cal BC;
- The destruction of the house between 3706 and 3628 cal BC;
- A well-defined date bracket for the accompanying pottery assemblage and the house format;
- A clear date for the grain cache.

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