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NOTE TECNICHE

— Radiocarbon Accelerator Dating: the work of the Oxford Accelerator Laboratory (R. E. M. HEDGES and R. A. HOUSLEY).

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Introduction

Since 1977 the technique of accelerator mass spectrometric AMS) radiocarbon dating has been developed and brought into use such that it is now functioning satisfactorily in a number of laboratories around the world. Current production of AMS dates stands at about 2000 measurements per year. In this paper we review the technique of AMS dating and outline the work, facilities and expertise of the Oxford accelerator laboratory with particular emphasis on the archaeological dating aspects. A detailed account of sampling considerations is included as a guide to potential users.

Scientific Background

This section explains how AMS dating works in general terms. For detailed technical accounts see Hedges (1981, 1985), or Hedges and Gowlett (1986).

WHAT IS ACCELERATOR MASS SPECTROMETRY?

Accelerator Mass Spectrometry is a technique developed over the last 10 years in which a tandem electrostatic accelerator is coupled to a mass spectrometric system so that isotopes at an ultra-trace level can be measured. The original and most important application has been to measure cosmogenically produced radionuclides, such as ^{10}Be, ^{14}C, ^{26}Al, or ^{36}Cl, since these have a natural abundance, relative to their common isotope, of 10^{-12} or less.

HOW DOES AMS WORK?

Any isotope is defined by its mass, M, and atomic number, Z. Mass spectrometry can, in principle, detect ions of a particular mass, but in practice the abundance of molecules or atoms with almost identical masses makes detection at levels below about 10^{-9} extremely difficult. Accelerating the ion beam to energies > 1 MeV/AMU enables multiply charged ions to be measured, eliminating molecules, and permits identification of the mass-selected particles through nuclear interactions which depend on Z. The main function of the system, then, is to filter out molecular ions and atomic ions of different mass, so that the particle flux to the final detector is reduced to a level where single particles events can be counted, analysed, and identified.

A second function of the accelerator is to provide high beam transmission. This is essential in order to obtain realistic count rates from ultra-trace elements. An equally important consideration is the generation of the negative ion beam, usually by a caesium sputter source. The characteristics of ion beams (e.g. efficiency of production) depend strongly on the particular ion in question.

WHAT IS AMS FOR?

Most *stable* isotopes tend to occur naturally in greater abundance than is suitable for AMS, so that most applications have been to measure long-lived *radionuclides*. (Shorter-lived nuclides are more easily measured through their radioactive decay). The majority of applications of AMS have been on the lighter nuclides ($M < 60$), although, for example, ^{129}I and ^{205}Pb are both studied. The main interest in the cosmogenic nuclides lies in their acting as tracers in the natural environment, in the information they provide about cosmic ray and other cosmogenic processes, and in their applications for dating.

Some application to stable isotopes has also been made. For example the relative abundance of osmium isotopes acts as a tracer for terrestrial versus cosmic origins in sediments.

The use of AMS for the general measurement of trace elements (10^{-9} to 10^{-12}) has not so far been developed, but in principle is possible.

THE OXFORD RADIOCARBON ACCELERATOR UNIT

The Oxford Radiocarbon Accelerator Unit was started in 1979 to develop and apply the new technique of Accelerator Mass Spectrometry specifically to radiocarbon dating in science-based archaeology. Support for this has come largely from the Science and Engineering Research Council. It was recognised that the thousand-fold reduction in sample size from the existing requirement promised major changes in both what is possible to date and in the general strategy of dating. Improvements in the methodology of dating are dependent on choices made in the selection of project, site, context and sample, and consideration of these choices should preferably be made *before* samples are taken.

The Unit is operated by the Research Laboratory for Archaeology (of the University of Oxford). Dating is undertaken on one AMS system, based on a 2MV Tandetron, and is dedicated to the measurement of ^{14}C. It has produced over 1000 radiocarbon dates in the last 3-4 years, of which over 90% are for research applied to archaeology. The expertise of the Unit extends from the strategic choice of archaeological dating, through the tactical selection of context and sample, chemical identification, extraction and purification of appropriate material, measurement by AMS, to the interpretation and publication of the result. The Unit also retains an interest in environmental questions relating to radiocarbon measurements, particularly to dating Quaternary events. It is prepared to develop other AMS-based techniques, when feasible, should the need arise.

Archaeological Radiocarbon Dating Research

The core of the work of the Unit has been either through its own 'in-house' dating programmes, or programmes of research undertaken in collaboration with archaeologists and scientists at U.K. Universities and Polytechnics elegible for SERC support through the Science-based Archaeology Committee. Future work, for which the Unit is increasing its dating capacity, is planned to include a wider spectrum of archaeological research, with additional funding from external sources.

Although the dating programmes are very diverse (including samples from 57 different countries, some 20 different types of material, and all archaeological periods over the last 45,000 years), their value depends greatly upon their overall coherency. To promote this, the Unit has in the past recognised certain themes as being of particular interest, and has given them priority. The themes have included: —

Study of different materials for dating, and their fractions;
Sources of contamination;
Studies of contextual and stratigraphic problems;
Correlation with other dating techniques;
Upper Palaeolithic cave sequences:
Late Palaeolithic open sites;
Development of agriculture and domestication of animals;
Late Palaeolithic fauna and artefacts (esp. in Britain);
Early man in the Americas;
Mesolithic and Neolithic skeletal remains (esp. in Britain);

A review of the results of the above projects can be found as a series of case studies in Gowlett (1987). These themes have accounted for about half of the measured dates. Other projects are envisaged in the future, whilst certain of the above themes may be continued.

Over 700 of the 1000 dates so far measured have been published as datelists in the journal *Archaeometry,* and it is the Unit's policy to publish a very high proportion of its work. The impact of AMS dating on archaeological research can be gauged, for example, in th proceedings of a Conference on this subject held in Oxford in 1985 (Gowlett and Hedges 1986).

The main strength of AMS radiocarbon dating lies in the fact that samples 1000 times smaller than previously can be dated. For example, the minimum sample size that can be dated needs to contain 1 milligram of carbon. This can usually be dated to an accuracy of ± 80 years BP, and sometimes better, if under about 10,000 years old. The maximum age of dating at present is about 40,000 years. This obviously enormously expands the scope for dating, and in particular the ability to select better samples. The selection may be for sites which previously yielded inadequate material for conventional dating, for samples with a much better association with the archaeological event of interest, or with greater inherent information, or for specific chemical or mechanical fractions from within a selected sample. In this way the general quality of archaeological dating can be greatly improved. The Unit has aimed deliberately to provide the best possible *quality* of dating, on the belief that questionable dates in the literature do more harm than good, but this effort must be matched by equally skillful and informed project, site, context and sample selection. This depends in part on the work of the Unit archaeologist, and in part on the appreciation of the new method by the archaeological community.

In general, archaeologists wishing to set up a collaborative dating programme, or merely wishing to apply for a number of accelerator dates should apply in the first place to the Unit, so that the feasibility of the project may be discussed. Assuming a given project is feasible there are several ways by which funding can be obtained, the procedure depending on the nature of the project. On the whole the Unit prefers to co-operate in dating projects which have a coherent theme of broad interest as opposed to ones concerned principally with individual sites.

Applications of accelerator dating in Italian Archaeology

A series of determinations have been run by the Oxford accelerator Unit specifically to date a number of sites in the Biferno Valley, Molise which were being investigated by Dr. G. Barker, the Director of the British School at Rome (Barker in press). Because none of the prehistoric sites were rich in charcoal, it was necessary to take advantage of the small sample size offered by accelerator dating in order to obtain a reasonable number of radiocarbon determinations. Three Neolithic sites, discovered in the lower Biferno valley were dated: B198 is near Larino, A184 is near Guglionesi, and A268, which is on the plain of Larino midway between Larino and Guglionesi. The AMS dates from Monte Maulo were as follows:

OxA-651	B198, I, 8, cow radius	6540+/−80 BP
OxA-652	B198, I, 10, cow scapula	6280+/−70 BP
OxA-653	B198, I, 10, cow bone	6210+/−70 BP

All the dates indicate occupation in the second half of the fifth millennium b.c. and come from a pit complex that is probably the remnant of a once larger site destroyed by ploughing. The pottery assemblage has very close similarities with the second phase of Impressed Wares development defined by Tinè on the Tavoliere plain to the south and named after the site of Guadone, and other similarities with the ensuing Masseria la Quercia phase, and, to the north, similarities with sites in Abruzzo such as Catignano. The Guadone phase on the Tavoliere dates to the sixth millennium b.c. and the Masseria la Quercia phase to the first half of the fifth millennium indicating considerable regional variation in the development of these styles. Catignano also dates to the second half of the fifth millennium.

The second Neolithic site, Colle del Fico produced a date:

OxA-654 A184, B 4, bone fragments 5840+/−70 BP

This site has a long pottery series, ranging from material similar to that at B198 to later Neolithic Serra d'Alto wares. The radiocarbon date is consistent with the later Neolithic wares.

The third Neolithic site, Piano di Larino, was also dated by the AMS technique:

OxA-655 A268, terrace 2, II, 2, bone fragments 5580+/−70 BP

Although the pottery was poor it was generally like the later wares at A184 and the radiocarbon date is in agreement with such a Later Neolithic interpretation.

The AMS dating technique also proved useful in dating a number of Bronze Age sites in the Biferno Valley including that of Monte Maggio, near Petrella

OxA-656	G1, V, 2, charcoal	3260+/−60 BP
OxA-657	G1, V, 3, animal bone	3040+/−60 BP
OxA-658	G1, VII, 4, animal bone	3070+/−70 BP
OxA-670	G1, VII, D3, animal bone	3050+/−80 BP
OxA-671	G1, VII, 6, D3, partially burnt animal bone	1950+/−160BP

Monte Maggio is an Apennine Bronze Age site in the middle valley (Barker 1976). Whilst OxA-671 was clearly an anomaly, perhaps because the bone was partially burnt, the remaining radiocarbon dates agreed very well with the expected age range of the last two or three centuries of the second millennium b.c., during which most of the Apennine Bronze Age sites seem to have been occupied.

Three sites with ' Subapennine ' pottery were also dated, all three dates were very consistent with the expected date of occupation at the end of the second millennium and the first two centuries of the first millennium b.c.

Masseria Mammarella -		
OxA-672	A113, I, 9, red deer antler	2990+/−80 BP
Matrice Villa, S. Maria delle Strade		
Oxa-676	740, charcoal from pit	2900+/−80 BP
Ancora, near Campomarino		
OxA-609	IX E 23 B V, charcoal	2990+/−80 BP

Finally an equally consistent series of AMS dates was obtained when the Iron age site of S. Margherita, 1 km north of Guglionesi was sampled:

OxA-673	A90 Pit B, charcoal	2500+/−80 BP
OxA-674	A90 I, 12, Pit 2, animal bone	2610+/−80 BP
OxA-675	A90 II, 5, animal bone	2480+/−80 BP

This particular locality had produced Daunian Iron Age pottery of the 7th-5th centuries BC, the same time range indicated by the radiocarbon dates.

On the whole the AMS dates obtained from the Biferno Valley, Molise have agreed well with the established chronology of the area and have shown that with careful selection good radiocarbon dates can be obtained. The Oxford Accelerator Unit is currently undertaking further work in Italy dating charred plant remains from the Early Neolithic contexts at Coppa Nevigata, near Manfredonia, and on dating the occupation of the Palaeolithic site of Petriolo, Provincia di Siena.

Non-archaeological applications of radiocarbon

The Unit has carried out radiocarbon measurements both for environmental and medical research, and hopes to expand these activities. Those interested in collaborative research, or in having measurements made on a purely commercial basis, should contact the Unit to discuss the application. At present the Unit has no facilities to make measurements on nuclides other than carbon 14.

Sample Requirements:

(i) Sampling selection.

It is imperative to pay special attention to sample selection when undertaking accelerator dating (Gillespie and Gowlett 1983, Gowlett 1984). Because of the greater choice of sample, it becomes more possible to date those samples which in themselves embody a high degree of archaeological information. For example, dating a specifically identified bone with evidence of human working has value regardless of precise context. In general, samples which rely on reliability of association for their dating value are more problematic. An example of this is a burial, where it is preferable to date the skeleton or grave goods, rather than dating pieces of associated charcoal where there is a possibility that it could be intrusive or residual. Therefore, other things being equal, the sample with greatest information should be chosen.

Reliability of context must be much more critically considered than previously. Some materials, or associations, will be much more reliable — for example, bones from articulated remains, or organic material sealed within mudbrick or fired clay — and it should be born in mind that samples of small physical size, such as seeds, single bones, or charcoal fragments, can be much more mobile than si commonly supposed. The Unit has found that one in five samples is intrusive, having moved either up or down, with the worst affected samples being single teeth and charcoal fragments. Contexts to be avoided include ditches with several recuts; areas of rodent disturbance; shallowly buried open sites, especially on sandy soils; talus slopes with loosely packed rocks; and areas of sites with signs of ancient digging.

The interplay of these two considerations with the particular dating questions relating to the project are likely to be the major determinants in the choice of sample(s). A third criterion is the susceptibility of the sample to contamination by modern carbon. This is especially important for material older than about 20,000 years, and will depend on the environment and the nature of the samples (e.g. the level of surviving collagen in bone).

In any case, the foregoing considerations underline the value of planning a sampling strategy whenever possible *before or during excavation* rather than afterwards.

(ii) Sample Size and Treatment.

The minimum useful amount of material, in general, is:

Charcoal	: 5 - 50	mg
Wood	: 20 - 50	mg
Bone	: 0.2 - 5	grams
Shell	: 50 - 100	mg

Samples should not be written on, and if preserpct bone should be chosen.

Samples should not be written on, and if preservation is essential, full details of the treatment should be given. *Some types of conservation treatment are liable to prevent a radiocarbon date from being measured.* Samples should be separately packed in well labelled containers. We recommend stout PVC or Polythene bags, or aluminium foil, or glass bottles with plastic tops. Paper, cotton wool, cork and other similar organic materials should be avoided. Contamination by handling should be avoided, but is only potentially serious for the very oldest samples.

Future prospects

In the terms of ture developments, both in general and specifically of the Oxford AMS system, there are a number of changes in the process of implementation. One promising line is the replacement of the graphite target with a carbon dioxide ion source. This would simplify the complicated and time consuming chemical preparation of graphite, reducing the workload of sample preparation and providing a good chance of reduced chemistry backgrounds. Such improvements are vital if there is to be any hope of dating samples older than 40,000 years. An associated benefit will be the ability to use even smaller samples than at present, however this carries the risk that contamination by modern carbon will become more significant unless strict control of processes and purity of chemicals are introduced.

Developments on the technical side must be paralleled by improvements on the archaeological side, both in terms of the coherence of dating strategies and on a tactical scale with strict and rigorous selection of suitable samples. The quality of a radiocarbon date is in part the quality of the measurement, but it is also dependent on the sample material, its context, and the relevance of the sample to the ' event ' to be measured. Thus in order to make full use of the potential of accelerator dating, the archaeological community will need adjust it's perceived wisdom on the suitability of certain samples for radiocarbon dating. Projects which cannot at present be done in any other way but by accelerator need to be sought out and given priority, for it is going to be through such applications that archaeology will benefit most. In conclusion, it has been shown over the past few years that accelerator dating works, the future will show the full archaeological potential of the method.

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BIBLIOGRAPHY

Datelists of Oxford accelerator dates published to present:

Archaeometry 1984, 26, 1 (Datelist 1)
Archaeometry 1985, 27, 2 'Datelist 2)
Archaeometry 1986, 28, 1 (Datelist 3)
Archaeometry 1986, 28, 2 (Datelist 4)
Archaeometry 1987, 29, 1 (Datelist 5)
Archaeometry 1987, 29, 2 (Datelist 6)

Barker, G. (1976) An Apennine bronze age site at Petrella, Molise, in *Papers of the British School at Rome* 44:133-156.

Barker, G. (in press) *The archaeology and history of a Mediterranean valley.* Cambridge: Cambridge University Press.

Gillespie, R. & Gowlett, J. A. J. (1983) Archaeological sampling for the new generation of radiocarbon techniques, *Oxford J. Archaeol.* 2 (3): 379-382.

Gowlett, J. A. J. (1987) The archaeology of radiocarbon accelerator dating, *J. World Archaeol.* 1 (2): 127-170.

Gowlett, J. A. J. and Hedges, R. E. M. (1986) *Archaeological Results from Accelerator dating,* Oxford University Committee for Archaeology Monograph Series, No. 11, Oxford.

Hedges, R. E. M. (1981) Radiocarbon dating with an accelerator: Review and preview. *Archaeometry* 23 (1): 3-18.

Hedges, R. E. M. (1981) Radiocarbon dating with an acce- *Sci. Prog. Oxford* 69: 409-427.

Hedges, R. E. M. & Gowlett, J. A. J. (1986) Radiocarbon dating by accelerator mass spectrometer. *Scientific American.* 254: 100-107.