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The Use of Radiocarbon (¹⁴C) to Identify Human Skeletal Materials of Forensic Science Interest

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ABSTRACT: The radiocarbon (¹⁴C) method is a well-known isotopic dating technique by which age can be assigned to organic materials, including human bone. Natural and anthropogenic anomalies in ¹⁴C activity in the biosphere over the last few centuries, including the presence of artificial or "bomb" ¹⁴C after 1950, can be used as an isotopic tracer to assign human bone samples with high degrees of probability to one of three temporal periods within the recent past: a Non-modern period (before about A.D. 1650) of no forensic science interest, a Pre-modern period (A.D. 1650 to 1950) of possible or potential forensic science interest, and a Modern period (A.D. 1950 to the present) of definite forensic science interest. We illustrate the use of the ¹⁴C method to assign human bone in five forensic science cases to one of these time periods.

KEYWORDS: physical anthropology, human identification, radiocarbon, dating, bone, skeletal remains

A primary concern of the coroner when dealing with skeletonized human remains is the determination whether or not the material is recent enough to be of forensic science interest. Bones that still retain odor or grease merit attention. Dried, bleached, or odorless skeletal remains present an ambiguous situation. Such remains may be as recent as several months if relevant factors (for example, temperature, location of body, size of body, animal and insect activity) favor rapid decomposition, or they may be of such antiquity that the coroner need not pursue forensic science investigation.

Bass [1] has noted the surprising lack of attention in the forensic science literature concerning the accuracy of analytical methods of inferring time since death for skeletonized human materials beyond a few months. Many studies [2-4] have focused on estimating age on an extended time scale, but these generally are not applicable to skeletal material from the recent past. The Berg [5], Knight and Lauder [6], and Castellano et al. [7] studies are among those which have evaluated biochemical and elemental constituents of recent bone to determine possible time-dependent indices that are useful for forensic science purposes. These studies suggested that elemental composition, nitrogen content, and amino acid concentration—both in terms of the number of amino acid residues and the presence of the

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amino acids hydroxyproline and proline—constitute the most useful analytical methods in distinguishing contemporary from noncontemporary bone.

Unfortunately, the available data point to significant variability in these components in bone samples of essentially identical age. Diagenetic effects, reflecting variations in mean annual temperature, bacterial, pH, ground water, and other environmental factors, can significantly influence the rate of any biochemical process. Only when bone samples derive from essentially identical biogeochemical environments can elemental and biochemical data in bone be used with any degree of confidence in making an accurate estimate of the time since death.

Several criteria might be offered to evaluate proposed methods of estimating time since death in excess of a few months for human bone. Preferred techniques would be those in which (1) the method of assigning age used is not affected by environmental variabilities, (2) the results could be critically supported on the basis of a body of well-established scientific data, and (3) analysis is relatively inexpensive and can be rapidly carried out. The first two criteria can be met by employing a method which uses a nuclear decay scheme. The best known isotopic dating technique that can be applied to bone is the radiocarbon method.

Radiocarbon Dating

Radiocarbon (^{14}C) dating is the best known and most widely used physical dating technique to assign age to organic materials. Although the method has made its major impact in prehistoric archaeological studies, it has been extensively used in a wide range of scientific fields, including biochemistry, environmental science, geochemistry, geology, geophysics, hydrology, and oceanography. There is an extensive literature which describes the basis of the method and its 40-year history of utilization (see Ref 8 for a summary of this literature).

To be able to assign an accurate age to a sample based on its residual ^{14}C activity, a series of assumptions must hold to reasonably close tolerances. There has been a great deal of research on the accuracy of the ^{14}C method in general as well as on the precision and accuracy of the inferred ages that may be expected from different types of samples derived from different geophysical and geochemical environments. Four decades of ^{14}C studies have demonstrated the general validity of ^{14}C -inferred ages on a worldwide basis. However, these same studies have also documented significant anomalies in ^{14}C data for some periods and sample types. These well-studied ^{14}C anomalies, which have been documented for the recent past, allow for the application of ^{14}C technology outlined in this paper [9].

Radiocarbon Dating of Bone

As we have noted, the most intensive use of ^{14}C data is associated with the dating of a wide range of organic materials associated with prehistoric archaeological contexts. However, in our discussion, we will focus our attention on human bone and utilize ^{14}C concentrations to distinguish modern from nonmodern samples.

There has been much discussion over the last decade concerning the problems in the ^{14}C dating of bone derived from paleoanthropological and archaeological contexts [9]. The focus of this literature is on determining which chemical fraction of a bone sample has been least affected by contamination as the result of being subjected to diagenetic effects over relatively long time periods. The studies reported in this paper are concerned with the dating of bone from the relatively recent past. The types of organic contamination which can affect the ^{14}C content of modern bone have generally been well studied. Fortunately, when proper pre-treatment techniques are carefully applied (such as isolating an organic carbon fraction from the bone), the accuracy of the ^{14}C analysis is generally very satisfactory, particularly on bone of relatively recent age.

Variations in Modern Radiocarbon Concentrations

Figure 1 plots variations in the ^{14}C activity between 1900 and 1980 in a series of terrestrial biospheric and atmospheric samples [11, 12]. The period from 1900 to 1954 is documented by the ^{14}C activity in a series of tree-ring samples. These samples can be used to measure decade-by-decade variations in ^{14}C activity. This is due to the fact that each tree ring reflects the ^{14}C activity in the atmosphere adjacent to the tree as averaged over a single season of growth. Data for the period from 1959 to 1982 are based on the ^{14}C activities measured in a series of samples of tropospheric carbon dioxide collected on an annual basis.

The ^{14}C values plotted in Fig. 1 are expressed with respect to one of the ^{14}C modern reference standards which provides the assumed ^{14}C concentration of contemporary, living materials in the biosphere. A ^{14}C age of "0 B.P." would be assigned to a sample which, within experimental error, exhibited a ^{14}C content equal to that of a modern reference standard which presumably reflects the contemporary ^{14}C equilibrium concentration. The assumed equilibrium ^{14}C activity has been set at zero in Fig. 1. Deviations from the assumed equilibrium concentration are expressed in percent above or below the activity of the modern standard. To convert percentage deviation to an approximate estimate of age for the recent past,

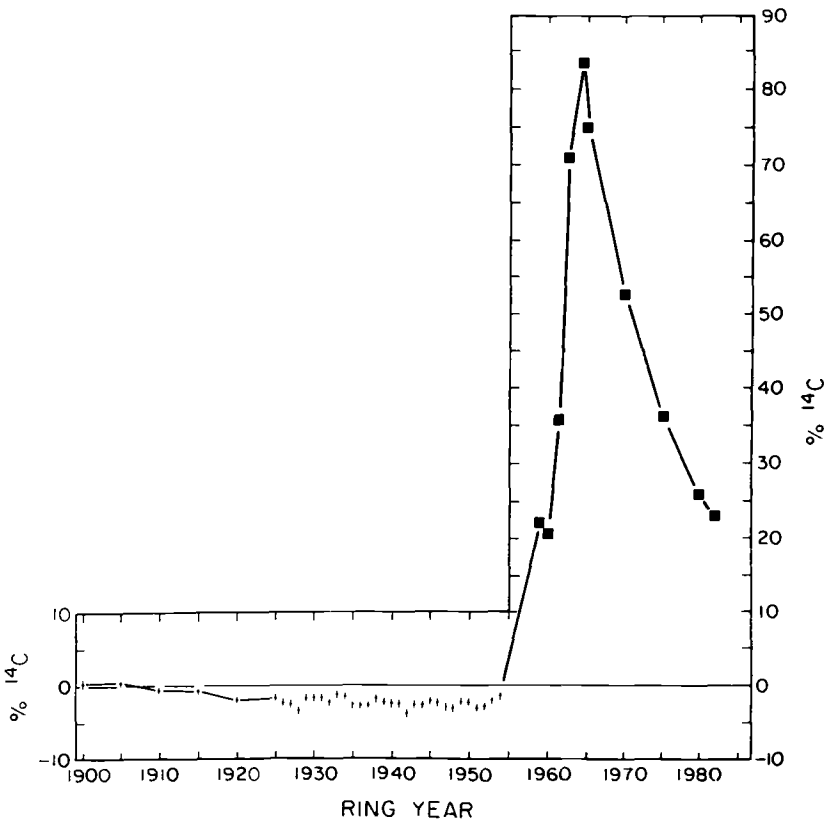


FIG. 1—Fossil [Industrial or Suess] (1900–1950) and Atomic Bomb [Libby] (1950–present) Effects on recent ^{14}C activities: ^{14}C activity in tree rings (1900–1955) and tropospheric carbon dioxide (1955–1983) expressed as percentage above or below 0.95 NBS oxalic acid standard (old). Tree-ring ^{14}C data (solid dots) from Ref 10. Tropospheric ^{14}C data (solid squares) taken from Ref 11; design of figure taken from Fig. 2.14 in Ref 8.

one would multiply the percent variation by 80 years. For example, a deviation of -2% from the assumed equilibrium, modern concentration would be equivalent to an age offset of about 160 years.

An examination of the data in Fig. 1 reveals that there have been two modern perturbations in ^{14}C activity over the last eight decades—both of which are *anthropogenic*, that is, caused by human agency. The period from about 1910 to 1950 is characterized by a *decrease* in ^{14}C concentrations in all terrestrial organics; the maximum decrease was about 3%. This depletion has been traced to the combustion of fossil fuels including coal, oil, and natural gas. Because of their great geological age (that is, hundreds of millions of years), fossil fuels contain no detectable amounts of ^{14}C and are referred to as “dead” in terms of their ^{14}C activity. The combustion of fossil fuels reduces ^{14}C concentrations since it adds carbon dioxide containing no measurable ^{14}C to the atmosphere. This phenomenon was first documented systematically in the early 1950s by Hans Suess, then at the U.S. Geological Survey [12]; consequently, the *fossil fuel effect* is sometimes referred to as the Suess effect.

Beginning about 1950, another set of events dramatically reversed the slow downward trend from the modern atmospheric and terrestrial equilibrium ^{14}C concentration. Beginning in the early 1950s, the testing of nuclear and thermonuclear weapons injected large amounts of artificial or “bomb” ^{14}C into the atmosphere [13]. This process resulted in a phenomenon known as the *atomic bomb effect*, also sometimes referred to as the Libby effect, named for the discoverer of the ^{14}C method, Willard F. Libby. Due to “bomb” ^{14}C , the ^{14}C activity in terrestrial organics rapidly rose to about 85% above the contemporary or modern reference level by 1963. An international agreement in 1963 halted atmospheric nuclear testing by most nations. This allowed ^{14}C to begin the process of reestablishing a new atmospheric ^{14}C equilibrium. It has been calculated that if no further artificial ^{14}C is produced, then a new equilibrium condition will be established at about 3% above the old, pre-1950 levels. However, by this time, the combustion of fossil fuels will have probably compensated for this increase if the present rate of fossil fuel consumption is continued.³

For the period before about 1900, a third phenomena has been identified that complicates the usefulness of ^{14}C data to make precise and specific age assignments over the last 300 years, prior to the period of the Atomic Bomb and Suess Effects. Figure 2, based on Stuiver

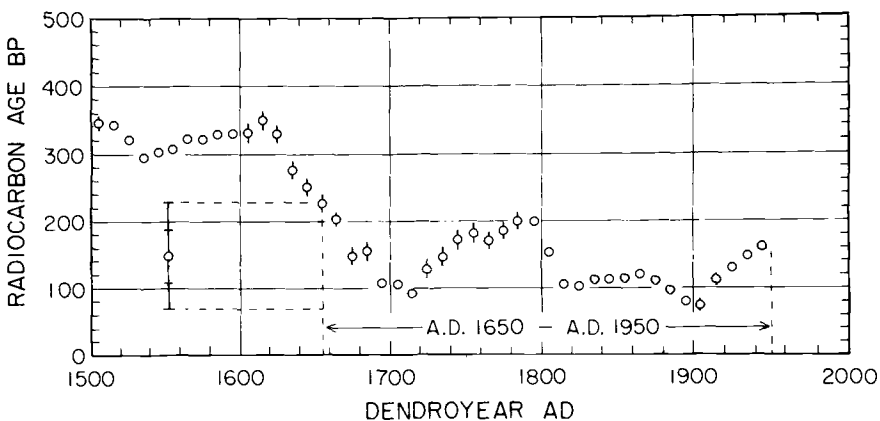


FIG. 2—Recent de Vries and Suess effects: 1500 to 1950. Data taken from Fig. 2a in Ref 14; design of figure taken from Fig. 2.13 in Ref 8.

³M. Stuiver, 1987, personal communication.

[14], presents the ^{14}C age of tree rings compared with their known age based on dendrochronological (tree-ring dating) criteria for the period from 1500 to 1950. Note that the ^{14}C data for the period of the Suess effect are represented in a different manner in Figs. 1 and 2. Figure 1 presents these data in terms of a deviation in ^{14}C activity *in percent* above or below an assumed modern equilibrium ^{14}C concentration in the biosphere. Figure 2 represents the actual inferred ^{14}C age *in years* before the present (B.P.) of known-age tree-ring samples. The 1900 to 1950 interval is characterized by a slow depletion in ^{14}C activity in the range of -1 to -3% . (Suess Effect period). These same data are represented in Fig. 2 as an increase in apparent ^{14}C age from about 80 years in 1900 to about 240 years in 1950. We previously noted that each 1% change in ^{14}C activity translates into a age difference of about 80 years for the recent past.

Figure 2 also represents a third type of ^{14}C anomaly that is revealed from an examination of the ^{14}C content of a series of known-age tree rings. These measurements have demonstrated that there have been periodic episodes of relatively rapid, short-term variation in terrestrial biospheric ^{14}C activity. For the period from about 1500 to 1900, Fig. 2 identifies several anomalies in the ^{14}C record. These anomalies are sometimes referred to as *de Vries Effects*, named for the pioneering Dutch researcher, Hessel de Vries, who first called attention to them. A number of causes for the de Vries Effects have been proposed. Relatively rapid modulation of the cosmic-ray flux by one or more components of the magnetic field of our sun has been suggested as probably playing an important role [15]. It might be noted that it has also been determined that the short-term or de Vries Effects are superimposed on a longer-term variation in ^{14}C activity. This is sometimes labeled the "major trend" in ^{14}C secular variation effects and has been linked to temporal variations in the intensity of earth's dipole field. It has been identified in ^{14}C measurements in tree rings spanning the last 8000 years. This major trend in the secular variation in ^{14}C activity is of primary interest to archaeologists, geophysicists, and others interested in Quaternary environmental changes, but does not concern the application being discussed here.

Combining the data in Figs. 1 and 2, we can identify three time segments which can be distinguished for the period of the recent past based on ^{14}C data: (I) a pre-1650 period, (II) a 1650- to 1950-period, and (III) a post-1950 period. Table 1 summarizes both the forensic and ^{14}C characteristics of these three temporal intervals. For our purposes, materials from Period I have been labeled as "Non-modern," those from Period II as "Pre-modern," and Period III as "Modern." Figure 2 illustrates why routine ^{14}C data cannot be used to assign a specific age equivalent in samples derived from the period from A.D. 1600 to 1955 on the basis of their ^{14}C content. A conventional ^{14}C value of 150 ± 50 ^{14}C years B.P. has been indicated with both a one and two sigma range in the experimental (statistical) variance [16].

It is clear from the ^{14}C /dendrochronological data that the actual age of materials exhibiting apparent ^{14}C values in the interval from about A.D. 1650 to 1950 cannot be assigned an actual age to better than about ± 300 years. The boundary between Periods I and II as defined in Table 1 will be somewhat indistinct, depending on the statistical variance assigned

TABLE 1—Modern periods distinguishable on the basis of ^{14}C data.

| Period | Forensic Science Characteristic | Interval | ^{14}C Characteristics |
|---------------|------------------------------------|----------------------|--|
| I Non-modern | no forensic science interest | before A.D. 1650 | period antecedent to the most recent de Vries Effect variation |
| II Pre-modern | possible forensic science interest | A.D. 1650- A.D. 1950 | period of the most recent de Vries Effect variation |
| III Modern | definite forensic science interest | A.D. 1950- present | period of bomb ^{14}C |

to the ^{14}C analysis. However, the rapid rise in ^{14}C activity beginning in 1955 results in a boundary between Periods II and III which is relatively distinct due to the fact that the ^{14}C signal for the period from 1950 to the present—that is, the presence of “bomb” ^{14}C —is unambiguous and well-documented.

Application to Forensic Science Cases

We have measured the ^{14}C concentrations in organic fractions prepared from five samples of human bone in which various coroner agencies have sought forensic anthropological consultation for the determination of the time since death. Using these cases, we have assessed the practical utility of the suggested approach of distinguishing non- or pre-modern from modern human bone samples on the basis of their ^{14}C concentrations. Table 2 identifies and summarizes the data obtained on these samples and assigns the bone sample to one of the three periods defined in Table 1. For Cases 1 through 3, the ^{14}C data are expressed in age expressions in ^{14}C years before present (with A.D. 1950 set as 0 B.P.). For Cases 4 and 5, ^{14}C data are expressed in percentage above modern (with 0% of modern the same ^{14}C activity as materials living in A.D. 1950 or of 0 B.P. age). These values register the amount of “bomb” ^{14}C in the bone.

Stable isotope data (^{13}C) were obtained for these bone samples to insure that no serious fractionation effects were present. All ^{13}C values are in the expected range. As is the convention in expressing ^{14}C results, ^{14}C values presented in Table 1 have been normalized to -25 per mil ^{13}C . The geophysical basis on which this normalization procedure is carried out is summarized in Refs 8 and 16.

Case 1 consists of adult human bones from what appeared to be an articulated burial recovered during excavation for a swimming pool in the backyard of a residence in Beverly Hills, Los Angeles County, California. Figure 3 illustrates the appearance of the bone fragments at the time of their recovery. They were presumed to be prehistoric due to their extreme lightness and porosity as well as the lack of any odor. This conclusion is supported by the ^{14}C data which show it to be a non-modern bone of no forensic science interest with an age of about 2000 years.

Case 2 concerned human bone which had been subjected to fire found as a result of a surface collection conducted by archaeologists during April 1985 near La Quinta, Riverside County, California. The reason for the analysis was that the area where the bone was found contained both archaeological remains (ceramic fragments, charcoal, and lithic flakes from

TABLE 2—Radiocarbon data on human bone of possible forensic science interest.^a

| Case | Sample | Locality in California | $^{14}\text{C}^b$ (Age/% modern) | $^{13}\text{C}^c$ | Period ^d |
|------|----------|------------------------|-------------------------------------|-------------------|---------------------|
| 1 | UCR-2125 | Beverly Hills | 2150 ± 60 | -26.21 | I |
| 2 | UCR-2079 | La Quinta | 720 ± 170 ^e | -20.29 | I |
| 3 | UCR-2160 | Fluor | 230 ± 50 | -19.95 | II |
| 4 | UCR-1800 | Indio | +9.1 ± 1.2% | -16.70 | III |
| 5 | UCR-2159 | Griffith Park | +43.8 ± 0.9% | -20.96 | III |

^a ^{14}C analysis carried out on total acid-insoluble organic fraction of bone samples.

^bFor Cases 1 through 3, ^{14}C data expressed in terms of ^{14}C years B.P. with A.D. 1950 = 0 B.P. Error is one sigma counting error of sample, background, and standard expressed in years. For Cases 4 and 5, ^{14}C data expressed in terms of percentage above modern reference standard, 0.95 NBS oxalic acid (old). Error is one sigma counting error of sample, background, and standard expressed in percent.

^cValues expressed in per mil with respect to PDB standard. Error on values ± 0.02 per mil.

^dSee Table 1.

^eRelatively large statistical error assigned to UCR-2079 reflects small amount of bone available for analysis.



FIG. 3—Case 1: bone fragments recovered from Beverly Hills, Los Angeles County, California excavation.

a presumed late-prehistoric cultural context) as well as modern trash items. There was no physical evidence that would conclusively determine the age of the bone. The ^{14}C analysis clearly indicates that the bone is non-modern and, with an age of about 700 years, of no forensic science interest.

Case 3 involves the recovery of skeletal material from one burial during archaeological salvage work being conducted at the Fluor site near Irvine, Orange County, California. The bulk of the skeletal materials—about 20 burials—appeared to be late prehistoric (A.D. 500 to contact) on the basis of the cultural materials found with the burials. The prehistoric materials were uniformly deteriorated, fragile, and dark in color. By contrast, Burial 3E, a subadult, appeared to be much more recent on the basis of morphological appearance. The bones showed a finer, smoother texture. The breaks in the bone appeared to be caused by mechanical stress and not as a result of deterioration. Finally, the color of the bones was much lighter than was the clearly prehistoric materials. With an age of about 200 ^{14}C years, there is some ambiguity as whether the burial should be assigned to Period II or the later portion of Period I. However, we conclude that the bone clearly does not constitute a situation where forensic attention is required.

Case 4 involved skeletal remains found in a desolate area of the Salton Sea near Indio, Riverside County, California. A single individual was represented by fragments of cranium, mandible, and long bone. The humeri showed extensive development of the deltoid muscles. The extreme robusticity, along with moderately heavy dental attrition, led to the possibility that the individual might be of some antiquity. However, the ^{14}C activity of the sample indicates that it contains bomb ^{14}C , assigning it to the early part of Period III, and thus of definite forensic science interest.

Case 5 involved human skeletal remains found in a plastic bag underneath a makeshift

concrete marker in a relatively inaccessible location high on a hill in the Griffith Park area of the City of Los Angeles. A crude inscription on the marker bore the legend "remains unfortunate prospector may he rest in peace 1983." The skeletal materials consisted of the partial remains of at least two young males, ages 15 to 17 and 20 to 26. The individuals had been buried at one or more locations elsewhere and then reinterred underneath the concrete marker. Only one skull was present (Fig. 4), which might go with either male or even may represent a third individual. Morphologically, the skull appeared to be Caucasoid. The nasal bones were projecting and the nasal aperture was narrow. The skull was orthognathous and the zygoma were not prominent. The dentition was not helpful in assessing time since death. No dental work was present, but many modern individuals of this age do not show dental work. Only slight attrition was present. Time of death would be in excess of several years since bones exhibited no odor. None of the bones appeared to have been bleached by the sun. The ^{14}C evidence assigned the bones clearly to our Period III and thus of definite forensic science interest.

Discussion

To determine whether an otherwise undocumented skeletonized human remains merit additional forensic science inquiry, ^{14}C analysis can be used to assign bone samples in most cases with relatively high degrees of probability to one of three temporal periods within the recent past. In our introduction, we noted three criteria that might be applied to evaluate any method of assigning age to modern bone. For the ^{14}C method, criteria (1) analysis not affected by environmental variables and (2) general accuracy of results supported by a recognized corpus of scientific data are, in our view, met and exceeded. Criterion 3, which involves costs along with relatively rapid availability of analytical results, may be a problem in some

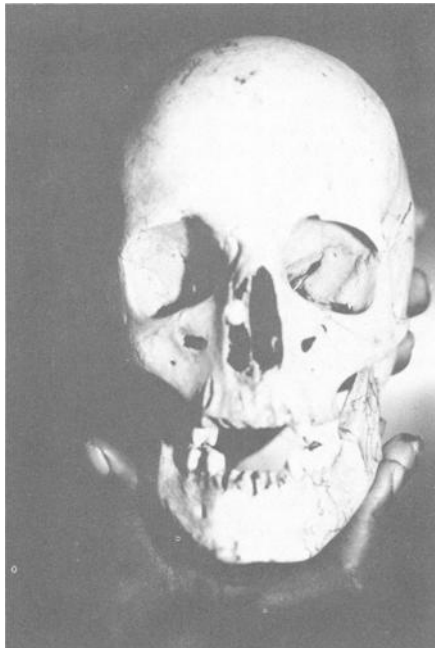


FIG. 4—Case 5: skull recovered from underneath makeshift concrete marker in Griffith Park area, Los Angeles County, California.

cases with the ^{14}C method. The current typical cost of a single, conventional ^{14}C analysis is in the range of \$300 to \$400. The typical length of time required to obtain a ^{14}C analysis ranges from several weeks to several months depending on technical factors. However, with special arrangements with certain ^{14}C laboratories, it is possible to complete a ^{14}C determination in less than 21 days with additional cost. Also note that the ^{14}C method is a destructive technique. Skeletal remains submitted to the laboratory will be consumed in the analysis.

The amount of bone material required for a ^{14}C analysis will be determined by (1) the organic carbon content of the bone and (2) the type of instrumentation used to undertake the ^{14}C analysis. For conventional or decay counting of ^{14}C , from 1 to 5 g of carbon are generally required. Since the organic carbon content of contemporary bone is typically on the order of 10%, it would be necessary to sacrifice from 10 to 50 g of bone for each analysis. Fortunately, the newly introduced accelerator mass spectrometry (AMS) method of measuring ^{14}C is capable of obtaining ^{14}C analysis on milligram amounts of carbon, and thus the required bone sample size can be reduced into the range of a few grams [17]. Unfortunately, the current cost of conducting AMS ^{14}C analysis is approximately double that of conventional decay counting, although that cost differential is expected to be reduced over the next decade.

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