

## BOMB RADIOCARBON DATING OF ANIMAL TISSUES AND HAIR

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**ABSTRACT.** Initially, radiocarbon dating by bomb  $^{14}\text{C}$  was used to check vintages of wine and whisky and to estimate the turnover times of carbon in various biological tissues. However, this technique has never been widely used for routine dating, although it has a wide field of application in geriatric medicine and forensic investigations. Fifteen years' experience in this field has shown the potential and limits of this technique. Taking into account the decisive biological factors, such as growth and aging, a complicated picture is obtained. Recent human bones cannot be dated with a constant precision. Despite an incomplete understanding of the process of incorporation of  $^{14}\text{C}$  into human bones, the present dating technique is still more precise than most estimates by geriatric experts, for conventional  $^{14}\text{C}$  dating follows that  $^{14}\text{C}$  dates of bone collagen represent the years of the termination of puberty rather than those of death.

Another application is the identification of furs of illegally hunted animals on the "Red List of threatened species" of the World Conservation Union (IUCN). For court cases, the year the animals were killed must be precisely determined. Due to the long and variable turnover time of more than one year of leather hair is the best dating material for animals.

### INTRODUCTION

Bomb radiocarbon dating of biological samples with ages of less than 50 years has not yet been widely used, although fields of application exist in forensic medicine and for the detection of illegally imported pelts of animals on the "Red List". This dating method is based on the  $^{14}\text{C}$  bomb tracer in the atmosphere and biosphere produced mainly by nuclear tests in the 1950s and 1960s. The atmospheric  $^{14}\text{C}$  input function is precisely known (D Harkness, personal communication 1999; Levin et al. 1985; Levin and Kromer 2000). Starting at 96.5 pMC in 1953, the specific  $^{14}\text{C}$  activity of atmospheric  $\text{CO}_2$  was twice as high in 1963–1964, at the peak of the nuclear bomb tests. It dropped exponentially after the atom test ban treaty and was about 110 pMC in 1999 (Figure 1). Plants and animals, as part of the biological cycle, have taken up this bomb  $^{14}\text{C}$  via the food chain.

One reason for the limited application of this dating method may be that there is little known about carbon uptake and residence time in animal tissues. There seem to be differences within and between organs of the animals as well as differences between individuals.

In forensic medicine, the collagen fraction of bone (Taylor 1982) is usually the most easily accessible dating material. Only in exceptional cases, material with a short carbon residence time of about one year (hair, nails) is available. Fresh bone contains about 1% organic carbon (TOC). The most accurate and reliable  $^{14}\text{C}$  ages are, however, obtained from the amino acids (Gillespie and Hedges 1983) extracted from collagen. The  $^{14}\text{C}$  value of the inorganic carbon fraction of bone is not usually suitable (e.g., Hassan et al. 1977).

In the 1960s, L'Orange and Zimen (1968a, 1968b) tried to calculate the annual radiation dose from bomb  $^{14}\text{C}$  for individual humans and estimated the biological half-life of  $^{14}\text{C}$  in collagen to  $22 \pm 2$  yr using an exponential model. The authors assumed that the food consisted of cereals (70%) with an age of one year and meat with an age of two or more years.

Stenhouse and Baxter (1979) also tried to calculate the annual radiation dose, and developed a realistic model for  $^{14}\text{C}$  uptake in animal tissues. They assumed a linear increase in  $^{14}\text{C}$  activity until the end of the puberty (about 19 years) and a variable composition and different ages of food. They used a mean biological half-life of carbon in collagen of 30 yr.

Taylor et al. (1989) were the first to estimate the “deposition period” of skeletal remains and the year of death using bomb  $^{14}\text{C}$  in bone collagen. This deposition period is defined in forensic medicine as the time between the finding of skeletal remains and the death of the individual. Taylor et al. concluded that only three ranges of the year of death can be distinguished by bomb  $^{14}\text{C}$ : before 1950, between 1950 and 1965, and afterwards.

The last work on this subject was by Lux and Rösing (1996). They presented  $^{14}\text{C}$  results for bone collagen from 48 samples of human skeletal remains. The years of birth and death of the corresponding individuals were known for all of these samples. The authors concluded that the period of deposition of skeletal material can be determined with a precision of  $\pm 2.5$  years by means of bomb  $^{14}\text{C}$ . We carried out the corresponding  $^{14}\text{C}$  analyses and extended this study to leather and hair of pelts of animals from the wild and from zoos. The outcome of our interpretation of these results is presented below. Hair allow the most reliable determination of the death of an animal.

### **SAMPLES AND TECHNIQUES**

Two kinds of materials were dated on the basis of bomb  $^{14}\text{C}$ . The first set of samples was taken from 48 human skeletal remains for which the dates of birth and death of the individuals were known. In a few cases information about the health history of the individuals was available. The birth dates range from 1901 to 1979 and the death dates from 1961 and 1992. The bone samples were degreased in a Soxhlet apparatus before the collagen was extracted by the method of Longin (1971). Afterwards  $\text{CO}_2$  was produced as counting gas by combustion. In addition, from 10 samples the apatite fraction also was studied. The specific  $^{14}\text{C}$  activity of these 58 samples was measured with proportional counters that require as little as 25 mg C.

The second set consisted of 19 samples taken from pelts of various carnivores (lion, jackal, panther, fox, wolf, marten) and herbivores (bear from Malaysia and from the Grand Chaco, beaver, bison, Indian deer, ibex, chamois, gorilla, gazelle). Fifteen samples were cut from pelts stored in the Zoologische Staatssammlung in Munich. Eight animals from those were imported alive and kept in the Munich zoo, four animals were born in this zoo and the others were shot by hunters. The year of death between 1962 and 1992 was always exactly known. The year of birth was roughly estimated and ranges from 1941 to 1968; it is always before the year the animal was imported between 1947 and 1992. Two samples were from wild pigs and one from a red deer shot by hunters in Lower Saxony. The  $^{14}\text{C}$  value was measured separately for the leather and hair of each sample after burning without any pretreatment.

### **RESULTS**

The  $^{14}\text{C}$  values for the human bone samples will be published elsewhere by B Lux. A representative selection of data from the collagen of human bones is shown in Figure 1. The corresponding individuals died between 1901 and 1944, 1955 and 1964, and 1975 and 1984.

The  $^{14}\text{C}$  values for the leather and hair samples are plotted (Figures 2 and 3, respectively), above the year of death of the corresponding individuals. All information can be requested from Section 3 of the Institute for Joint Geoscientific Research, Hannover.

### **INTERPRETATION**

Because different aspects must be taken into consideration for human collagen and pelts, they will be discussed separately.

### Radiocarbon Values of Human Bone Collagen

I used the model of Stenhouse and Baxster (1979) for the interpretation of the  $^{14}\text{C}$  values for collagen but take into consideration the different rates of growth of females and males. The curve for the input of  $^{14}\text{C}$  into atmospheric  $\text{CO}_2$  was provided by Douglas Harkness (personal communication 1999), Glasgow, which fits well with other available data (Levin et al. 1994; Levin and Kromer 2000). According to the mentioned model, the uptake of carbon occurs in two phases:

#### *The Growth Phase*

In humans, the main uptake of carbon occurs between birth and the end of puberty (at about 19 years old). Bone carbon increases by about 5% per year. It is not considered that the  $^{14}\text{C}$  activity may be unevenly distributed in the bone collagen and may change along the growth direction of the bones.

#### *Aging Phase*

During the aging phase a steady-state exchange of carbon in the body is assumed between the radiocarbon ingested in the food and the loss by radioactive decay. During this phase the specific  $^{14}\text{C}$  activity in the body adjusts to that of the atmospheric  $\text{CO}_2$  more slowly than during the growth phase. The biological mean residence time ( $\text{MRT}_{\text{biol}}$ ) of the carbon is calculated from the carbon exchange rate  $r$ :  $\text{MRT}_{\text{biol}} = 1/r$ .

The modeled relationship between the year of death and the specific  $^{14}\text{C}$  activity of human bone collagen is shown in Figure 1 for a number of years of birth. The lowest curve is representative for all individuals who became 19 years old in 1954 (when the  $^{14}\text{C}$  activity in the atmosphere began to increase) or before (i.e., date of birth before 1935). It was found that the best fit of the curve to the  $^{14}\text{C}$  data is obtained for a carbon exchange rate of 1.5%, corresponding to a biological MRT of 67 yr.

As the plotted curves are for specific years 10 years apart, and the  $^{14}\text{C}$  values are for most years in-between, most of the  $^{14}\text{C}$  values do not fit the modeled curves. Large deviations may have the following causes:

Old individuals are sometimes treated with medicine to reactivate the growth of bone. This treatment is apparently successful, which is why high  $^{14}\text{C}$  values in the collagen were found (Lux and Rösing 1996). In one case, this treatment is certified. The health history of individuals and their medical treatment is normally unknown for skeletal remains.

The specific  $^{14}\text{C}$  activity may not be uniformly distributed in all of the bones in the body and may change along the growth direction. There was no possibility to collect corresponding samples to check this hypothesis. Another example is that collagen from skulls of old individuals did not contain any bomb  $^{14}\text{C}$  (K van der Borg 2000, personal communication).

The specific  $^{14}\text{C}$  activity of collagen depends on the kind of food eaten. Vegetarians may eat food with an age not older than one year while non-vegetarians eat meat that may have a biological MRT of several years. It is not usually possible to evaluate the individual food history even of known persons.

The weak relationship observed between the  $^{14}\text{C}$  value of atmospheric  $\text{CO}_2$  and that of the bone collagen is indeed an oversimplification and does not allow precise dating for forensic medicine. The following example demonstrates this. The measured  $^{14}\text{C}$  value of the collagen fraction of a bone was  $115 \pm 0.5$  pMC. The skeletal remains were found 1995. We use the 2-sigma criterion. The possible ranges of the years of death are shown in Figure 1 and compiled in Table 1. For an anthropologically

determined age at death of 20–30 years, the year of birth must have been between 1943 and 1948 and the burial time was 22–27 years. The precision is about three years.

This example shows that each burial time must be individually estimated. It is the more precise the more accurately the age at death of the individual is known. The burial time can, however, seldom be estimated with a precision of less than three years and may be as large as several decades. The assumption by Lux and Rösing (1996) that the precision is a constant  $\pm 2.5$  years is misleading. It is

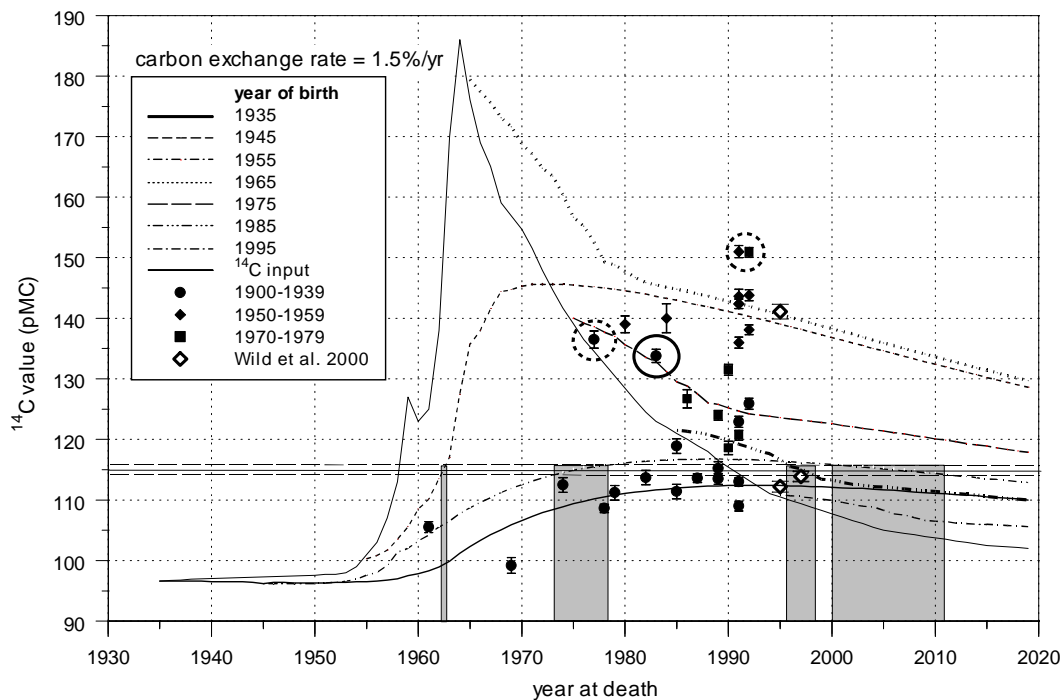


Figure 1 Modeled relationship between the year of death and the specific  $^{14}\text{C}$  activity of human bone collagen for several years of birth (1935, 1945, 1955, 1965, 1975, 1985, and 1995). The measured  $^{14}\text{C}$  values of the samples from individuals born between 1900 and 1939, 1950, and 1959 as well as 1970 and 1979 are shown. The outlier values may belong to individuals with bone diseases (e.g., osteoporosis) that were treated with medicine (circled; the value in the solid circle is certified for that). The examples of Wild et al. (2000) are shown as open rectangles.

Table 1  $^{14}\text{C}$  activity of collagen:  $115 \pm 1$  pMC; age at death 20–30 yr, year of discovery 1995

Assumed year of birth	Corresponding year of death	Maximum age at death (years)	Calculated burial time before 1995
1943	1973–1974	30–31	21 to 22
1945	1973–1977 & 2000–2010	28–32	22 to 18; –5 to –15
1948	1968–1969	20–21	27 to 26
1955	1962–1963	7–8	33 to 32
1965	Not possible	—	—
1975	Not possible	—	—
1985	1996–1998	—	–1 to –3
1995	Not possible	—	—

also open to question whether the dating precision increases with the number of  $^{14}\text{C}$  analyses of bones from the same individual (Lux and Rösing 1996). In any case, the bomb  $^{14}\text{C}$  dating method is also applicable for the first decades of the twentieth century.

The impact of biological processes on the estimates of burial period is shown by other examples. L'Orange and Zimen (1968b) determined a mean specific  $^{14}\text{C}$  activity of  $113.6 \pm 0.6$  pMC for bone collagen from six individuals who died in 1967 and had ages between 56 and 88 years. According to the model curve for 1935 (Figure 1), all measured  $^{14}\text{C}$  values were too large by about 9 pMC. Wild, Arlamovsky et al. (2000) presented four other examples, which are also shown in Figure 1. The  $^{14}\text{C}$  values of the bone collagen of a one-year-old baby born in 1994, of a 30-year-old male, and a 73-year-old woman who died between 1995 and 1997 correspond well with the modeled  $^{14}\text{C}$  activities. The  $^{14}\text{C}$  value for a 46-year-old female who died in 1997 is, however, about 8 pMC too small. Obviously there are exceptions that cannot yet be explained.

### Implications for the Common $^{14}\text{C}$ Dating of Human Bone Collagen

The confirmation that the main uptake of carbon in bone collagen ceases at 19 years and the subsequent carbon exchange occurs at a low rate of about 1.5% affects the conventional  $^{14}\text{C}$  dating of bone collagen. Any conventional  $^{14}\text{C}$  age  $t$  of human bone collagen has to be corrected to take into account the fact that the main  $^{14}\text{C}$  uptake ceases in the adult human. The correction term  $E_{\text{corr}}$  depends on the age  $t_{\text{life}}$  of the individual at death. The real  $^{14}\text{C}$  age  $t_{\text{real}}$  is obtained from  $t_{\text{real}} = t - E_{\text{corr}}$ . This correction is important for dating historic events related to dated bones. Figure 2 shows, for example, that 32 years have to be subtracted from the  $^{14}\text{C}$  age of bone collagen of an individual who was 65 years old at death.

### Bomb $^{14}\text{C}$ Dating of Leather and Hair from Pelts

The  $^{14}\text{C}$  values of the leather from 19 pelts from carnivores and herbivores in their relation to the year of death are shown in Figure 3. I assume that the main uptake of carbon also occurs during the growth phase similar to the collagen of human bones. Mammals usually approach their adult size within the first year of life.

The straight solid bars show when the animals were imported or born in a zoo. Data points with dotted bars belong to animals that were shot and the age could only be estimated based on the  $^{14}\text{C}$  value. In all cases realistic ages at death were obtained. An exception is sample No. 17 from a beaver pelt. It was apparently a young animal and the reservoir effect may play a role.

The mean residence time of carbon (MRT) was estimated from the  $^{14}\text{C}$  values for samples from animals born before 1954 as in this case the year of birth must be exactly known. I calculated the dashed curve for an exchange rate  $r = 5\%$  or a  $\text{MRT} = 20$  yr. Sample No. 2 is from an animal from the southern hemisphere for which the  $^{14}\text{C}$  input curve deviates from the used one. Sample No. 11 was from a 12-year-old black panther that was feed with frozen meat from national strategic reserves. Skin No. 3 was of a lion baby that died shortly after birth. The  $^{14}\text{C}$  value reflects that of the mother lion.

It is apparent that the  $^{14}\text{C}$  values of the leather is a complicated function of the dates of birth and death as well as the kind of food. Hence, they are not suitable for a reliable and precise determination of the death of the animals using bomb  $^{14}\text{C}$ .

Hair is usually replaced twice a year. The  $^{14}\text{C}$  activities of the corresponding samples (Figure 4) fit well to the  $^{14}\text{C}$  input curve of atmospheric  $\text{CO}_2$  if a bias of one year is assumed. This bias reflect the mean age of the food of one year.

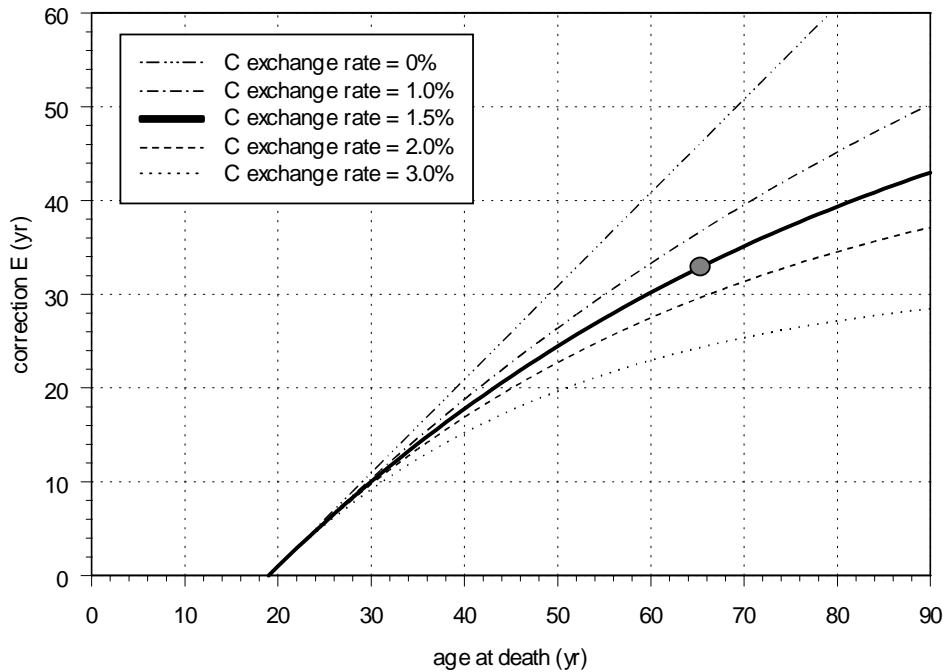


Figure 2 Correction term  $E_{\text{corr}}$  for conventional  $^{14}\text{C}$  ages of human bone collagen as a function of the age at death of the individual and different carbon exchange rates. The most common one is 1.5%.

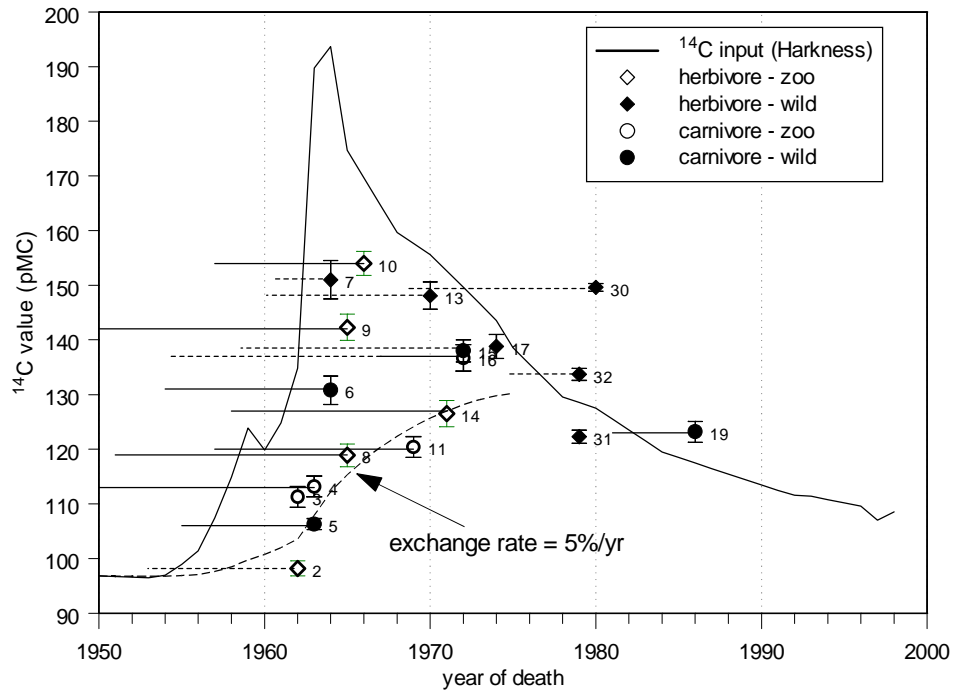


Figure 3  $^{14}\text{C}$  values for the leather of 19 pelts as a function of the year of death. The dashed line was calculated for a carbon exchange rate of 5% per year.

The  $^{14}\text{C}$  values for the hair are closer to the  $^{14}\text{C}$  input curve than those for the leather. Surprisingly, the  $^{14}\text{C}$  values for hair of most of the carnivores also fit this picture. There are four exceptions. The age of the feed meat of more than one year is reflected by the  $^{14}\text{C}$  value of a black panther (No. 11) and a red fox from Libya (No. 15). The  $^{14}\text{C}$  value for a beaver (No. 17) is too low because its food consists of aquatic plants (reservoir effect) and roots several years old. The outlier value for a bear from Malaysia (No. 14) that lived 13 years in a zoo cannot be explained.

On the basis of these results, the  $^{14}\text{C}$  value for hair can be used to determine the year of the animal's death with a precision of about  $\pm 2$  years.

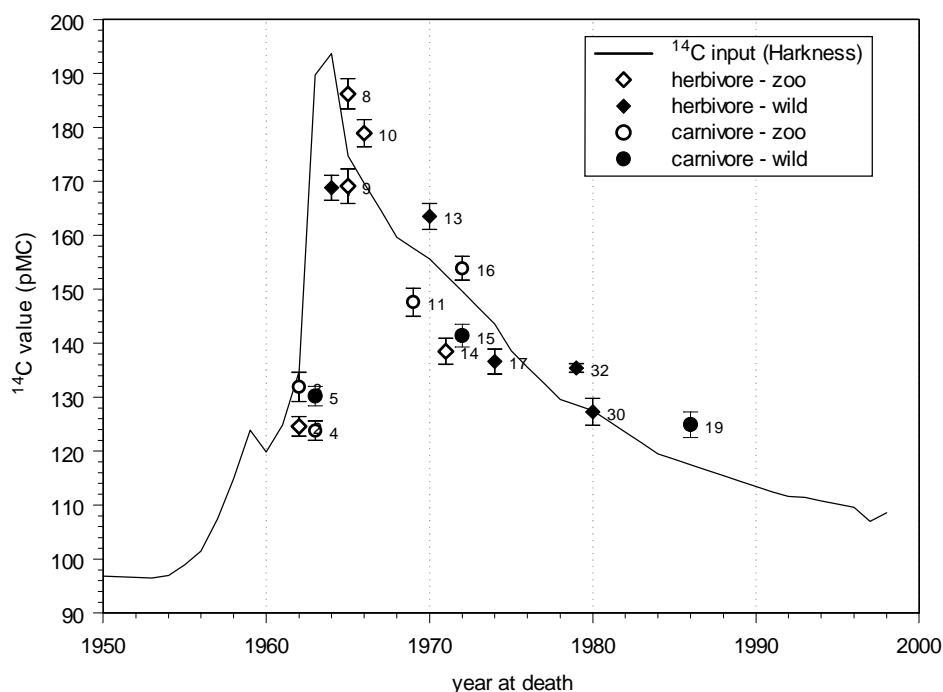


Figure 4  $^{14}\text{C}$  values for hair from 19 pelts as a function of the year of death

## CONCLUSION

The attempts to date the collagen fraction of human bones by means of bomb  $^{14}\text{C}$  show that the precision ranges from at least three years to several decades. This low precision limits the application of this method in forensic medicine. Improvements in the method should be possible if sample collection takes into account the growth direction of bones.

A correction of conventional  $^{14}\text{C}$  dates for bone collagen is required because the main carbon uptake ceases at 19 years with the termination of puberty. The maximum correction is the difference between the age of the individual at death and at puberty and is important if a historical event has to be dated by means of bone collagen.

The  $^{14}\text{C}$  activity of hair from pelts of animals on the "Red List" yields precise dates if a bias of one year for the carbon uptake is taken into account. Leather is not suitable for bomb  $^{14}\text{C}$  dating.

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