ARCHAEOLOGICAL IMPLICATIONS OF NATURAL CARBON-14 VARIATIONS

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CONTENTS

1. INTRODUCTION

2. THE RELEVANCE OF A CALIBRATION

3. THE NORTH AMERICAN CALIBRATION CURVES

4. THE CHARACTERISTICS OF THE CALIBRATION CURVES

5. THE USE OF CALIBRATION CURVES

6. EPILOGUE

7. REFERENCES

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1. INTRODUCTION

The method of dating archaeological finds by means of natural radiocarbon is based on three principles:

(i) The radioactive isotope ^{14}C is continuously being produced in the atmosphere by cosmic radiation; as $^{14}CO_2$ it mixes well with the inactive atmospheric carbon dioxide and is taken up by the oceans and by plants during assimilation; because of the continuity of the processes of production and decay during a geologic time which is long compared to the ^{14}C half-life, a state of equilibrium has been reached; as a result the ^{14}C content of the atmosphere is constant and known.

(ii) During the life-time of vegetable and animal tissues an equilibrium exists between the ^{14}C content of the organism and that in the atmosphere; after its death the isotopic exchange ceases and the ^{14}C concentration only decreases by radioactive decay.

(iii) The rate of radioactive decay is constant and known; consequently, by measuring the specific ^{14}C activity in "dead" material, the time elapsed since the organism ceased taking up ^{14}C (the "age") can be determined; the conventionally used half-life of ^{14}C is 5568 years, although we presently know that 5730 ± 40 years is a better value; using the conventional half-life and the internationally adopted standard ^{14}C activity (NBS oxalic acid), ages (in years *B*efore *P*resent) count back from AD 1950.

Based on these principles and conventions a ^{14}C chronology has been established for the prehistorical cultures from the Late Palaeolithic to the Roman times, which is based on many thousands of radiocarbon dates (Lanting & Mook, 1977).

De Vries (1958) was the first to report variations in the atmospheric ^{14}C content which he indirectly observed in tree-rings from European and North American wood. Since then over thousand ^{14}C analyses have extensively confirmed the fact that the natural ^{14}C level has not been constant. These measurements have largely been carried out on North American dendrochronologically dated wood from Sequoia and Bristlecone Pine by the radiocarbon laboraties of the University of California at San Diego in La Jolla, the University of Arizona in Tucson and the University of Pennsylvania in Philadelphia; the results have on various occasions been reported by Suess (1970), Damon (1972) and Ralph (1973) and co-workers.

During recent years a few laboratories (Groningen, Seattle, Belfast, Heidelberg) have come to dispose of counter equipments allowing much more precise ^{14}C measurements than the usual $\pm$ 40 years. The medium-term variations, claimed by Suess to exist in almost any part of the 8000-years calibration curve, were confirmed with high precision for the period 3800-3200 BC (de Jong et al., 1979). At the Radiocarbon Conference in Bern/Heidelberg in 1979 Stuiver (Seattle), Pearson (Belfast) and Bruns (Heidelberg) showed similar variations to have existed during more recent periods. It seems that during the few years to come the archaeological world will be confronted with a wealth of precise ^{14}C analyses on – partly floating – tree-ring chronologies, enabling calibrations essentially better than before.

In order to be able to judge the present situation and the perspectives, we have to discuss the relevance of a dendrochronolocical calibration of the ^{14}C timescale in some detail.

2. THE RELEVANCE OF A CALIBRATION

For the greater part of the ^{14}C samples, whether they are of archaeological or geological interest, there is no need for comparing radiocarbon dates with "true" or astronomical ages. Radiocarbon has presented its own consistent timescale. In some instances, however, the relation between both is of great relevance.

1. *The correlation of prehistoric cultural phases in the radiocarbon chronology with historically dated events* showed severe discrepancies, already in an early stage of the development of radiocarbon dating. It is well known, for instance, that radiocarbon dates the 1st Egyptian Dynasty about 700 years too late. Vogel (1969) discussed the correlation between the British Wessex culture and the Shaft Grave Period in Mycenae. He pointed out that, after applying a correction to the ^{14}C dates, the early Mycenae (Middle Helladic) is correlated with the later Wessex II instead of the early Wessex. In general, the fact that ^{14}C ages of over 3000 years BP are too small, may alter existing ideas about centres of cul-

tural development and diffusion.

2. *The relation between radiocarbon dating and other independent physical dating methods* is still uncertain. Archaeometric dating on relatively young material, as there are the methods based on thermoluminescence, aminoacid racemization, archaeomagnetism and fission tracks, can still not compete in accuracy with radiocarbon dating. Independent geological dating methods using ^{40}K-^{40}Ar or non-equilibrium decay series (^{230}Th-^{231}Pa; ^{230}Th-U) still have insignificant if any overlap in datable time range with ^{14}C or are insufficiently accurate to act as a cross-check.

3. *The true duration and the close succession of cultural developments* can be wrongly indicated by the conventional ^{14}C dates. The duration of a cultural phase as concluded from these dates is too large during a period where the slope of the correction curve (fig. 2) is larger than the straight line indicated, and conversely. Furthermore, a part of the correction curve having a steep slope (during this period the ^{14}C content of the atmosphere was increasing) can present itself as a hiatus in a series of ^{14}C datings and thus in a cultural development. This stretching of the ^{14}C timescale was seriously considered by Lanting et al. (1973) in relation with the continuity of the Beaker chronology (that is, the transitions PFB – AOO – BB) for the Middle and Lower Rhine area around 4000 years BP.

4. *Histograms* have been constructed by Geyh and Streif (1970) and by Roeleveld (1974) presenting the periods of formation of peat deposits in the North Sea coastal region. A non-linearity of the ^{14}C timescale will present itself as a dense assemblage of ^{14}C dates during a period of decreasing atmospheric ^{14}C concentration. Ignoring this might erroneously lead to conclude to a high prehistoric activity during this time.

5. *A floating dendrochronologically dated sequence* or, in certain cases, *a stratigraphical* sequence of samples allows a very precise dating, if a ^{14}C calibration curve is available. Examples have been presented by Ferguson et al. (1966) and Mook et al. (1972) studying the stratified Swiss lake dwellings (cf. Waterbolk, Niederwill Monograph II, in prep.). The principle is that a special and irregular pattern shown by the sequence of ^{14}C ages is fit to the same irregularity in the ^{14}C correction curve.

3. THE NORTH AMERICAN CALIBRATION CURVES

Almost all measurements checking radiocarbon ages of dendrochronologically dated tree-rings have been performed on North American wood, although the study for secular variations (the "De Vries effect") as revealed by the Sequoia gigantea was started in Europe (Willis et al., 1960; Lerman et al., 1970). Since then almost 1000 tree-ring samples were dated in the U.S. laboratories cited in the introduction, mostly from the bristlecone pine (Pinus aristata) (Ferguson, 1970). The recently revised results of Suess (1978) are compiled in fig. 1. The majority of the tree-ring samples cover a period of 10 years. The standard deviations in the ^{14}C results are not indicated but are between $\pm$ 40 and $\pm$ 100 years.

The three laboratories have drawn their own curves. Suess (1970) originally drew a curve by hand through his points, Damon et al. (1972) computed a third order polynomial through 25 year averages of four laboratories including Yale, while Ralph et al. (1973) smoothed the data of the three laboratories through a 100 years moving average procedure. Ralph et al. (1973) derived an equation to convert radiocarbon into historical ages (revised by us to apply to conventional ages BP):

$$T_{conv} = 1907 - 0.892\ T_D - 6.97 \times 10^{-5} T_D^2 - 1.15 \times 10^{-8}\ T_D^3$$

where T_{conv} is the conventional ($T_{\frac{1}{2}}$ = 5568 years) ^{14}C age and T_D is the historical age in AD/BC. Finally, Ralph and Damon separately presented correction tables which resulted from their graphs. The general trend of the existing curves is very similar, while the discrepancies largely are a matter of detail. Later on, however, we will point out that often these details are more relevant than the general trend.

4. THE CHARACTERISTICS OF THE CALIBRATION CURVES

We would like to distinguish three different types of secular variations:

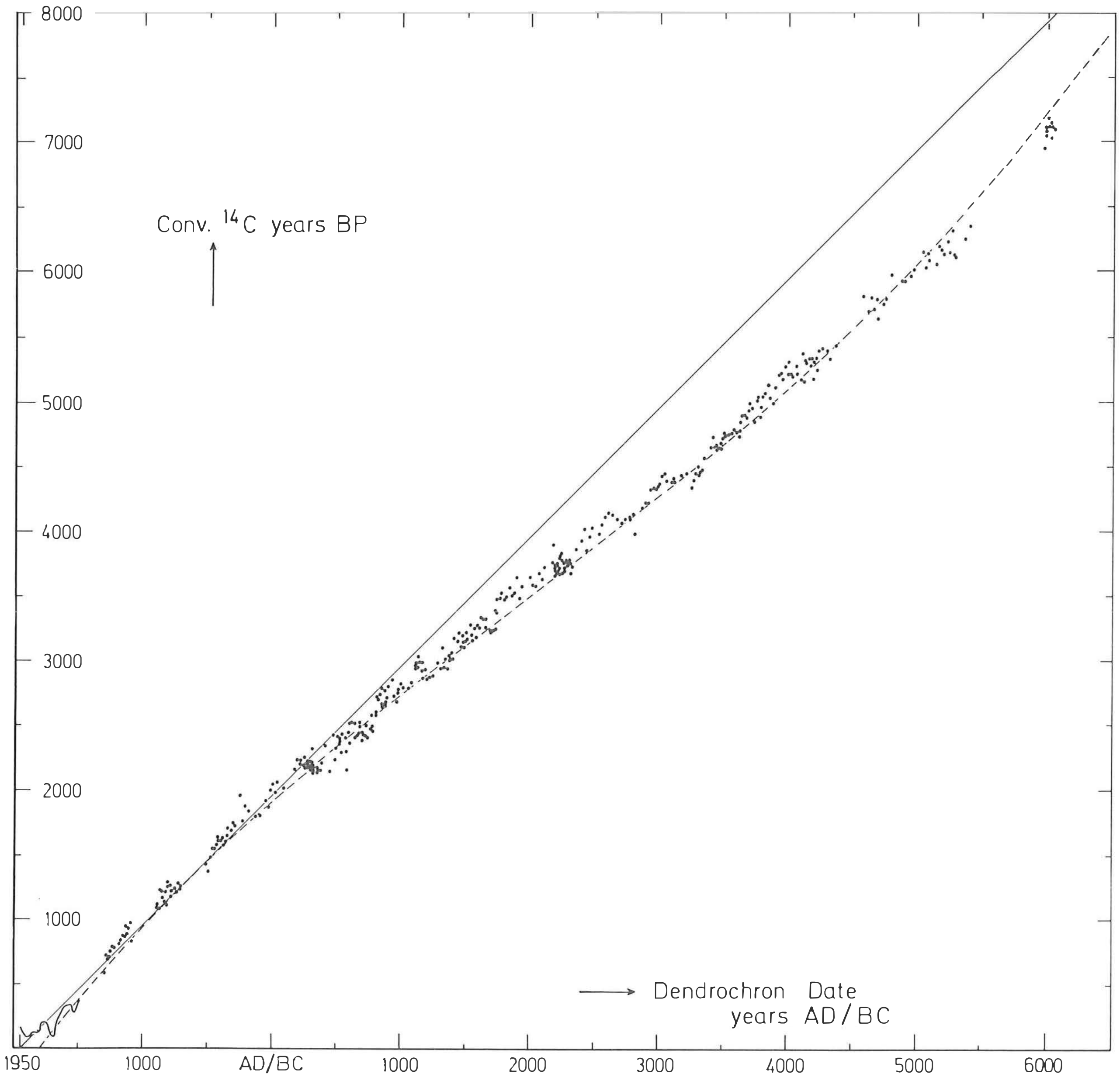


Fig. 1. Graph of Suess' measurements on Bristlecone pine (Suess, 1978). For reasons of clarity the standard deviations (between ± 40 and ± 100 years) have been left out. The dashed line represents the third order polynomial calculated by Ralph et al., 1973). The curve for the 19th and 20th century is taken from highprecision analyses by Stuiver (1978).

(i) *A long-term trend.* As concluded before the slowly changing ^{14}C concentration in the atmosphere (a nearly perfect sinusoidal curve with a period of almost 9000 years) is confirmed by all existing data. By applying this correction to ^{14}C dates from Egypt a reasonable agreement with the historical dates is obtained.

It is important to notice that in the range where the application of the "better" half-life of 5730 years becomes significant (before 3000 BP), the natural ^{14}C trend causes even larger deviations. This makes the use of this half-life, instead of the conventional 5568 years, superfluous. In order to avoid any confusion in the meaning of ^{14}C ages, the 5730 half-life, therefore, is not to be used.

The long-term trend can be accounted for by an equivalent variation in the earth magnetic field affecting the intensity of cosmic radiation in the atmosphere.

(ii) *Medium-term variations*, the so-called "Suess wiggles". As mentioned in the introduction, these variations have now been established to exist beyond doubt. They may amount to an apparent change in ^{14}C ages of a few hundred years within a historical period of less than half a century (cf. de Jong et al., 1979: a drop of almost 250 ^{14}C years during 3400-3350 BC). These variations can be explained by changes in the ^{14}C production rate by about 40% at intervals of about 160 years. The shape of the variations appears to be very similar during entirely different periods. Changes in the solar activity are presumably being held responsible.

(iii) *Short-term fluctuations*. In 1978 Stuiver reported evidence for a correlation between the atmospheric ^{14}C content as observed in tree-rings and the 11-year sunspot cycle. This was further confirmed by Tans et al. (1979). During a period of high solar activity the geomagnetic field intensity is increased, so that the atmosphere is more efficiently shielded against the cosmic radiation. The effect is, however, very small and amounts to about an amplitude of 3‰, which is equivalent to 24 years. This conclusion certainly means a relief to the archaeologists, whenever one-year samples as grain, twigs, etcetera are considered to be dated (Waterbolk, 1971).

5. THE USE OF CALIBRATION CURVES

The purpose of finding the "true" calibration curve with all short- and longer-term variations can only be achieved by measuring thousands of *single tree-rings* with an essentially better precision than the usual standard deviation of $\pm$ 50 years. Any statistical treatment of existing data in order to obtain a better curve from imprecise measurements, as has been suggested (Renfrew and Clark, 1974), is subjective and misleading as long as the quantitative geophysical insight in the shape and size of the variations is not taken into account. At this point we want to remind that the larger errors in Damons curve and tables around 5500 B.P. are not purely statistical but rather due to larger ^{14}C fluctuations during this period of low geomagnetic field intensity (Damon et al., 1972). The "error" to be assigned to a ^{14}C result in this case thus depends on the time span covered by the sample. The uncertainty in age of "short-lived" samples very much depends on the amplitude of the short fluctuations, that of the "long-lived", on the contrary, not. For the latter a smoothed correction curve can be applied with a higher accuracy than is indicated by Damon's tables. On the contrary, the standard deviations quoted in the Masca tables (Ralph et al., 1973) do not take into account the uncertainties in short-lived samples.

Generally speaking, the actual calibration curve to be used, provided the precise true curve is known, depends on the time range represented by a sample. For material covering many years, as charcoal or peat, the detailed curve has to undergo a smoothing to an extent depending on the sample.

The amount of wood necessary for an accurate ^{14}C measurement depends on the chemical pretreatment procedure and the degree of accuracy wanted. According to our experience, the average yield of dry material after standard treatment with 4% hydrochloric acid and 1% sodium hydroxide solution is between 50 and 60%. For obtaining a precision of 5‰ ($\pm$ 40 years) in a counting period of 48 hours about 4 g of dry recent wood before pretreatment is needed. A precision of $\pm$ 8 years (1‰) requires 100 grams of dry wood. This is easily seen from the equation : $\sigma/A = 1/\sqrt{At}$, where σ is the standard deviation, t is the counting time and A is the net counting rate of the sample (the background is considered as being negligibly small). Wood which is 6000 years old and, consequently, contains about half the recent amount of ^{14}C requires another factor of 2 increase in sample size.

In order to illustrate the use of calibration curves, provided the above mentioned "true curve" is avaible, a few graphs are shown from results obtained in our laboratory. De Jong et al. (1979) have accurately ($\pm$ 10 years) measured tree-ring samples from German oak material, each sample covering one or two years from a "floating" chronology at 3200-3900 BC. The 1-year curve (fig. 2a and 3a) is obtained by computer construction of a Spline-function through the single results. The 20-, 50- and 100 year curves of fig. 2 and 3 consist of moving averages and represent the proper calibration curves to be used for samples covering 20, 50 and 100 years respectively. The consequences of

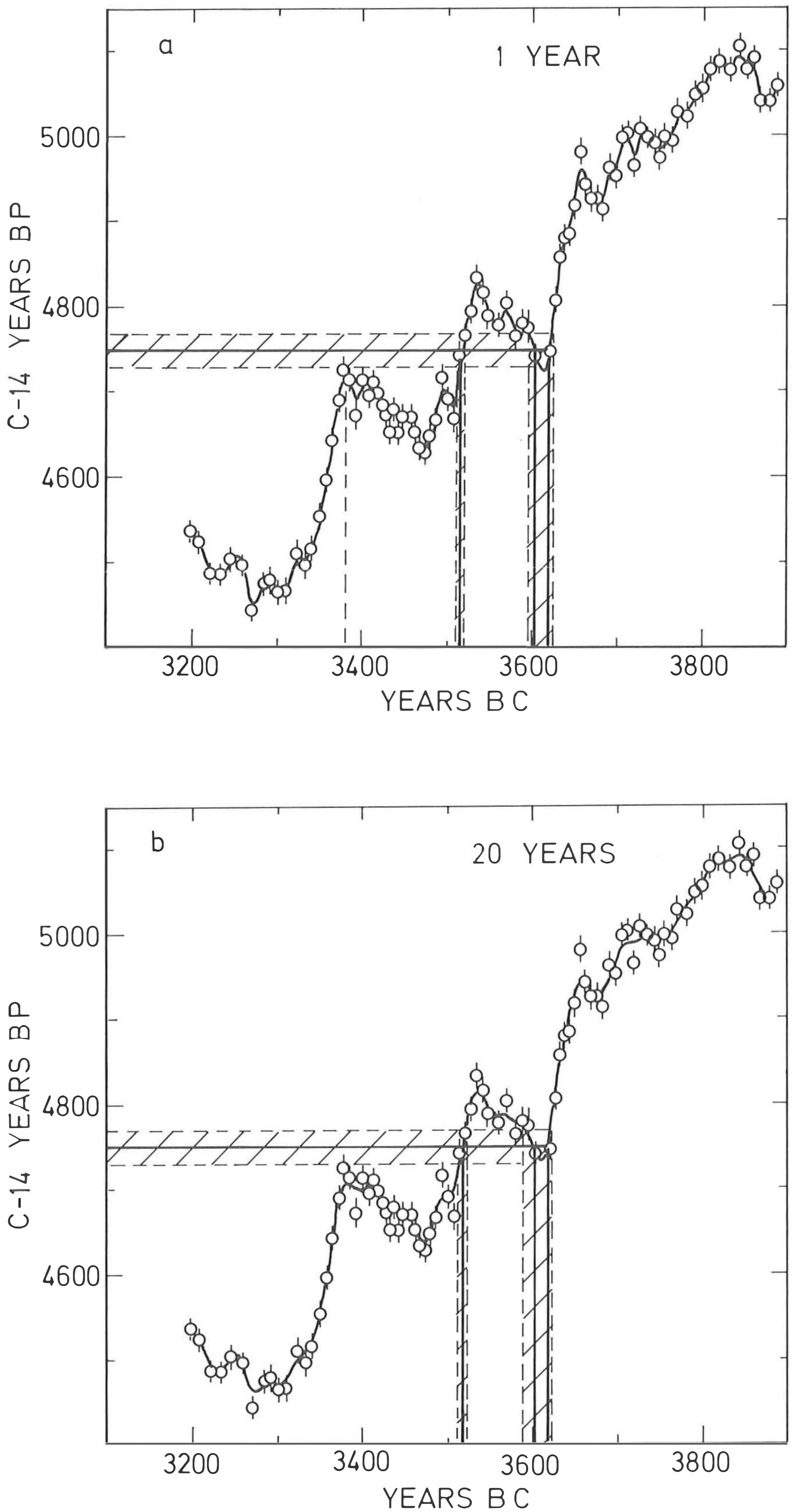
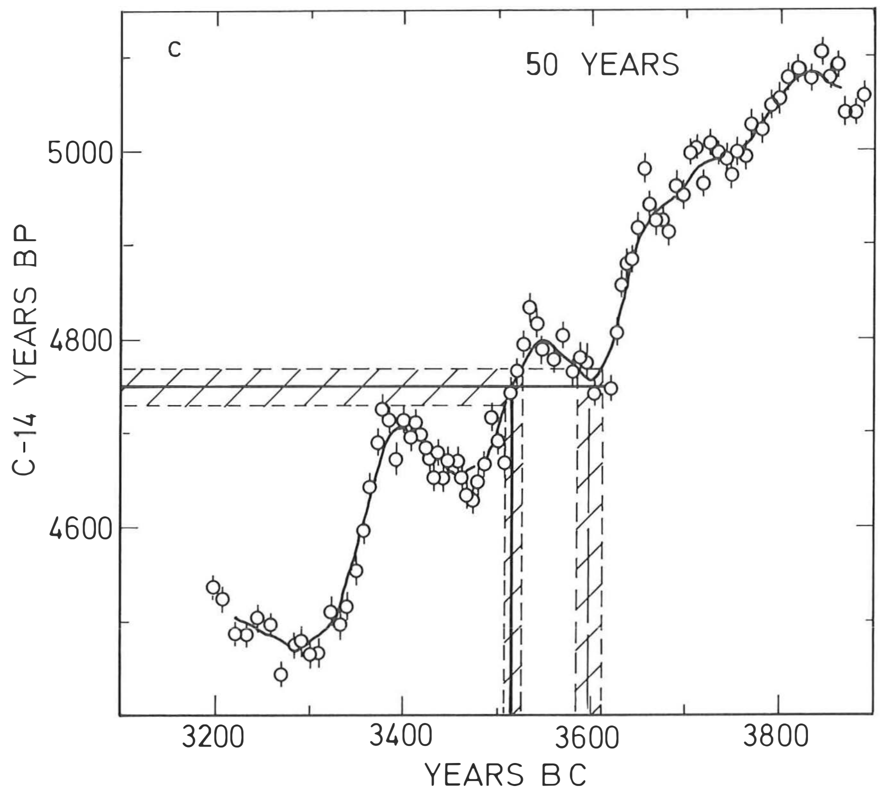
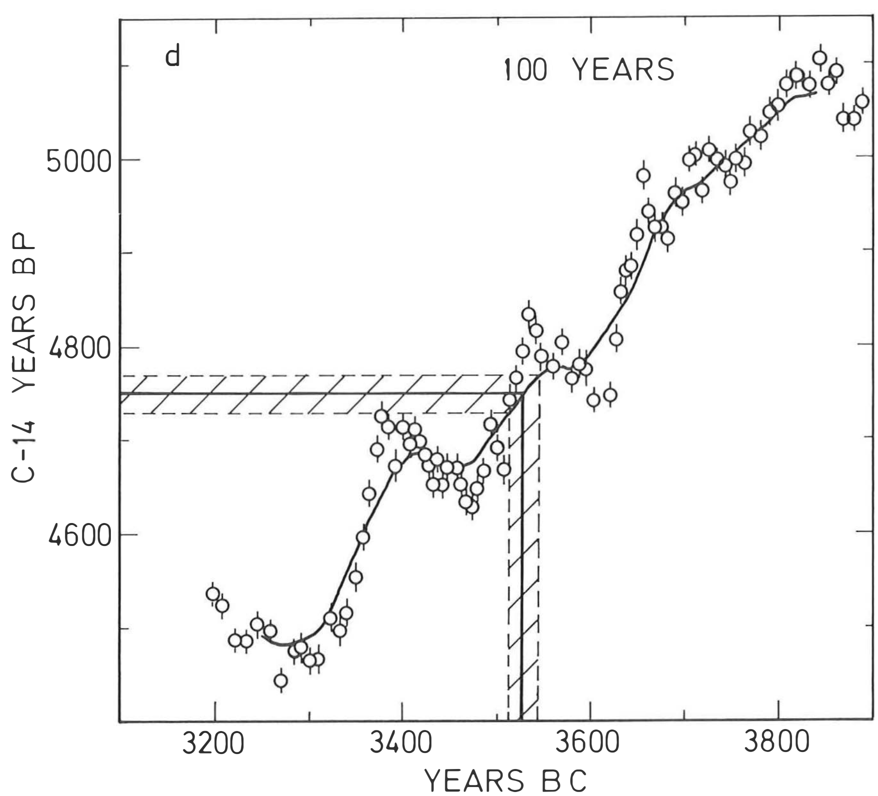


Fig. 2. Calibration curves from precise ^{14}C analyses on single or double tree-rings from Beckers floating chronology (Donau 7). The horizontal scale is calibrated by "wiggle matching" with Suess' results on the absolutely dated Bristlecone pine (De Jong et al., 1979). From the original data a Spline-function was calculated and drawn by computer (a). The curves b, c and d are drawn by calculating moving averages over 20, 50 and 100 years BC, respectively, and apply to ^{14}C samples representing 20, 50 and 100 years of growth.

As an illustration a calibration is shown for a conventional age of

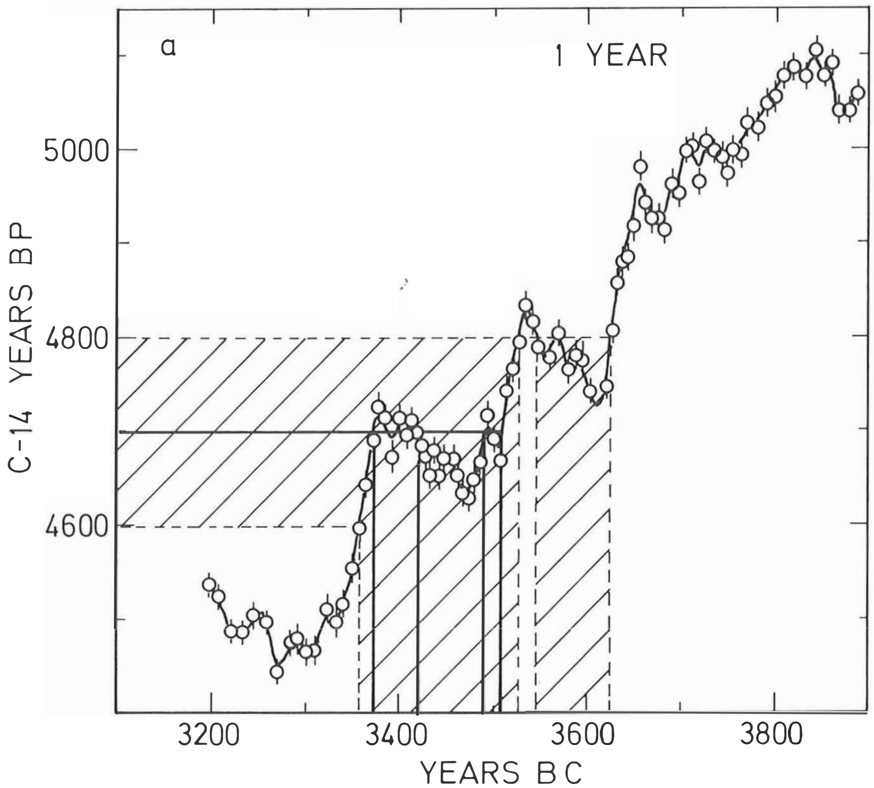




4750 ± 20 BP. The hatched areas are the regions of uncertainty applying to one standard deviation.

It appears that dating a sample covering 100 years gives the best accuracy in historical age; however, this only applies to the unrealistic case that each year of growth is represented to the same amount in the sample.

In our construction of the historical ages the statistical uncertainty of the curves has not been taken into account. The 1-year curves of fig. 2 and 3 have an inaccuracy of about ± 10 years, whereas the averaging (smoothing) procedure makes the curve more precise.



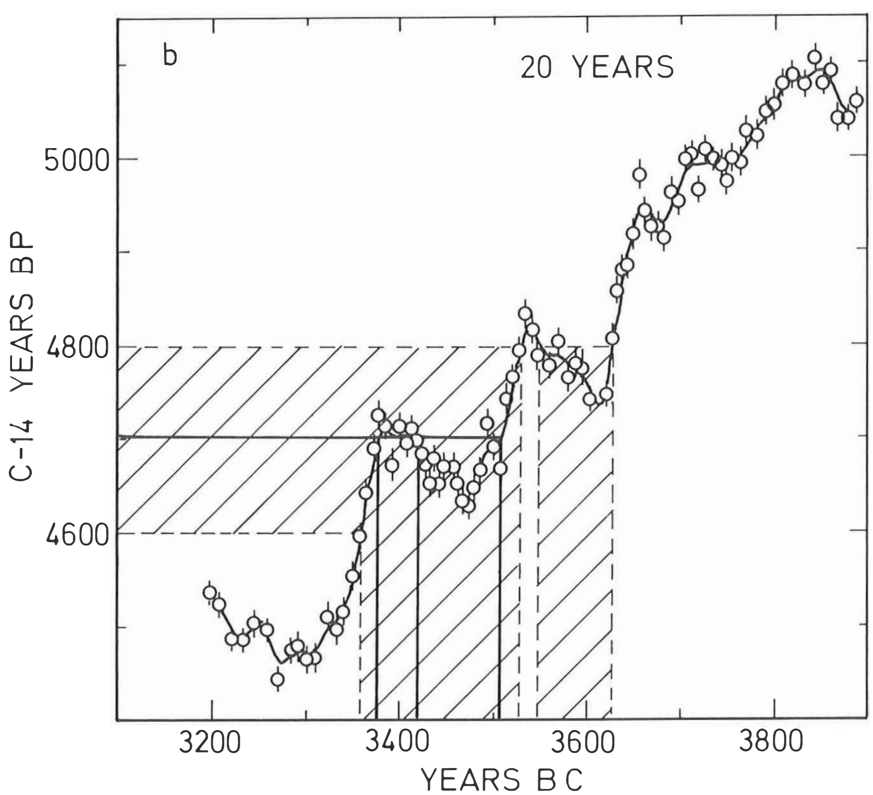
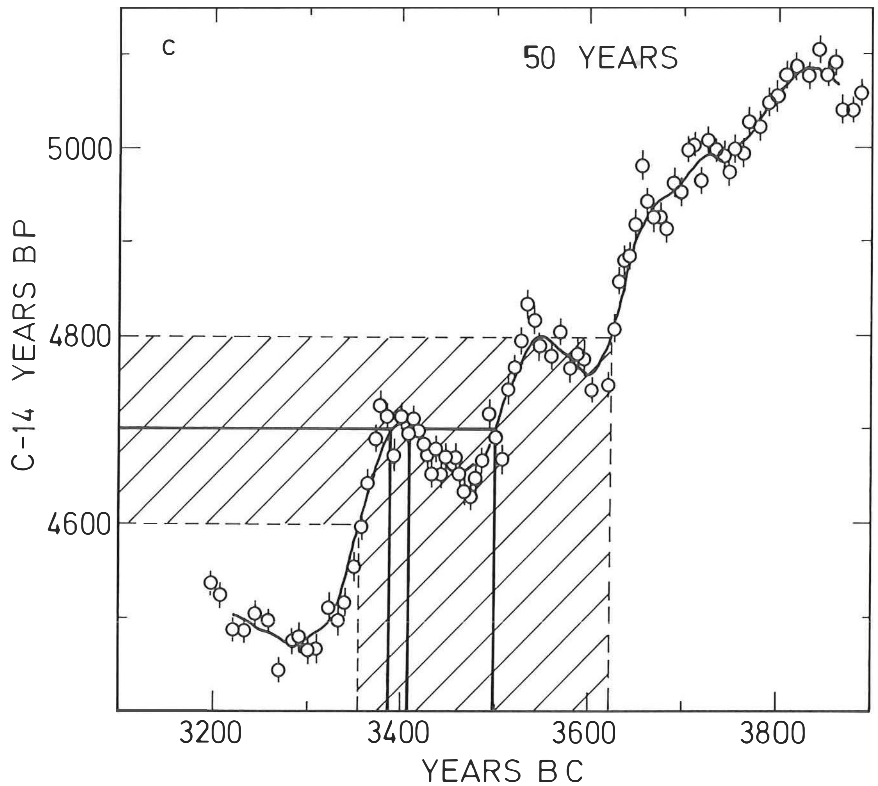
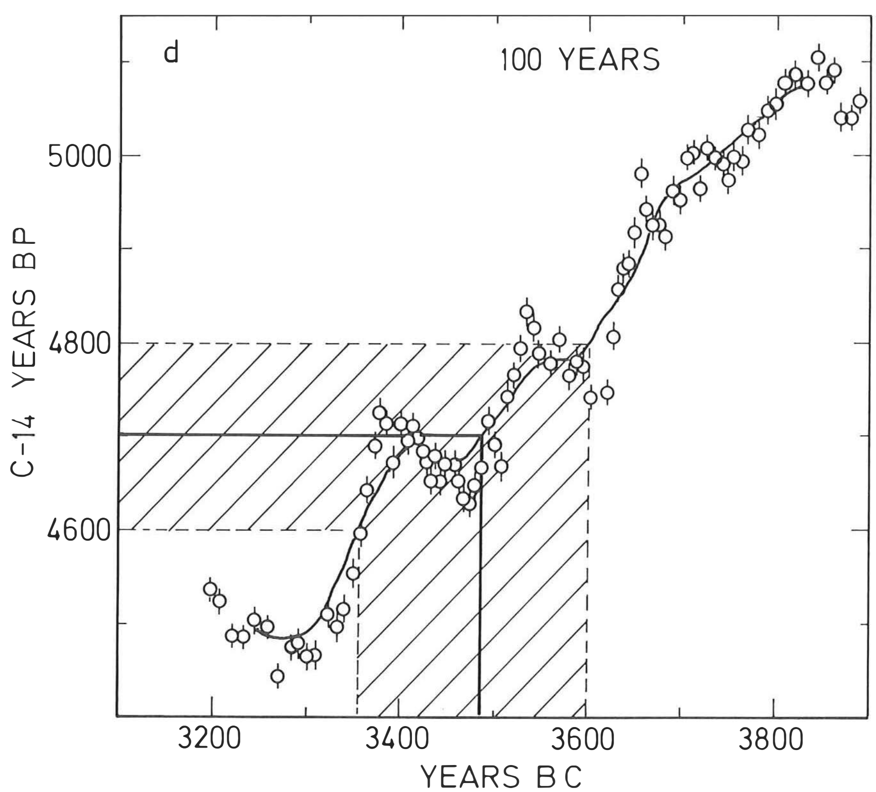


Fig. 3. Same curves as in fig. 2. Here a conventional age of 4700 ± 100 BP is calibrated. Again figures a, b, c, d represent the graphs to be applied if the samples cover growth periods of 1, 20, 50 and 100 years respectively. It is obvious that, compared to the example of fig. 2, a





less precise analysis requires less certainty about the growth period represented by the sample. The 100 year curve agrees in general with that given by Ralph et al. (1973) in the Masca paper. Our curve shows somewhat more detail.

selecting the improper calibration curve is shown for two arbitrary ^{14}C ages: 4750 ± 20 BP (fig. 2) and 4700 ± 100 BP (fig. 3). It appears, that in the case of a 100-year sample which is measured to ± 20 years the 100-year curve should be used. If the analysis is imprecise (± 100 years) it does not make much difference, what curve is used. On the contrary, if the sample originates from one year of growth, the use of the 100-year curve (cf. the Masca correction curve) would lead to a too simple impression about the true age.

From the foregoing we can conclude that (i) a calibration curve should be used which applies to the number of years represented by the samples, (ii) condition (i) is the more essential the better precision is obtained with the ^{14}C analysis, (iii) the conditions (i) and (ii) depend on the irregularity of the 1-year calibration curve (if the curve is an almost straight line, either curve will be satisfactory), (iv) the proper correction curve can easily be calculated and plotted by computer, once the single-year data are known.

6. EPILOGUE

Part of this paper (Mook, 1977) was presented at a special meeting on dendrochronology in Mainz in 1974 which Waterbolk and the first author attended. The situation with respect to the calibration of the ^{14}C timescale has changed markedly since then. A few laboratories have independently decided to devote part of their effort to high-precision ^{14}C measurements. We can therefore expect that the progress in ^{14}C calibration will be significant during the first years to come, much earlier than we anticipated a few years ago.

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