

Dating methods

Time is a very strange quantity, which tends to swell or shrink in particular situations and is apparently able to scroll in both directions, i.e. into the future and in the past; but this is not true because the equations of motion does not come back (as in a film screened in reverse) and measure the time back is very different from using a clock. The idea, apparently, to create a system of absolute dating is much more complex than it seemed at first in the fifties of last century, when it was discovered the radiocarbon method. Measure time has always been one of the activities where humankind were given greater importance, whereby they continually come up with sophisticated methods, starting with those who use the astronomical phenomena which, with their frequency, are probably the primary cause of our feeling (psychological) of the passage of time. Generally the idea is always the same: find a periodic phenomenon and count how many times it is repeated during the time interval that you want to measure. The greater will be the period, the time clock is much less sensitive. This is the reason why modern clocks use clock with 32768 oscillations per second, reaching truly remarkable sensitivity. If the time interval to be measured is long, it is necessary to use special measurement systems and use a calendar to plan future activities, as is done on computers that have a clock and a calendar available.

To measure a time that has passed, we need a very different strategy, to use methods that are based on the observation of phenomena very slow, started well before the date you want to estimate: this must be assured. An emblematic case is that of a sediment, or even that of a tree, which has a progressive development with a periodicity that is determined by the seasons, depending upon climatic conditions. In the case of a tree counting growth rings allows the calculation of the time that has passed since a starting date; It is, however, incorrect to believe that the identification of a periodic phenomenon that has taken place in the past allows to make a measure of time as we do with today watches. In the measurement of time that is passing you can make use of the observation of the event for which you want to measure the duration (for example an athlete passes the finish line) this is not true for events that have already taken place. The dating's in fact relate to objects and not to events. It is therefore necessary to find a connection between an event and an object that existed at that time, this operation will never be direct but will require some intermediate steps that will, often strongly, increase the uncertainty of the estimates.

Men have continuously developed methods to build chronology (and where possible calendars) that are essential tools to reconstruct the events occurred for a long time in the past, but also to understand and better organize those on horseback with the present. A chronology is a scale that can be referenced in the positioning in the time of events of interest, it is an important tool for a historian, but also for those scientists who study past events, such as the geological formations or astronomical events that are of interest to the history of the Earth and the people who lived in it. It is well known that to make a measurement, it is necessary to have a scale, it is clear, therefore, how the methods devised by historians are, at least in their rationale, very similar to those of scientists. Also for dating is necessary, as we shall see, create a reference scale (which does not define 'calendar' in order not to create confusion); It is therefore interesting to study the characteristics of a scale that make it more or less useful in making a measurement. Take for example a ruler: it consists of many adjacent divisions that indicate increasing values of length. As in stratigraphy the individual layers correspond to periods-sometimes well-known sometimes less-in the ruler you can take smaller intervals, thus increasing the sensitivity. You still can't go too much in this game as under millimeter the eye has difficulty to finds exactly the mark that match the measurement. Similar things happen for the chronologies (but also for stratigraphy) that are characterized by intervals which cannot be too small as it is often difficult to have a detailed grid of events, especially if you go too far back in time¹. The scale need to

¹ Currently uses the Gregorian calendar, which are well-known relationships with those who preceded it in the immediate. There are calendars, used in the past, often built on the basis of astronomical knowledge, the existence of

have a minimum value (in many cases the null value that is zero), a maximum value and ranges. If the intervals are all equal a linear scale. This also happens to the histories but could be characterized by intervals of time not all equal and an arbitrary starting point². Any error on the position of the initial value introduces a systematic error in the calculation of all values that refer to that chronology, though this error is eliminated when determining a time interval between two events that refer to the same chronology.

An assumption shared by scientists and historians is that the time scale is linear; It may happen, however, that the intervals are not all exactly equal or, like a ruler which expands with increasing temperature, the interval between two marks is no longer a millimeter. This introduces a progressive offset of all results that is difficult to correct, these happened in previous calendars, which have been accumulated over time with many small displacements, so that it needed in 1582 to realign dates deleting days. It was necessary to correct some ancient chronologies, when it was realized that some elements were omitted or incorrectly evaluated.

The chronology system needed always of external verification to improve its operation and gain the trust of scholars. It is not easy to enter from the outside into a chronology, by assigning a date that is more credible than that calculated with the same chronology. In fact, it is necessary to use a method that refers to a scale that is somewhat more credible than one built with patience by scholars: this is what we decided to do in the last sixty years with radiocarbon dating and other dating methods. From these considerations comes the distinction that is made in archaeology between absolute and relative dating, giving to the radiocarbon a meaning of method for absolute dating which, as we shall see, it is not correct. It is useful, however, emphasize that all the dates set within a chronology, take a relative value, while that produced by an independent method provide an absolute value.

Lastly, understand that is a very different thing to date an object that cannot be placed in connection with a chronology (or a strata in the case of a finding of an excavation); in the latter case, the use of dating can be almost obligatory. The dating techniques for authenticating artifacts is becoming very common practice, even if the risks are not lower than those using a chronology without making indirect verifications.

The radiocarbon dating

The radiocarbon method is born from the idea of creating an absolute dating method reliant on the mechanisms by which the nuclei of atoms are bound together, which cannot easily be changed. The most well-founded part of the radiocarbon method is that linked to the fact that only one of the isotopes³ of carbon-¹⁴C – is radioactive so its presence can be easily highlighted through a measure of activity. Following the disintegration, the ¹⁴C transforms into ¹⁴N (stable isotope of the element with atomic number greater than one unit) with the emission of a radiation β^- .

The **radioactivity** or, more simply the **activity of a radioactive source**, is the number of disintegrations occurring in unit of time. It is measured in **Bequerel**, which corresponds to **one disintegration per second (1 Bq)**. The disintegration of a radioactive nucleus is a random process and then regulated by a precise

which is evident from the sources (or from results of archaeological research), that can be placed in relation to the newest ones only approximated.

² like the Gregorian calendar, which we use for dating an event, which has as its starting point that, by Convention, is deemed to be the date of Christ's birth.

³ an isotope is one of the possible configurations of the nucleus of an element. Because the nuclei are composed of protons and neutrons, and the first are constant for a given element the various isotopes differ only in the number of neutrons. In the case of the three isotopes of carbon they have one neutron in addition going from ¹²C to ¹³C up to ¹⁴C therefore has two more neutrons of ¹²C.

probability distribution⁴: therefore we cannot in any way to know when, exactly, a particular nucleus will disintegrate. However, since this is a probabilistic phenomenon, we can assume that the average number of disintegrations per unit of time (and therefore the activity) is proportional to the number of radioactive nuclei present, so higher is the mass of the source greater is the activity. It will be so useful a quantity that measure the activity per unit mass: the **specific activity** (dpm/g or q/gram). However, even in the presence of a few milligrams of substance and while taking into account the rarity of certain radioisotopes, we are always dealing with millions, if not billions of atoms, so we can know how many atoms (although not what) will disintegrate in a certain time interval with good precision.

Why a nucleus is unstable and decays, related to well-known mechanisms-that hold together the particles that constitute a nucleus; it is logical therefore to think that the probability that a given nucleus decays is constant and does not change over time. From this simple consideration, using a bit of mathematics, it is easy to get to what is called the law of radioactive decay, which, ultimately, connects the temporal law with that of unstable nuclei of a particular element decay (see Figure 1):

$$A(t) = A_0 e^{-\frac{t}{\tau}} \quad (\text{x. 1});$$

where A_0 is the activity of the radioactive element to the initial time (fixed to zero) and τ is the mean life of the nucleus which is linked, as mentioned above, the probability that the nucleus decays in a certain time interval. What is the basis of dating with ^{14}C is the belief that the radioactive elements decay, now as in the past, with a well-defined temporal law. Unfortunately this law is not linear because the number of unstable nuclei decrease, while the probability remains cost, therefore the number of nuclei that decay at a certain time interval, say for example the mean life⁵, it is less and less as time passes. This, well founded, conviction leads to assume that, if we knew the initial activity of a source and measure the current one, would it be possible to make some kind of track back feature that allows us to estimate the time that has passed. The calculation is simple:

$$t = \tau \log \frac{A_0}{A(t)} \quad (\text{x. 2}).$$

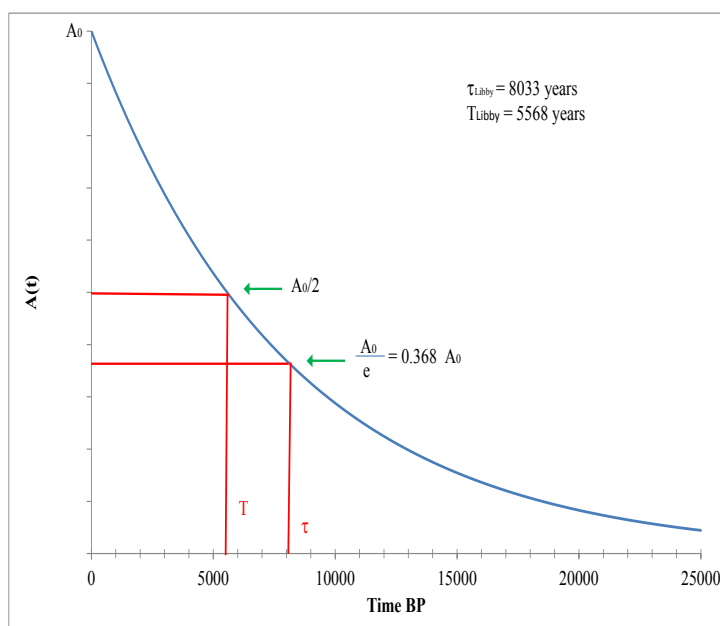


Figure 1 – temporal law of decay of ^{14}C

The ^{14}C has an mean life of 8267 years which corresponds to a half-life of 5730 years⁶. It is necessary to emphasize the fact that ^{14}C is produced in the upper atmosphere from a particular reaction involving nitrogen atoms and neutrons ($^{14}\text{N} (n, p) \rightarrow ^{14}\text{C}$) produced by high-energy nuclear particles that originate from the cosmic space (cosmic rays). The ^{14}C is not-like most of the isotopes-produced in nuclear reactions (mostly occurred in a remote past) or in the decay of nuclei that are part of the families that constitute the natural radioactivity: the ^{14}C is cosmogenic, i.e. continuously produced as a result of the presence of cosmic rays.

⁴ a random variable, as in this case the number of disintegration, is characterized by a function, called distribution, that gives for each possible variable value its occurrence probability.

⁵ it is clear that the mean life may be taken as the unit of measure of time.

⁶ The conventional dates are still calculated using the value determined from Libby who is 5568 ± 30 years.

This has many consequences and not all positive. But the fact that it is continuously produced is positive because it allows you to suggest that its concentration in the upper atmosphere is more or less constant, although it is a radioactive element. If happen its segregation in a compartment, which is not open to new income of ^{14}C from the outside, its activity will begin to decrease with the law that we described earlier. It is therefore necessary to determine the initial activity (A_0) and how ^{14}C income in a system, before it not come into contact with more carbon from the atmosphere.

At this point the radiocarbon method is complicated, however, becoming fascinated by the various solutions adopted to remove various obstacles. Willard Libby was one who worked in a radiochemical laboratory strongly involved in the development of nuclear weapons, in a historical period in which they tried to find out what risks you ran with the use of atomic bombs. In those years he did research on the mechanisms by which radioisotopes present in the atmosphere could follow up on Earth and be ingested by humans. Curiously these researches, promoted for reasons of war, allowed him to develop the radiocarbon method. In fact the inventor of the method is Libby, that earned him the Nobel Prize in 1960.

Living beings are a real carbon pump as it is for them to produce many of the molecular structures of which they are composed. As an example we can say that carbon is virtually non-existent on the solid Earth (minerals and rocks) and is a very significant biological systems ($\approx 21\%$), it is easy, then, to assume that if the carbon comes in contact with the surface of the earth it will be mostly caught by living organisms selectively. The path the carbon from upper limits of the atmosphere to living organisms is obviously much more complex than one might suppose from this simple consideration. The first step is the one that connects the upper atmosphere with the surface of the Earth. The presence of a continuous mixing of the gases that make up the atmosphere is due mostly to evaporation from the surface of the seas (which occupy the entire surface's 71%) that causes carbon dioxide (CO_2) in the atmosphere is continuously with the remix. In this mechanism play a significant role the rains that water atmosphere continuously move towards the sea. It's the CO_2 the main transportation vehicle of the carbon towards the Earth's surface is easily justified by the fact that practically all the carbon that is formed in the upper atmosphere quickly turns into CO_2 -through the reaction $\text{C} + \text{O}_2 \rightarrow \text{CO}_2$ ⁷ -because exchanges on the sea surface is this to bring the ^{14}C in the hydrosphere (i.e. in large tank that is represented by the water of the seas and oceans which store about 95% of the ^{14}C , see Figure 2). It may sound like science fiction if there were many experimental results showing that atmosphere, that contains a concentration of 0.03% in volume of CO_2 , and hydrosphere are really in equilibrium and that they have been since the atmosphere was formed in its current composition, about three million years ago. Dissolved in water the CO_2 balance comes in with bicarbonate ions (HCO_3^-) and carbon (CO_3^{2-}) that can convey carbon to the flora and fauna of the sea. As we shall see this is going to be a problem in the dating of aquatic origin (shells, fish, etc.) and can also influence the dating of living beings that have ingested them. As always happens in science what is a problem, when it is solved with a complete clarification of the mechanisms, becomes subsequently a possibility of making new studies and open new fields: in this case for anthropology.

The CO_2 in the atmosphere enters a fundamental mechanism for living beings: photosynthesis. Through this channel comes in living systems (which are part of the biosphere, see Figure 2) the majority of carbon: photosynthesis can, therefore, be considered the true ^{14}C pump to living beings.

In the case of animals that happens only indirectly, through the supply of vegetables. To clarify all these steps you must use a schema (Figure 3) that highlights all exchanges that occur and how we can say with certainty that the proportion of ^{14}C present in an animal is known.

⁷ The formation of CO_2 involve three isotopes of carbon, each according to its abundance of 98.9% for the ^{12}C and 1.1% for ^{13}C . That of ^{14}C is, as seen above, variable

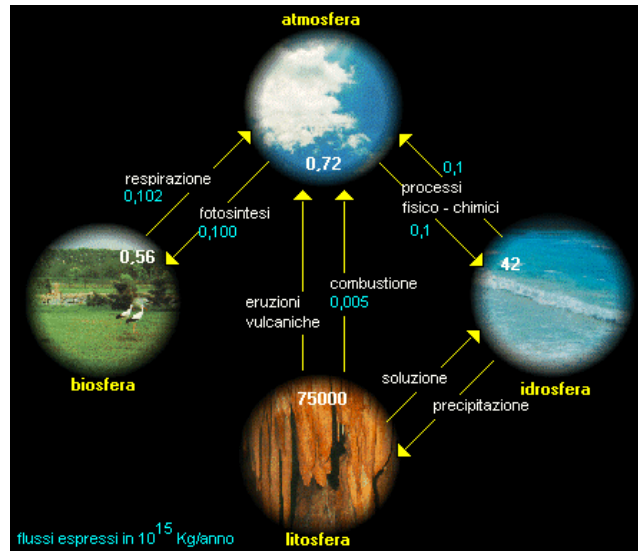


Figure 2 - Schematic of carbon exchange mechanisms between compartments where can logically be divided the land. For each compartment also refer carbon amount contained among that.⁸

Each of these steps raises issues that must be resolved, such as whether the carbon exchanges between the air and plants create a substantial equilibrium between the proportion of ¹⁴C in the atmosphere and in plants. It can be shown that, despite being of the order of the mass of carbon traded daily, for a long time (typical of plants), the concentration of ¹⁴C in plants and in the air are strongly interlinked.

The obstacles are not finished, there is indeed a problem in carbon assimilation by plants due to the fact that the ¹²C isotope by far more abundant, tends to be favored in the various reactions of photosynthesis that plants contain a fraction of ¹²C higher than the atmosphere. This phenomenon is not tied solely to exchange resulting from photosynthesis, but in a different way, it happens to all exchanges of carbon even those with the hydrosphere. Fortunately it is possible to demonstrate that the proportion of ¹³C, the third isotope of carbon, which is also present in nature in amounts well below those of the ¹²C, has similar behavior, so it is possible to correct this systematic shift

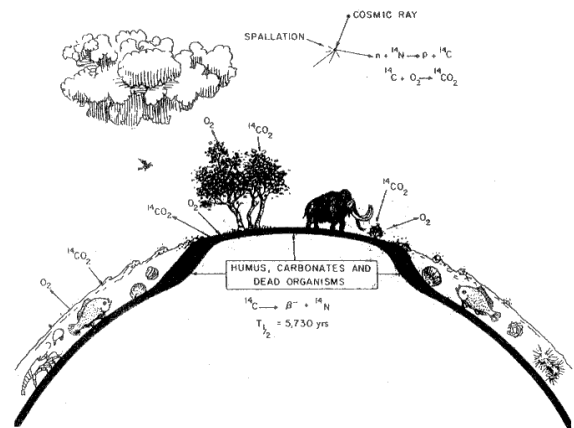


Figure 3 – Logical scheme of exchanges that enable living beings to assimilate ¹⁴C

If this condition exists in dynamic equilibrium of ¹⁴C in the various sectors (as outlined in Figure 2) you could, even now, to measure the initial activity by using a standard and should be calculated from the measured activity the date that corresponds to the death of the living being or loss of contact with the outside of the system that contains ¹⁴C. This is precisely what is done using the Modern Reference Standard (MRS) which is a sample of oxalic acid, by the National Bureau of standards in Washington, from a lot of sugar beet produced in a certain region in a given year. So you can calculate the conventional date using a timescale that begins from the moment the radiocarbon method was introduced, namely the 1950. The conventional

⁸ The large amount of carbon present in the lithosphere is stored in tanks that are not readily available for exchanges with the atmosphere and the biosphere. As they represent a problem for radiocarbon dating because they contain large amounts of virtually carbon ¹⁴C, said 'off' carbon because the ¹⁴C content in it is completely decayed.

date is the result of corrections like that due to isotopic fractionation of which we will discuss later. As you can see radiocarbon method is not a easy-to-read watch, below we will try to illustrate various aspects of the method from the description of how it is applied in practice.

As it is done dating with radiocarbon

The phase preliminary, to the measure, has a fundamental importance in the quality of the results that may be obtained with this method. It is very important to find the sample to date and it must be representative of the set that you want to date, be it a layer (or context) or a artefact that you want to determine the age. For example dating a layer you must ensure that the sample belong with certainty to the layer. In the case of an artefact we must ensure the selected sample is representative of the materials that make up the whole object (which is not always easy). There is always lurking the problem of contamination which can considerably alter the dating results. For example, if the 1% of the carbon extracted from the sample to date has a different source, suppose comes from a tank, how that geological⁹-which contains 'off-carbon', the calculated date will be reduced by about 80 years, being the mean life of ^{14}C of 8033 years¹⁰. If, instead, would be erroneously added of carbon from organic remains, the failure would be much smaller, but much greater as the sample to date is ancient; to give an example for a sample of the late Republican age, 78 BC, the error, for a 1% contamination by recent carbon, would be only 6 years. From these examples, the consideration that the most dangerous contaminations are those with materials from the geosphere. The interest to get a result and the conditions under which it is forced to operate push sometimes to take risks, often we get the aid by sample treatment methods which ensure that carbon retrieved is part of the original and does not contain carbon that was added later. From the sample collected, with due caution, and brought in the laboratory, is extracted the carbon present in it with pretreatment techniques, developed considerably in recent decades, often making treasure of previous errors, and using knowledge about materials and growth processes of living systems, which are strongly increased with the development of modern material science and biology. Sample pretreatment is the stage that requires more attention because its considerable cost. The techniques to be used depend on both the nature of the sample chosen by the next experimental technique that you have decided to apply. For example in the dating of the bones you need to know how to extract the collagen that contains carbon useful for dating, you cannot trust the remaining carbon that could be from diagenesis. There are, of course, treatment techniques that are proven and drive the choice of the sample. Below are listed the quantities required for the various materials dating by AMS that is currently most used technique:

Charcoal	50-100 g
Peat	50 g
Lake sediments and fossils	100-1000 g
Shells, carbonate	20-50 g
Textile fibers	7 mg
Vegetable seeds	0.15 mg
Food remains	1.2 g
Remains of funeral pyres	1.2 mg
Furnaces Coal	0.4 mg

As you can see we tend generally to date non-precious materials, perhaps discharged, still available in layers identified in the archaeological excavations.

⁹ the organic remains contained in or included in deposits minerals that have been for a very long time off from contact with the atmosphere no longer contain ^{14}C because it has all turned into radioactive carbon process retrieved from these remains is said to be "off".

¹⁰ it is possible to prove that if α is the error on $A_0/A(t)$ ratio (see the equation x. 2) the error on the date will be $\alpha \tau$, where τ is the mean life of ^{14}C .

For the determination of ^{14}C in the sample to date, you can follow two different paths, which bring both to analogous results. The most traditional are a measure of specific activity of carbon extracted using very sophisticated measurement techniques, with the use of special gas proportional counter or liquid scintillators. The number of disintegrations in one source is greater, the higher the mass, you must then perform a normalization in order to properly use the law of decay. To use the radiometric method, it must therefore know the mass of the sample, which represents a further small complications. A sample have a smaller activity if it is an old, basically making a measurement with the radiometric method, you might find in terms of count less than one count per second (we pay 0.01 Bq) whereby, it would be necessary to wait a 10^6 s (11 days) to get a precision of 1%, which corresponds to 10^4 counts revealed. Radioactivity measurement has an uncertainty inherent in the fact that, even apart from other measurement errors, there is a statistical threshold determined by the fact that the counts revealed are random events characterized by a probability distribution¹¹, which has a standard deviation equal to the square root of the measured counts. The relative uncertainty is \sqrt{N}/N to which a precision of 1% corresponds to 10000 counts: bringing to a very long counting time. When you have a big amount of material that may not be a problem, although chemically treat a large mass of material is neither easy nor cheap.

You might think that the radiocarbon technique has limitations, in that you can go back in time as much as you want, is only a problem reaching a sufficient statistical accuracy. Instead things are different because there is always the possibility, albeit remote, that the background radiation (largely due to the presence of cosmic rays, getting them!!), not completely canceled by counters shields, can produce spurious counts in the counter. This also allows us to fix another experimental limit of radiocarbon dating that is the upper limit (i.e. backwards in time) that you can reach. We start from the simple consideration that, after they have passed four mean live time – 32132 years - estimated activity of one gram of carbon is 0.0016 Bq, or about one count every minute. To be sure that, a so low counting rate¹² -really derived from ^{14}C the background counting rate should be significantly lower, e.g. 2-3 counts each sixteen minutes. This leads to an upper limit of 30000 years that can be overcome easily by placing larger quantities of materials or using very well shielded systems that have a number of background counts even lower. In a nutshell, it can be used the following relation in which the so called 2σ criteria is used that minimizes the probability of random result, i.e. spurious, to 13.5%, where σ_f is the background standard deviation $t_{\max} = \tau \ln\left(\frac{A_0}{2\sigma_f}\right)$ with $\sigma_f = \sqrt{\frac{N_f}{t_{\text{mis}}}}$ where t_{mis} is the measurement time.

The fraction of an isotope, in particular of ^{14}C respect to ^{12}C , is a very useful figure that can be used to characterize the materials, as it is a kind of fingerprint that in many cases allows you to identify which were the source materials of an artifact. It is the ratio of the masses of two isotopes of a given element which can be determined by a mass spectrometer. Using an accelerator mass spectrometer (AMS) you can directly measure the isotopic fractions in a sample of carbon: you can then use this measure to determine the concentration of ^{14}C in a sample and then date. The use of the accelerator is required to have sensitivities that are necessary to determine the small fractions of ^{14}C that are present in the samples to be dated, the accelerator makes it possible to reveal an extremely small number of atoms of ^{14}C and directly measure the isotopic fraction $^{14}\text{C}/^{12}\text{C}$ -in order to be able to directly compare with that of modern standard and thus to obtain the conventional date. In this case, it should be noted that, as the fraction of atoms of ^{12}C in a modern and ancient sample are equal, because this isotope is stable, it turns out that:

$$\frac{\left(\frac{C^{14}}{C^{12}}\right)}{\left(\frac{C^{14}}{C^{12}}\right)_0} = \frac{C^{14}}{(C^{14})_0} \quad (\text{x } 3);$$

¹¹ Poisson distribution

¹² The counting rate is the number of counts per unit of time

then the specific activity being proportional to the number of atoms of ^{14}C the equation x 2 can be rewritten:

$$t = \tau \log \frac{(C^{14})_0}{C^{14}} \quad (\text{x } 4).$$

So by measuring with a mass spectrometer the isotopic fractions of ^{14}C present in the sample to be dated and in the modern standards it is possible to calculate the date.

The isotopic fractionation

This problem is well known and studied, to become with time a resource for studying the eating behaviors of individuals and populations, but for other archaeological and anthropological studies. The slight weakening of the two isotopes of carbon present with isotopic low fractions can be quantified using the $\delta^{13}\text{C}$ by referring to a geological standards made up of calcite derived from marine fossil, Pee Dee Belemnite (PDB) formation of South Carolina presenting a fraction $^{13}\text{C}/^{12}\text{C}$ of 0.011237:

$$\delta^{13}\text{C} = \left(\frac{\left(\frac{C^{13}}{C^{12}} \right)_{camp} - \left(\frac{C^{13}}{C^{12}} \right)_{PDB}}{\left(\frac{C^{13}}{C^{12}} \right)_{PDB}} \right) \times 1000 \quad (\text{x } 4).$$

The values of $\delta^{13}\text{C}$ are calculated per thousand from the value for marine carbonates (the standard has in fact that source). The values are generally negative, the CO_2 in the atmosphere presents a fraction of C^{13} slightly (-8%). As mentioned above, the correction on the fraction of ^{14}C can be calculated as $\delta^{14}\text{C} = 2 \times \delta^{13}\text{C}$, i.e. the values of δ for the two isotopes are exactly twice the one another. This may seem odd if you consider that the different behavior of isotopes is related solely to their atomic weight, so being the three carbon isotopes different to a unit of atomic weight (a neutron in addition) the increase for the ^{14}C must be twice that of the ^{13}C which is due, in turn such a change of one unit of atomic weight than the ^{12}C .

The measured activity must therefore be corrected to take into account the isotopic fractionation, which refers to the value of $\delta^{13}\text{C}$ of wood to which is assigned the conventional value - 25 ‰. The expression with which you apply that correction has a somewhat convoluted shape because the values of $\delta^{13}\text{C}$ are expressed per thousand and the correction can only be is a multiplication factor close to unity. In practice it is more helpful to look at how it can affect this correction

$$\Delta t = \tau \ln \left(\left(1 - 2 \frac{(25 + \delta^{13}\text{C})}{1000} \right)^{-1} \right) \approx 2 \frac{\tau}{1000} (25 + \delta^{13}\text{C}) = 2 \times 8,033 \times (25 + \delta^{13}\text{C}) \quad (5 \text{ x}).$$

In the table are the values of $\delta^{13}\text{C}$ and the corresponding corrections of dates (Δt) for some typical materials used for dating, the uncertainties indicated in the table apply only in the case of measures with the radiometric method as in the case of measures AMS the $\delta^{13}\text{C}$ is experimentally determined and have an uncertainty associated with the measure.

This is the reason why in conventional date calculation is applied the correction for the effect of isotopic fractionation, while for calibrations are not variations of ^{14}C in the atmosphere ($\Delta^{14}\text{C}$) which will be discussed later.

Table I- $\delta^{13}\text{C}$ and correction in years for samples extracted from some materials

MATERIAL	$\delta^{13}\text{C}$	Δt
MARINE CARBONATES AND PBD	1 ‰	429 ± 50
BONE HYDROXYAPATITE	0 ‰	412 ± 70
FRESHWATER SHELLS	-6 ‰	311 ± 50

BONE COLLAGEN	-7 ‰	295 ± 50
CO ₂ IN THE ATMOSPHERE	-8 ‰	278 ± 35
CEREALS (MAIZE, SORGHUM, ETC.)	-10 ‰	245 ± 50
MARINE PLANTS	-12 ‰	212 ± 70
STRAW AND FLAX	-14 ‰	179 ± 70
FRESHWATER PLANTS	-16 ‰	145 ± 130
BONE COLLAGEN, CELLULOSE	-19 ‰	97 ± 35
WHEAT, OATS, RICE, ETC.	-23 ‰	32 ± 70
WOOD AND CHARCOAL	-25 ‰	0 ± 70
LEAVES, STRAW, PEAT AND HUMUS	-27 ‰	32 ± 95

Errors in the measurement of the conventional date

Various reports with which calculates the conventional date are all exponential, so uncertainty about time is proportional to the relative error on the measurement of the activity of the sample (A) or, in the case of AMS, the determination of isotopic fraction $^{14}\text{C}/^{12}\text{C}$:

$$\Delta t = \tau \Delta/y = \tau \sigma_{in}/A. \quad (x 6)$$

Then with a little patience you can contain the statistical error and thereby reduce uncertainty about formal date, just think that a 1% error leads to an uncertainty in the conventional date of about 80 years. However, it is clear that when the ^{14}C decreases, and therefore is less its isotopic fraction, it is more difficult to maintain equal uncertainty levels, keeping in mind that the statistical error of measurement (σ_{stat}) is added with that measure quadratically (σ_{size}) and tool (σ_{strum}):

$$\sigma_{mis} = \sqrt{\sigma_{stat}^2 + \sigma_{mis}^2 + \sigma_{strum}^2} \quad (7 x)$$

and that while the statistical error can be reduced by counting for longer times, this is not the case for other errors. The uncertainty about the oldest dates, therefore, tends to grow although it is not possible to give a numerical value of this increase because it is highly dependent on conditions.

From conventional to the calibrated date

To pass from conventional date to the calibrated some correction methods which are often sophisticated and not easy to understand must be used. It worth to note at the outset that in providing the result of a measurement must always be given the date with its uncertainty interval in order to permit recalculation of calibration corrections, using the methods judged most appropriate. In any case, it is useful to point out that the calibrations are used to improve the accuracy of the results, so you can make an estimate nearest to the true, that would be the most probable on the base of measurements and our knowledge of systematic errors. Calibrations are also characterized by uncertainty ranges that determine an overall increase of error to give the date, in particular measurement errors (σ_{size}) are quadatically added to those introduced with the calibrations (σ_{cal}).

Calibration of initial concentration of ^{14}C

The radiocarbon method is based on the fact that the concentration of atoms of ^{14}C in the atmosphere remain constant over time. A possible variation leads to errors that are easily quantifiable. For example, if A_0 has changed of 1% this will produce a systematic displacement of all dates to higher values of 1%, then the

absolute value will grow linearly going back in time. For example the time measured will be higher five years in 1450 BP and ten years in 950 BP, then we are facing a systematic non-negligible error.

As early as 1958 De Vries emphasized that the concentration of radiocarbon in the 16th and 18th century was approximately 2% higher than that of the 19th century, attributing these changes to climatic variations. Subsequent research led to the discovery that the dating of artifacts from Egypt in the third millennium BC were moved forward in time a few hundred years. Following researches to see if such errors repeats in other ages, and what was the origin, made the idea of reconstructing the evolution of the concentration of ^{14}C in the atmosphere using biological systems that preserve track of those changes. Almost immediately were identified tree rings that have the unique characteristic of forming and die within a year and then remain trapped inside the stem without coming into contact with the atmosphere: a real fortune. Of all this there are numerous evidence that Plant Physiologists may explain in detail, besides the use of dendrochronology for dating the plants is well established for some time that has been evolving in recent decades with the development of methods for dating.

The systematic reconstruction of concentrations of ^{14}C in the past took a job of about 30 years, with the participation of numerous research groups that could verify with cross measures the accuracy of the results. Thanks to the presence of long-lived trees such as *Pinus Aristata*, *Sequoia Gigantea* and the Oaks, it was possible to determine an uninterrupted series of concentrations of up to 12,000 years ago. The resulting framework is rather complex because, as you can see from Figure 4, there were slow changes that seem to outline a periodic trends as short time fluctuations, also very significant. It is clear that long-term changes can be explained with great changes occurred in the past on Earth (such as changes in Earth magnetic field, which in a remote past has also reversed) while short term fluctuations are due to variations in activity of stars (including mainly those of the Sun) which are definitely also happened in the recent past. In Figure 4 the $\Delta^{14}\text{C}$, expressing these changes in for thousand, and defined as follows:

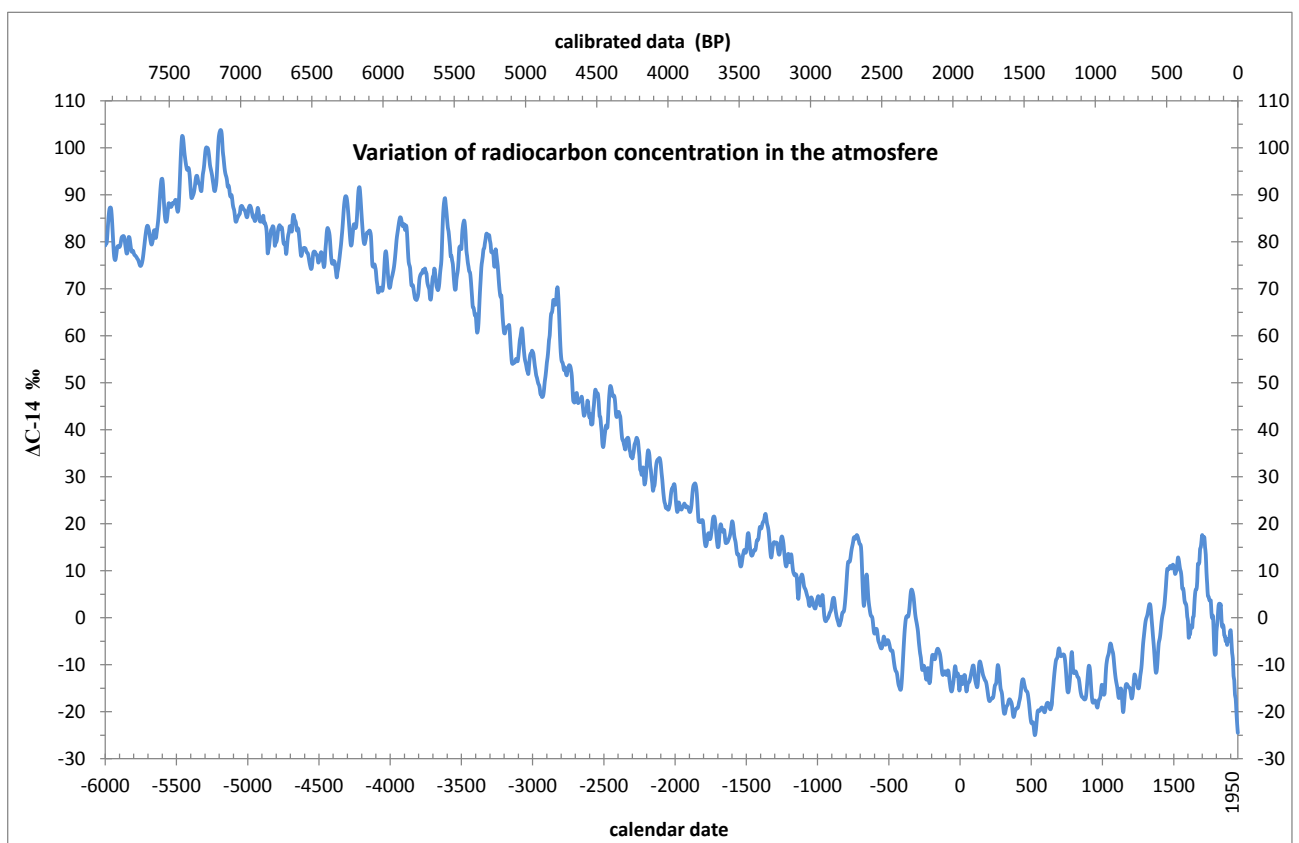


Figure 4- $\Delta^{14}\text{C}$ see equation x. 8) in the Earth's atmosphere over the past 9,000 years.

$$\Delta^{14}\text{C} = \frac{C_0 - C_0^*}{C_0^*} \times 1000 \quad (8 \text{ x})$$

where C_0 is the concentrations measured using tree rings and C_0^* is that found in Modern Reference Standard, which we have already spoken.

A first consideration - reassuring - is that in recent years 4000 fluctuations were all below 3% for which errors are contained. The second is that the presence of fluctuations creates a strong ambiguity in dating a sample, because conventional date, considering the uncertainty due to measurement errors and calibration, can match multiple calendar dates.

Using absolute chronological determinations on tree rings from the USA and Europe, it was possible to extend calibrations until 8329 BC, with a good degree of accuracy. Subsequent research, using the chronology of corals, dated with the Uranium-Thorium, allowed to extend calibration up to 13,635 BC and finally by the floating chronology of layered sediments (marine varves) from the Cariaco basin was able to get up to 24,000 BP. Such calibration applies, of course, the only living things that were for a long time in contact with atmospheric air, but cannot be applied to marine living organisms. Accurate researches using shells, corals and other marine species have made it possible to confirm that the hypothesis that there was also a dynamic equilibrium in the past between the largest reservoir of ^{14}C , i.e. the oceans, and the atmosphere is fully confirmed, as well as the fact that the concentration in the seas is slightly lower (approximately 5%). This indirect confirmation of the results obtained by dendrochronology (Figure 5) significantly strengthens confidence in the radiocarbon technique.

The knowledge of the $\Delta^{14}\text{C}$ allows us to calculate the calibrated radiocarbon dates, i.e. correct for the concentration of ^{14}C in the period in which the living system has accumulated carbon. The calibrated date differs from the conventional one (corrected for the actual value of the lifetime) the amount $\Delta t_{\text{cal}} = \tau \ln(1 + \Delta^{14}\text{C}/1000) \approx \Delta \tau \Delta^{14}\text{C}/1000$.

Figure 6 shows the calibration curve constructed with the most recent data available in the literature¹³, it was superimposed on the $\Delta^{14}\text{C}$ curve in the atmosphere in order to highlight better how are calibrations criticism at sudden variations in concentrations of ^{14}C .

Recent variations of the concentration of ^{14}C in the atmosphere

To some extent, as in a 'thriller', discovering the radiocarbon dating method is matched by a sudden change of the situation that makes more difficult the application of this method to recent date and for use its in the near future. To make the order, it is necessary to start from the end of the calibration curve of Figure 6. In an inset window in the figure shows the trend of the past 400 years, the calibration curve looks like anything but not a linear calibration, in fact there are continuous variations that create ambiguity to get a simple solution. In particular, there is a recommendation not to use the radiocarbon method for the past three centuries; It would seem a drastic limitation if you are not taking into account that it would be more used to authentic or other uses not strictly scientific, in that the datings are particularly useful as historical sources will deplete. Always looking at Figure 6, but would it be possible to do so, from Figure 5, that shows the same curve, you can observe that $\Delta^{14}\text{C}$ since the beginning of the 20th century suddenly decreases; This could be caused by the increase of coal 'off' in the atmosphere, due to the massive increase in combustion of coal and

¹³ PJ Reimer, MGL Baillie, and Bard, Bayliss, j.w. Beck, C Bertrand, PG Blackwell, EC Buck, Burr G, KB Cutler, Damon, PE RL Edwards RG, Fairbanks, m. Friedrich, TP, KA Guilderson Hughen, B Kromer, FG, McCormac S Manning, C Bronk Ramsey, RW Reimer, Remmele, JR S Southon, M Stuiver, S T, FW Taylor, J van der Plicht, and EC Weyhenmeyer (2004) Radiocarbon, 46:1029-1058.

hydrocarbons which occurred in that period. The fact is notable because it happens immediately after a resounding fact that changes dramatically the situation: a series of nuclear tests that last for about twenty years. In Figure 7 shows the trend of $\Delta^{14}\text{C}$ until 2001: you can see how the concentration of ^{14}C in the atmosphere grows suddenly coming to values never seen before. It is immediately apparent that the arrival of ^{14}C in the atmosphere takes place a few years delay: only after 1955 the $\Delta^{14}\text{C}$ quickly begins to grow, when bombs were detonated by time. However, after 1968 the concentration begins to decrease with an exponential trend that should predict that within a few decades the variations of carbon will be, as usual, those due to production by cosmic rays and not those induced by man. The curve appears indented as in recent decades have become very frequent determination of concentration of ^{14}C in the atmosphere, even monthly, so even small changes are evident, perhaps due to statistical fluctuations.

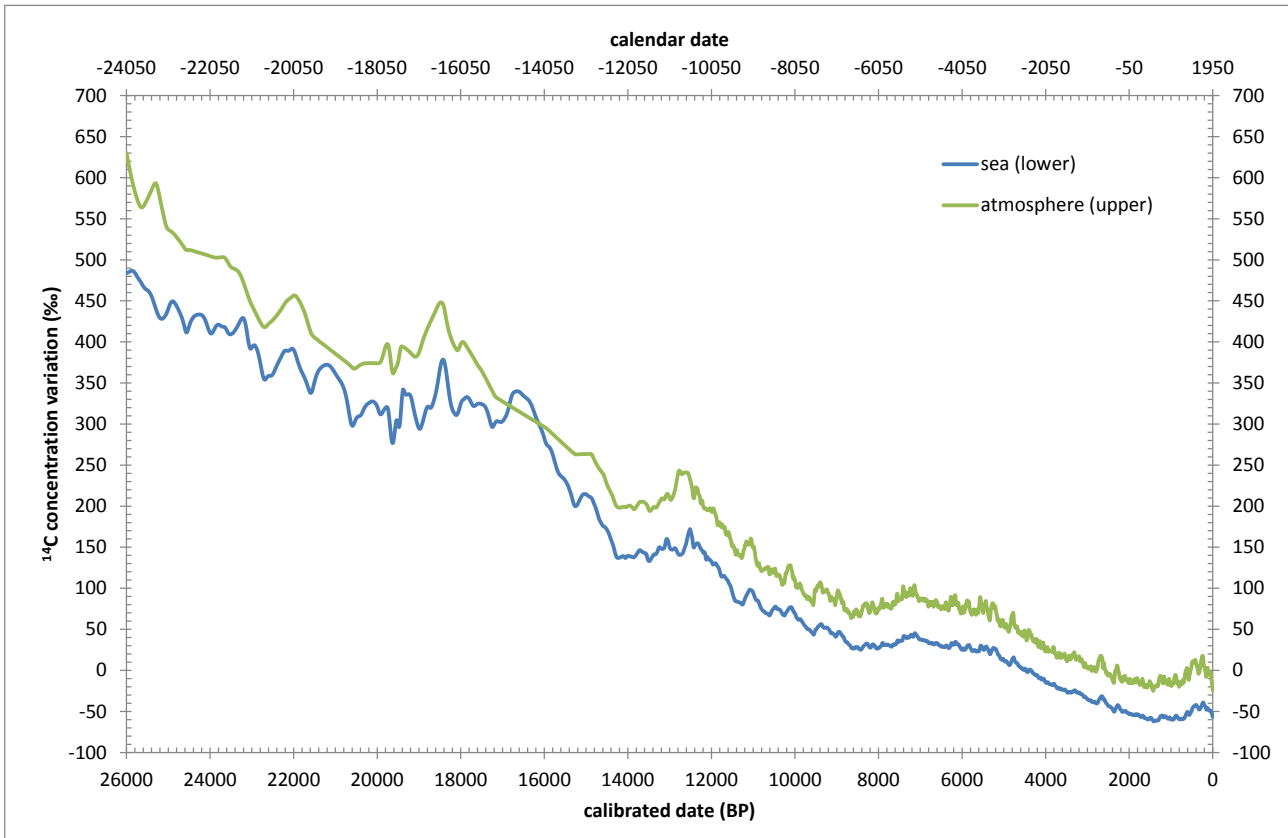


Fig. 5- $\Delta^{14}\text{C}$ in the sea compared with that in the atmosphere to show how they are related (related) between them.

Scientists are active people they have not do not claim for the great disaster of the nuclear age, indeed have sought new ways to take advantage of the new situation that had been created. In particular the large slope of the curve for about a decade after the 1950s allow to identify with high precision the year of manufacture of food (vintage, whiskey, etc.) through the direct determination of the concentration of ^{14}C that is characteristic, in those years, for a specific year. Both the climb and downhill curve allowed to refine knowledge of the dynamics of balance between the large tanks of ^{14}C , e.g. between the atmosphere and the hydrosphere (where is ending much of the ^{14}C produced during nuclear explosions), in explanation of his gradual disappearance from the atmosphere (fig. 7).

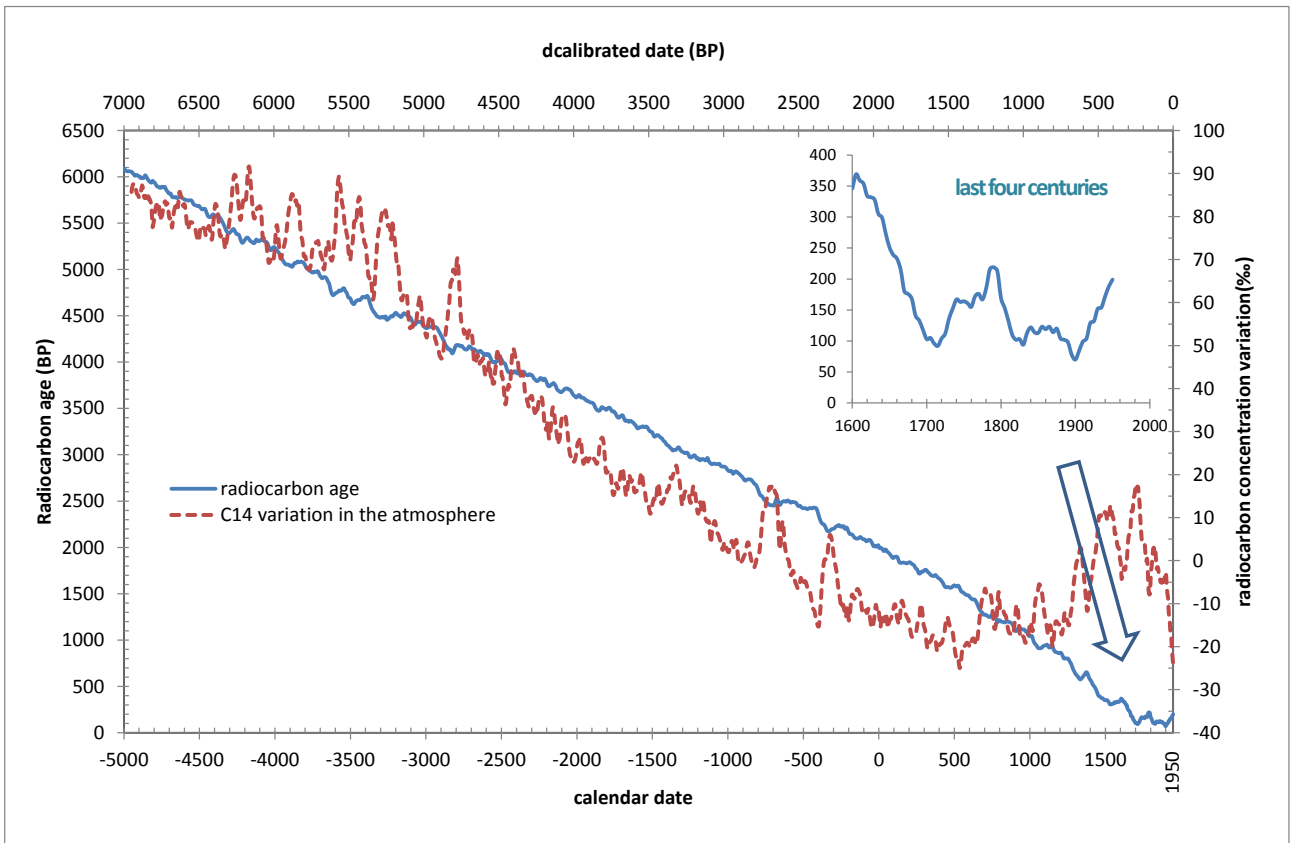


Fig. 6 calibration curve of radiocarbon age in the last 7,000 years. Overlay (with reference to the right vertical axis) $\Delta^{14}\text{C}$ in the atmosphere, to highlight how the latter variations affect calibration.

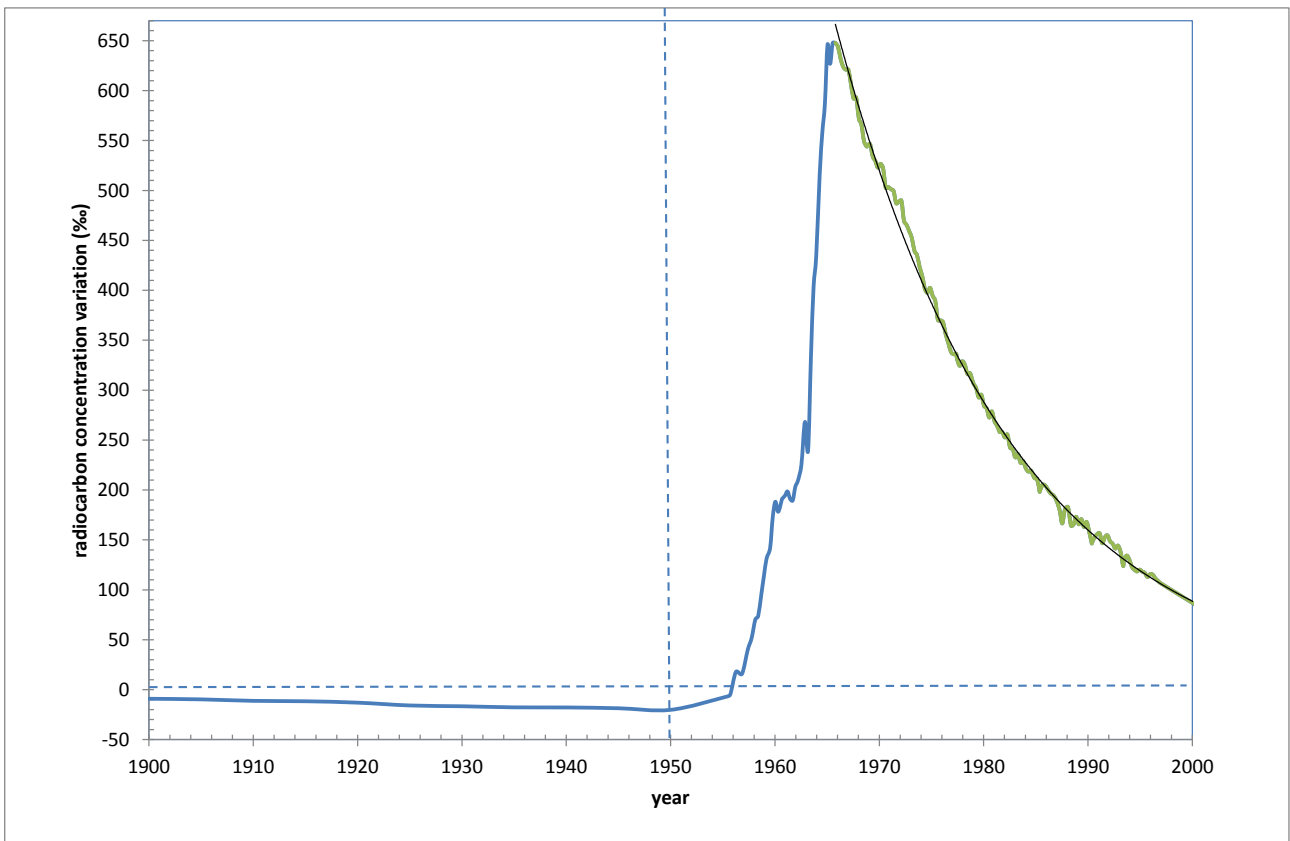


Fig. 7 – variation of $\Delta^{14}\text{C}$ due to nuclear explosions in the atmosphere (bomb effect).

How do you determine the calendar date

The decision to introduce a new scale of values, those BP, it may seem inappropriate if you consider a set of circumstances that would create confusion if you had decided to assign a calendar date from the beginning to the radiocarbon date. It should be noted that conventional dates, the result of a measurement, are often very different from those obtained after calibrating, as can be seen from Figure 6 by comparing conventional dates (also known as radiocarbon age) with the corresponding values, calibrated, located on the upper horizontal axis. For this the website of Oxford University laboratory¹⁴ - which is a real authority in the field of archaeometric research - states pique that the date produced by the radiocarbon method is not a calendar date. Always looking at Figure 6 you can notice that there is a unique and simple method to pass between calibrated and calendar date just subtract to 1950 the calibrated expressed in years BP. The result is the calendar date by placing to the negative numbers (taken in absolute value) the suffix a.C. (or BC in the international literature) while to the others the suffix d.C. (or A.D.).

As mentioned previously, the conventional date is characterized by an error, indicated by the sign \pm , which allows to calculate the interval in which the result fall with a given confidence level¹⁵. To some extent this is a sign that marks the difference between a calendar date and the result of applying a method of dating¹⁶. In particular it is assumed that the uncertainty about the measure of a radiocarbon age has a normal distribution (or Gaussian). This assumption is justified by the fact that there are several factors that contribute to this uncertainty, not only due to intrinsic decay distribution (that is Poisson and not normal distribution).

Calibration, therefore, allow to identify the correspondence between the result obtained with radiocarbon method and probable date of the calendar. Taking a step back, we can see how the data for calibrations look: so we understand better how to proceed.

Table II – calibration Data for the value of the initial concentration.

Calibrated date (BP)	¹⁴ C age (BP)	σ (BP)	Calendar Date
1975	2019	13	-25
1970	2014	14	-20
1965	2006	12	-15
1960	2014	13	-10
1955	2025	12	-5
1950	2012	12	0
1945	1992	12	5
1940	1991	13	10
1935	1994	13	15
1930	1988	14	20
1925	1977	13	25
....

¹⁴ <http://C14.arch.ox.ac.uk/calibration.html>

¹⁵ If α is the sum of the probabilities (also called cumulative probability) that the observed value falls within a given range, then the confidence interval (e.g. corresponding to two standard deviations around the mean) is that in which there is a cumulative probability $1-\alpha$ to find the result of subsequent measurements made under identical conditions. Suppose α to be 0.046 (which can be considered a quantification of risk considered acceptable) then the confidence was 95.4%.

¹⁶ Even historians, when sources are uncertain, sometimes use a time interval typically defined by extreme values within which date must necessarily fall. In this case, the interval has not a statistical significance, as for that of an estimated date by a dating method.

The first column shows the calibrated date (i.e. corrected for variation of concentration of ^{14}C in the atmosphere), the second shows the conventional date (or radiocarbon date) and the third the standard deviation measuring uncertainty on calibration. The last column shows, for completeness, the corresponding calendar date that matches the calibrated date.

These data are easily available in the net at several sites, those in the table are taken from the aforementioned article, Reimer et al. 2009. To stay updated on the calibration data is useful to consult one of several sites that calculate online the calibration, such as that of Oxford University laboratory that has developed a program for the calibration of the conventional dates: OxCal 4.1. In Figure 8 shows the visual interface of the program, which comes with three windows that you need to fill in: in the first you must provide a name for the file in which to store the calibration data, in the second - in a window called 'DATE' - the measured radiocarbon date, in the third (indicated by sign \pm) finally, insert the measurement error (the standard deviation). Then there is the possibility, by selecting them in the 'curve', using other calibration data instead of the newer, chosen automatically. The calculation provides the calendar date with uncertainty, which of course includes the calibration. Uncertainty about the determination of the date must be coupled quadratically with the calibration (also reported in table II, in the third column):

$$\sigma_t = \sqrt{\sigma_m^2 + \sigma_c^2} \quad (\times 9)$$

It is so defined an interval in which falls the calendar date sought with a probability of 95.4%, which is very high. A simple consideration that stems from what we have said is that calibration uncertainty increases, as it adds the part due to the calibration, but gets a value which is the closest to the real thing as possible, which for a time is essential.

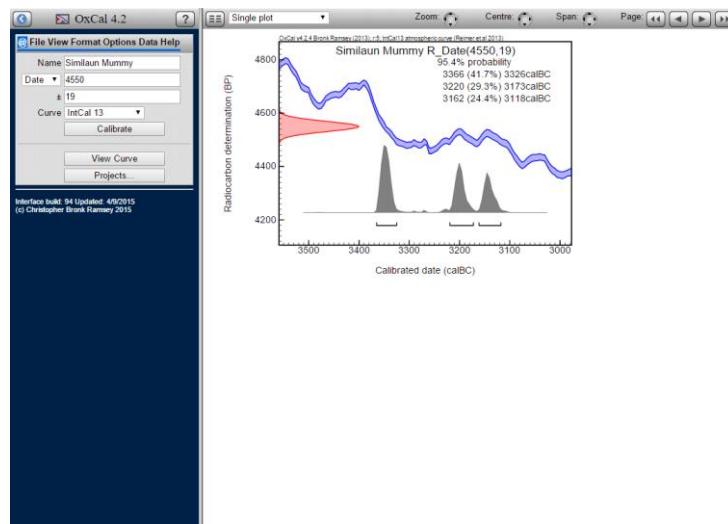


Figure 8 Visual interface of OxCal 4.2 program

The estimate of the date can then be refined by - selecting a special-button on the left side of the window that give the calibration result, Figure 8 below - calculating the intervals at which it can actually fall, and their percentages of probability. At this point it is necessary to make a concrete example: take the steps to estimate the age of the Similaun Mummy, fortuitously discovered in 1991 in the Ötztal Alps. The conventional date of 4550 ± 19 BP. Entering these date into the program OxCal 4.1 (latest version mentioned) you get an interval 3366-3118 BC with 95.4% confidence. If enabled, as stated before, the procedure detailed analysis shows that it is possible to estimate that leaves an uncertainty margin 32% (generally not recommended) that identifies three date ranges: 3335-3360 BC, with a probability of 34.1%, 3191-3210 BC 20.2% and probability, 3137-3152, 13.9% probability BC. There is then a four interval that overall leave just under 5% of uncertainty are: 3324-3366 BC, probability, 40.9% 3225-3231 b.c., 0.6% probability, 3173-3220 BC,

probability and 29.4% 3118-3162 BC, probability of 24.5%. How to read all this information? the recommended choice is to take a comprehensive range that includes all the ranges in which they can place the data sought. The data provided detailed analysis can be used for further considerations on the basis of other evidence or results found with other research.

Curiosity pushes us now understand better how these intervals were found. To do this you need to use a graphic (Figure 9) that allows a better understanding of the method followed. The graph shows a section of the calibration curve which allows to calculate the calendar date.

You will notice that the situation is somewhat complex because, given the uncertainty about the date measured, there is a single range of values to take into account. The chart on the left shows the curve that gives the probability distribution for the conventional date, from which you can calculate the two intervals of values highlighted by horizontal rectangles, corresponding to one and two standard deviations (σ). These rectangles intersect the calibration curve at several points, particularly the widest (2σ) intersects the calibration curve in seven points with bottom side and one in the upper one. To the eight points of intersection correspond the four intervals listed above. In particular the first two intersections delimit the first interval, the one with higher probability (40.9%); to calculate this probability, however, it is necessary to go ahead another step. The program OxCal calculates, along with the detail data discussed above, a table which gives the probability of having the date we are looking for in each of a hundred little intervals which divided the total (3366-3118 a.C.) calendar dates that hypothetically are possible. The little intervals are chosen to match those with which the calibration values were determined for, i.e. $\Delta^{14}\text{C}$ values (see for example Figure 4), choose smaller would do interpolations that don't improve the accuracy of calibration, choose the largest would be possible but practically not useful for analysis.

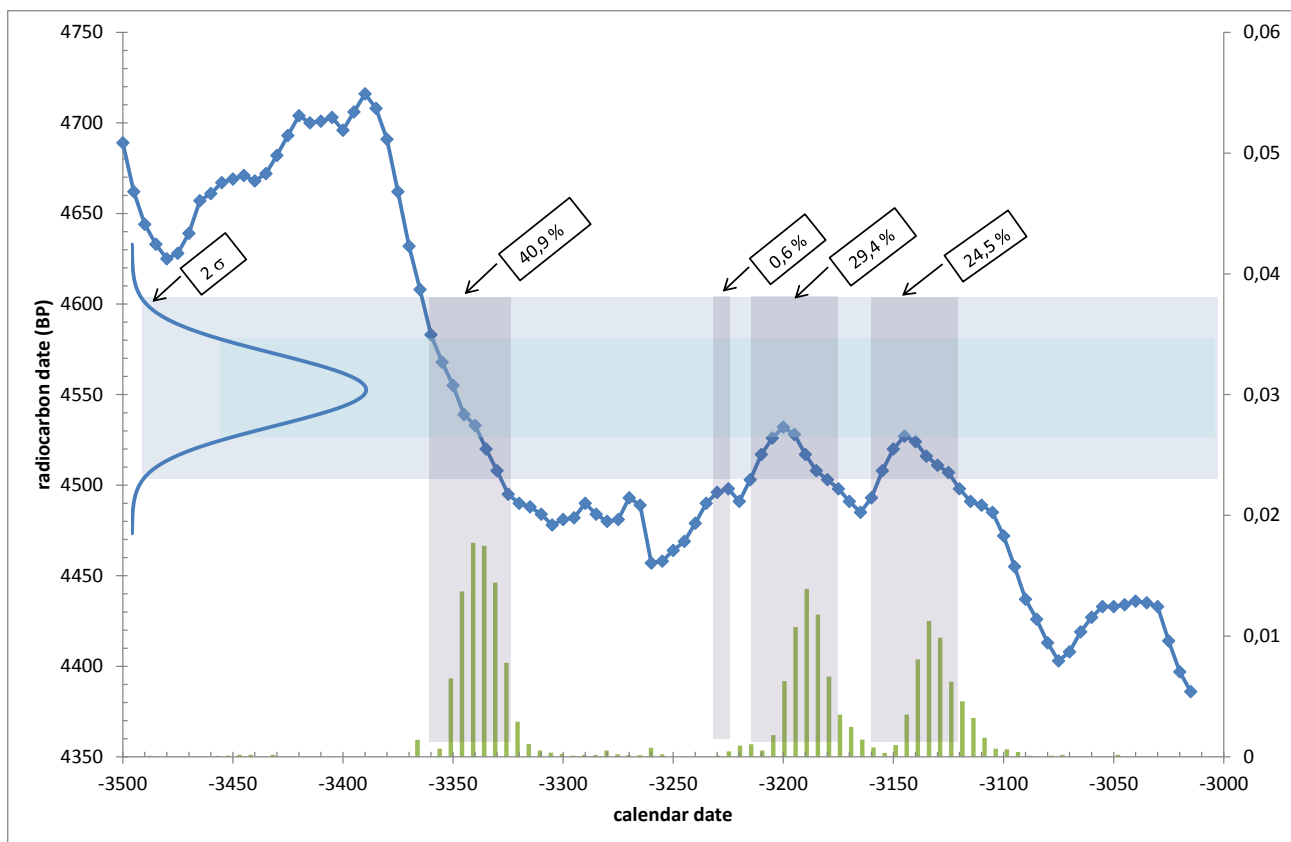


Figure 9 – Examples of the method in the calibration of the conventional date in the case of the mummy of Similaun.

Always in Figure 9, below, shows the histogram of probability of the date, using the table provided by the calibration program. It may be noted that in the histogram are three peaks that correspond roughly to three of the four vertical rectangles whose sides correspond to points, where the calibration curve intersects the horizontal rectangle delimiting the interval 2σ which reduces the uncertainty in the calibration result to less than 5%. It understands how we may lose in this reasoning but you must just reflect for a moment and try to generate the four vertical rectangles. In particular the fourth rectangle does not correspond to a peak of the histogram but to an accidental raising of the calibrating curve, which makes possible the intersection of 2σ rectangle. The probability for each date interval is calculated by adding the elements of the histogram (i.e. their values) included in it, so if the date interval corresponds to a marked histogram peak probability will be greater. It is clear that some dates intervals, which, however, belong to the total that come as result of dating, have low probability (the probability is less than 5% as a whole), then it is highly unlikely that the searched date falls within them.

Summing up the calibration, which is a vital step when you use the radiocarbon method allows to estimate a interval of values in which falls the date sought with a given probability (commonly that corresponds to a interval of 2σ). It is interesting to reflect on the marked difference with what happens to the conventional date where you can talk about an error, representing the uncertainty with a well-known distribution – that Gauss or normal – while in the case of the interval supplied with calibration is not entirely reasonable to talk about error: rather than uncertainty about the estimate. It is clear that seem sophisms but instead are useful reflections that allow you to examine a way of thinking that we can call 'probabilistic' which is that you work with in the field of dating.

Radiocarbon laboratories.

The AMS is gradually becoming the reference technique with radiocarbon dating, despite the high cost of the system and the bulk of the necessary instruments. It happened that the production of dates has become a high-quality service offered by some very specialized centers, which can have very competitive operating costs because they can save on management, also having a significant number of users. This is associated with the fact that a large Center can do multiple controls on the quality of the results and stay up to date on technical improvements and continuation of experiments will improve the reliability of the results obtained. The cost to get a date, are so greatly reduced that it makes no sense to create a Center for dating to economize on costs. This does not mean that it is not useful to create specialized centers that use services offered internationally and provide end users with better quality results. We are in a logic, which we can call globalization, which shows light and shadows but which is now the one that prevails in the area of services of the highest quality. A description of the experimental apparatus can be regarded as a mere curiosity: it's much better to focus on measurement techniques and how to avoid the errors that are always lurking. It is clear that a sophisticated instrumentation, as it may be, fails to remedy certain errors that were committed during the selection of samples and their preparation before sending to the Center. Finally, it will not be easy to correct mistakes committed in the conceptual planning phase of research, due to a deficient knowledge of the limits and characteristics of dating technique.

There are now numerous laboratories around the world that provide radiocarbon dating with both radiometric as *Accelerator Mass Spectrometry* (AMS) techniques.

RADIOCARBON LABORATORIES

This is *Radiocarbon's* annual list of active radiocarbon laboratories.

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