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P. J. Reimer, T. A. Brown, R. W. Reimer

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Radiocarbon

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Paula J. Reimer[1,2], Thomas A. Brown[1], Ron W. Reimer[2,3]

[1]Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, Livermore, CA, 94550, USA
[2]Queen's University Belfast, Belfast BT7 1NN, UK
[3]Ocean Sciences Department, University of California – Santa Cruz, Santa Cruz, CA 92697 USA

Abstract

The definitive paper by Stuiver and Polach (1977) established the conventions for reporting of ^{14}C data for chronological and geophysical studies based on the radioactive decay of ^{14}C in the sample since the year of sample death or formation. Several ways of reporting ^{14}C activity levels relative to a standard were also established, but no specific instructions were given for reporting nuclear weapons testing (post-bomb) ^{14}C levels in samples. Because the use of post-bomb ^{14}C is becoming more prevalent in forensics, biology, and geosciences, a convention needs to be adopted. We advocate the use of *fraction modern* with a new symbol $F^{14}C$ to prevent confusion with the previously used Fm, which may or may not have been fractionation corrected. We also discuss the calibration of post-bomb ^{14}C samples and the available datasets and compilations, but do not give a recommendation for a particular dataset.

Reporting of post-bomb ^{14}C data

Atmospheric nuclear weapons testing doubled the amount of ^{14}C in the atmosphere in the late 1950s and early 1960s. The use of this nuclear weapons testing (post-bomb) ^{14}C spike to provide age information in forensics, environmental forensics, biology and the geosciences has accelerated over the last few years (e.g. Campana and Jones, 1998; Kaplan, 2003; Kirner et al., 1997; Reddy et al., 2003; Wild et al., 1998) but there is no consensus as to what data should be reported in such studies. ^{14}C measurements of these samples cannot be considered indicative of an age. The ^{14}C content of post-bomb samples must be interpreted in relation to the ^{14}C content of the atmosphere or ocean reservoir, which has very little to do with the radioactive decay of ^{14}C. Negative radiocarbon ages have been utilized for the convenience of calibration with existing computer programs (Goslar et al., 2003). While this works mathematically, it is philosophically objectionable, because the decay of radiocarbon used to calculate the radiocarbon age is unrelated to time of formation of a post-bomb sample. Negative radiocarbon ages could also provoke a misunderstanding or mistrust of ^{14}C analyses in general.

The basic information needed for comparing the ^{14}C content in a post-bomb sample at the time of growth or formation to that of the atmosphere or ocean is the ratio of the sample activity to the standard activity measured in the same year, both activities background corrected and $\delta^{13}C$-normalized, which is equivalent to A_{SN}/A_{ON} in the notation of Stuiver and Polach (1977). The decay counting activity ratio is equivalent to the ratio of the sample $^{14}C/^{13}C$ (or $^{14}C/^{12}C$) isotope ratio to the standard $^{14}C/^{13}C$ (or $^{14}C/^{12}C$) isotope ratio

measured by accelerator mass spectrometry (AMS) in the same year, both ratios background corrected and $\delta^{13}C$-normalized, which is also known as *fraction modern* or F_m (Donahue et al., 1990). Unfortunately the term *fraction modern* has been used with and without $\delta^{13}C$-normalization of the sample activity. The term *Percent modern* (pM) can cause confusion since *"absolute" percent Modern* is also in use for geochemical and equilibria studies and the symbol is widely used to stand for picomoles. The terms $\Delta^{14}C$ and $D^{14}C$ are a step away from the basic data of interest in that they represent fractional deviation from the standard activity. Also, there is potential for confusion of $\Delta^{14}C$ with Δ, which is age-corrected for year of sample growth.

$\Delta^{14}C$ is a very useful way of reporting ^{14}C measurements for geochemical studies, including comparisons to model results. Unfortunately, under Stuiver and Polach's definition, $\Delta^{14}C$ is based on A_{SN}/A_{abs}, and the value obtained for a sample grown/formed in a particular year depends on the year in which it is measured; e.g., a sample grown/formed in 1962 will give a different $\Delta^{14}C$ if measured today versus if it had been measured in 1962. Hence, relating the $\Delta^{14}C$ value measured today of a forensics sample which grew/formed in an unknown year, to bomb-^{14}C records, based on samples measured at various times and expressed in $\Delta^{14}C$ units, is problematic. For forensics and similar studies such difficulties would be avoided if the ^{14}C values obtained for the unknown samples and for the bomb-^{14}C records were expressed as ratios that do not change with time (i.e., A_{SN}/A_{ON} rather than A_{SN}/A_{abs}). While the difference between A_{SN}/A_{abs} and A_{SN}/A_{ON} is small at present, it will become more important as time progresses. The ratio A_{SN}/A_{ON} has also been given the symbol $^{14}a_N$ (Mook and van der Plicht, 1999) but this nomenclature has not been widely adopted. We suspect this is due to a reluctance to depart from the Stuiver and Polach (1977) definitions, and because the symbols do not convey the information that carbon is involved. We propose to establish $F^{14}C$ as an unequivocal term that is in keeping with the Stuiver and Polach (1977) A_{SN}/A_{ON} definition, yet conveys the information needed for atom counting and decay counting measurements in bomb-^{14}C based studies.

It is worth noting at this point that $\delta^{13}C$-normalization differs between radiocarbon methods that measure the $^{14}C/^{12}C$ activity or isotope ratio (all radiometric methods and many AMS systems) or $^{14}C/^{13}C$ isotope ratio (some AMS systems). While the laboratories generally supply data normalized to –25 ‰ with respect to VPDB, in some cases the $\delta^{13}C$ is measured or estimated at a later time and retroactive corrections must be made. Because of this difference, a correction for a 1 ‰ shift in $\delta^{13}C$ results in a correction factor to $F^{14}C$ of approximately 0.002 for a $^{14}C/^{12}C$ activity or isotope ratio measurement or 0.001 for a $^{14}C/^{13}C$ isotope ratio measurement. This is equivalent to approximately a 16-year and 8-year correction to the radiocarbon age, respectively. For clarity we reiterate the basic equations for $\delta^{13}C$-normalization of the sample for 1) $^{14}C/^{12}C$ measurements (Stuiver and Robinson, 1974) and 2) $^{14}C/^{13}C$ measurements (Brown, 1994; Donahue et al., 1990), substituting $F^{14}C$ for A_{SN}/A_{ON}:

$$1)\ F^{14}C = (A_S/0.95\ A_{OX}) \cdot (0.975/0.981)^2 \cdot [(1+\delta^{13}C_{OX}/1000)/(1+\delta^{13}C_S/1000)]^2$$

where A is the activity or $^{14}C/^{12}C$ isotope ratio, and subscripts $_S$ and $_{OX}$ refer to sample and oxalic acid standard, respectively;

2) $F^{14}C = (R_S/0.95\ R_{OX}) \cdot (0.975/0.981)^2 \cdot (1+\delta^{13}C_{OX}/1000)\ /(1+\delta^{13}C_S/1000)$

where R is the $^{14}C/^{13}C$ isotope ratio and subscripts $_S$ and $_{OX}$ as above.

Therefore, if a sample has been normalized with an estimated value of $\delta^{13}C$ and the oxalic acid normalized to $\delta^{13}C_{OX} = -19$ ‰, then the following formulae apply to the retroactive correction for a measured $\delta^{13}C_S$:

1′) $F^{14}C = F^{14}C_{est} \cdot [(1+\delta^{13}C_{est}/1000)/(1+\delta^{13}C_S/1000)]^2$; for $^{14}C/^{12}C$ measurements,

and

2′) $F^{14}C = F^{14}C_{est} \cdot (1+\delta^{13}C_{est}/1000)/(1+\delta^{13}C_S/1000)$; for $^{14}C/^{13}C$ measurements.

Note that these corrections can be applied to radiocarbon ages, since $t = -8033 \cdot \ln(F^{14}C)$.

Calibration of post-bomb ^{14}C data

Comparison of atmospheric $^{14}CO_2$ records indicates that the distribution of bomb ^{14}C at the height of nuclear testing was not nearly as uniform as pre-bomb ^{14}C (Levin and Kromer, 1997; Manning and Melhuish, 1994; Nydal and Lovseth, 1983; Tans, 1981). In addition CO_2 from fossil fuel, which is depleted in ^{14}C, is non-uniformly distributed and can be a substantial contribution of carbon to a sample (Levin et al., 2003). In the tropics ^{14}C-enriched CO_2 released from the terrestrial biosphere may result in slightly elevated ^{14}C levels compared to mid-northern hemispheric ones in recent decades (Levin and Hesshaimer, 2000; Randerson et al., 2002). Therefore a regional, or even a local, atmospheric ^{14}C dataset is the ideal for calibration of a post-bomb ^{14}C measurement. However, it is not feasible to develop a local calibration dataset in most cases. A number of post-bomb atmospheric ^{14}C records are available (Levin and Kromer, 1997; Levin and Kromer, this issue; Manning and Melhuish, 1994; Nydal and Lovseth, 1983). These long-term observations provide the best record of atmospheric ^{14}C values at their respective locations.

Tree-rings and other organic material also provide a record of growing season averaged ^{14}C, provided mobile carbon compounds are removed during pretreatment (Stuiver and Quay, 1981). Hua and Barbetti (this issue) have compiled zonal averages of ^{14}C data derived from atmospheric, tree-ring and organic materials for the Southern Hemisphere and three zones in the Northern Hemisphere including a zone following the Northern Hemisphere summer Inter-Tropical Convergence Zone (ITCZ). These compilations, together with the summer means from the atmospheric observations (Levin and Kromer, this issue), should provide adequate calibration for most purposes. However, while the Southern Hemisphere is represented by one zonal compilation, mixing is likely to have an

influence along the ITCZ. Growing season differences should also be considered especially for the tropics and for high latitude sites, and during periods of rapid change in the atmospheric ^{14}C levels. Subannual measurements may be necessary to capture the rapid response of tree cellulose to atmospheric ^{14}C levels (Grootes et al., 1989).

Marine datasets derived from coral, coraline sponges, fish ootoliths, and shell chronologies are also available for post-bomb calibration of marine samples but show higher regional variation (Druffel, 1996; Druffel and Griffin, 1995; Fallon et al., 2003; Guilderson et al., 2000; Nydal et al., 1984; Weidman and Jones, 1993).

In addition to needing a calibration dataset that reflects the ^{14}C content of the atmosphere or ocean in the locality of the sample growth, it is necessary to consider that some types of samples may have incorporated carbon from numerous sources. Modern diets and petroleum based carbon compounds can introduce additional uncertainty in the calibration. Turnover time of human or animal tissues is dependent on the type of tissue involved and may be affected by age or health of the organism (Geyh, 2001; Harkness and Walton, 1972; Lovell et al., 2002; Stenhouse and Baxter, 1977). Proximity to discharge from nuclear reactors or medical waste incinerators can introduce additional pulses of ^{14}C, which may not be observed in the regional or zonal calibration datasets (Cook et al., 1995; Trumbore et al., 2002) although atmospheric mixing may be rapid enough in some cases to dilute a pulse beyond detection (McGee et al., 2004).

Post-bomb calibration programs

Because the ^{14}C content of the atmosphere changed rapidly, especially during the years immediately preceding the nuclear test ban treaty, computer programs that are used to calibrate post-bomb ^{14}C data must step through the calibration dataset in smaller increments than is normally done in calibration programs, as noted by Puchegger et al. (2000). The resulting calibrated age ranges are thus given in smaller increments. It must be realized that these narrow ranges may not be completely realistic given the uncertainties discussed above. The calibration program assumes that the sample is from a system closed to carbon exchange after its formation. It is therefore not appropriate for use on open systems such as soil carbon where more complex modeling is required to understand the carbon dynamics (Trumbore, 2000).

We have constructed a post-bomb calibration program with graphical user interface for use on Macintosh OSX or Windows operating systems. The program CaliBomb allows the selection of calibration datasets or a user-defined local dataset. It is up to the user to choose or construct the appropriate dataset for the region of interest. The datasets and compilations provided have been extended into the past with tree-ring measurements from the appropriate hemisphere (McCormac et al., 2002; Stuiver et al., 1998) to provide seamless calibration for modern samples. A moving average of the dataset may be used to approximate the length of time over which the sample accumulated carbon. An example of the output is given in Figure 1.

Conclusion

It is recommended that $F^{14}C$ be used to report ^{14}C measurements of post-bomb samples. As with all ^{14}C measurements, the measured or estimated $\delta^{13}C$ should be reported. The atmospheric post-bomb calibration datasets and compilations discussed above and the program CaliBomb are available on the Radiocarbon website at www.radiocarbon.org or at www.calib.org.

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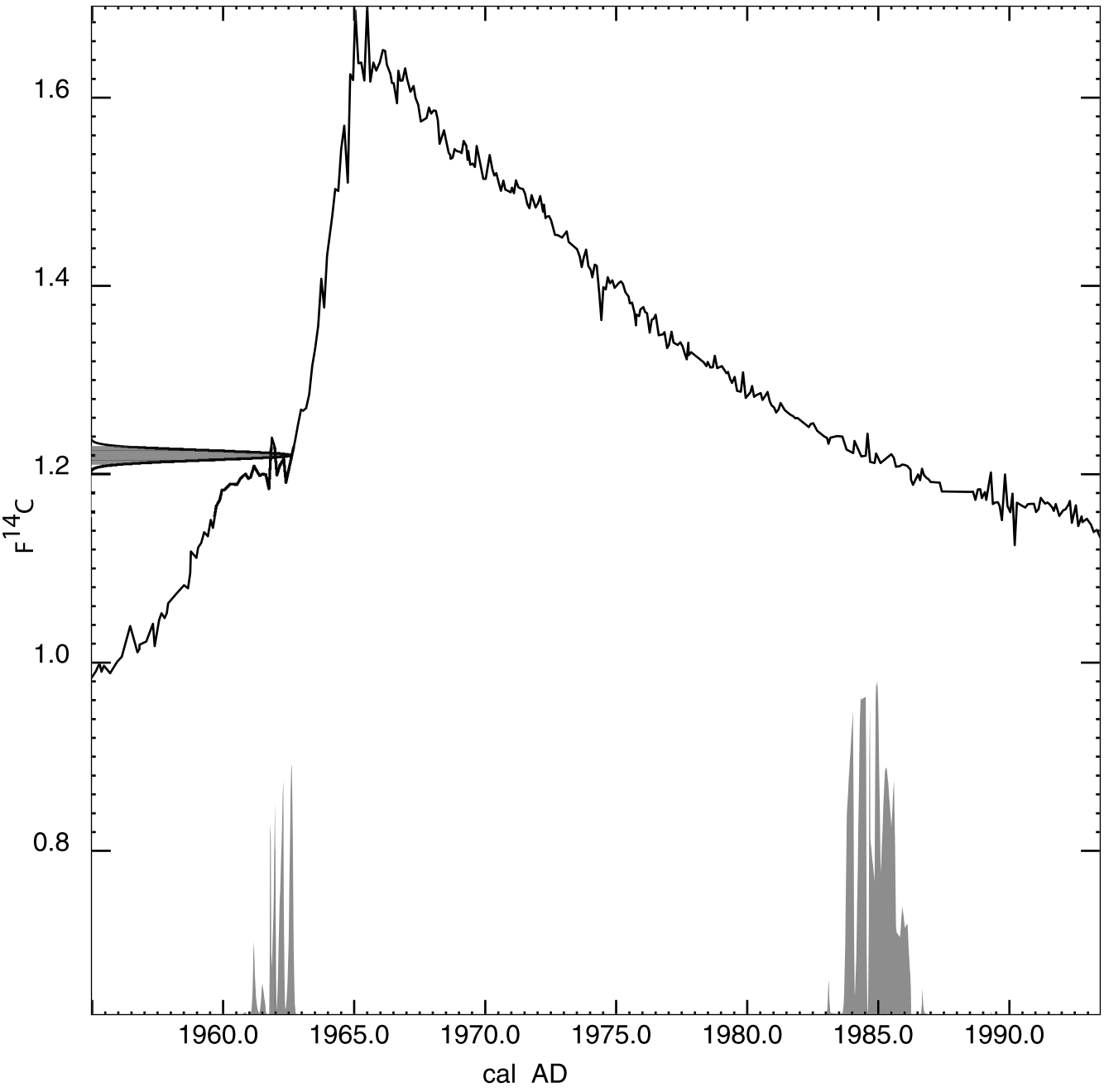
Figure captions

Figure 1. Output from the program CaliBomb for the calibration of a hypothetical sample with $F^{14}C = 1.220 +/- .005$. The Southern Hemisphere post-bomb dataset from Wellington, New Zealand (Manning and Melhuish, 1994) was converted to $F^{14}C$ for this purpose, assuming the atmospheric samples were measured in the year of collection. The two sigma calibrated probability ranges are shown on the calendar axis.


1.6
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