



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Discussion: Reporting and calibration of post-bomb ^{14}C data

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

October 12, 2004

Radiocarbon

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

Discussion: Reporting and calibration of post-bomb ^{14}C data
Paula J. Reimer^{1,2}, Thomas A. Brown¹, Ron W. Reimer^{2,3}

¹Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, Livermore, CA, 94550, USA

²Queen's University Belfast, Belfast BT7 1NN, UK

³Ocean 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 $F^{14}\text{C}$ to prevent confusion with the previously used F_m , 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 $\delta^{13}\text{C}$ -normalized, which is equivalent to $A_{\text{SN}}/A_{\text{ON}}$ in the notation of Stuiver and Polach (1977). The decay counting activity ratio is equivalent to the ratio of the sample $^{14}\text{C}/^{13}\text{C}$ (or $^{14}\text{C}/^{12}\text{C}$) isotope ratio to the standard $^{14}\text{C}/^{13}\text{C}$ (or $^{14}\text{C}/^{12}\text{C}$) isotope ratio

measured by accelerator mass spectrometry (AMS) in the same year, both ratios background corrected and $\delta^{13}\text{C}$ -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 $\delta^{13}\text{C}$ -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 $\Delta^{14}\text{C}$ 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 $\Delta^{14}\text{C}$ with Δ , which is age-corrected for year of sample growth.

$\Delta^{14}\text{C}$ 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, $\Delta^{14}\text{C}$ is based on $A_{\text{SN}}/A_{\text{abs}}$, 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 $\Delta^{14}\text{C}$ if measured today versus if it had been measured in 1962. Hence, relating the $\Delta^{14}\text{C}$ 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 $\Delta^{14}\text{C}$ 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., $A_{\text{SN}}/A_{\text{ON}}$ rather than $A_{\text{SN}}/A_{\text{abs}}$). While the difference between $A_{\text{SN}}/A_{\text{abs}}$ and $A_{\text{SN}}/A_{\text{ON}}$ is small at present, it will become more important as time progresses. The ratio $A_{\text{SN}}/A_{\text{ON}}$ has also been given the symbol $^{14}a_{\text{N}}$ (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 $F^{14}\text{C}$ as an unequivocal term that is in keeping with the Stuiver and Polach (1977) $A_{\text{SN}}/A_{\text{ON}}$ 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 $\delta^{13}\text{C}$ -normalization differs between radiocarbon methods that measure the $^{14}\text{C}/^{12}\text{C}$ activity or isotope ratio (all radiometric methods and many AMS systems) or $^{14}\text{C}/^{13}\text{C}$ isotope ratio (some AMS systems). While the laboratories generally supply data normalized to -25‰ with respect to VPDB, in some cases the $\delta^{13}\text{C}$ 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 $\delta^{13}\text{C}$ results in a correction factor to $F^{14}\text{C}$ of approximately 0.002 for a $^{14}\text{C}/^{12}\text{C}$ activity or isotope ratio measurement or 0.001 for a $^{14}\text{C}/^{13}\text{C}$ 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 $\delta^{13}\text{C}$ -normalization of the sample for 1) $^{14}\text{C}/^{12}\text{C}$ measurements (Stuiver and Robinson, 1974) and 2) $^{14}\text{C}/^{13}\text{C}$ measurements (Brown, 1994; Donahue et al., 1990), substituting $F^{14}\text{C}$ for $A_{\text{SN}}/A_{\text{ON}}$:

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

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

$$2) F^{14}\text{C} = (R_s/0.95 R_{\text{OX}}) \cdot (0.975/0.981)^2 \cdot (1+\delta^{13}\text{C}_{\text{OX}}/1000) / (1+\delta^{13}\text{C}_s/1000)$$

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

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

$$1') F^{14}\text{C} = F^{14}\text{C}_{\text{est}} \cdot [(1+\delta^{13}\text{C}_{\text{est}}/1000)/(1+\delta^{13}\text{C}_s/1000)]^2; \text{ for } ^{14}\text{C}/^{12}\text{C} \text{ measurements,}$$

and

$$2') F^{14}\text{C} = F^{14}\text{C}_{\text{est}} \cdot (1+\delta^{13}\text{C}_{\text{est}}/1000)/(1+\delta^{13}\text{C}_s/1000); \text{ for } ^{14}\text{C}/^{13}\text{C} \text{ measurements.}$$

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

Calibration of post-bomb ^{14}C data

Comparison of atmospheric $^{14}\text{CO}_2$ 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, coralline sponges, fish otoliths, 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 $F^{14}\text{C}$ be used to report ^{14}C measurements of post-bomb samples. As with all ^{14}C measurements, the measured or estimated $\delta^{13}\text{C}$ 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.

Acknowledgements

This work was funded in part by NSF grant ATM-0407554. A portion of this work was performed under the auspices of the U. S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

References:

- Brown, TA. 1994. "Radiocarbon dating of pollen by accelerator mass spectrometry." Unpublished Ph.D. thesis, University of Washington.
- Campana, SE, and Jones, CM. 1998. Radiocarbon from nuclear testing applied to age validation of black drum, *Pogonias cromis*. *Fishery Bulletin* **96**, 185-92.
- Cook, GT, Begg, FH, Naysmith, P, Scott, EM, and McCartney, M. 1995. Anthropogenic C-14 marine geochemistry in the vicinity of a nuclear fuel reprocessing plant. *Radiocarbon* **37**, 459-67.
- Donahue, DJ, Linick, TW, and Jull, AJT. 1990. Isotope-ratio and background corrections for accelerator mass spectrometry radiocarbon measurements. *Radiocarbon* **32**, 135-42.
- Druffel, ERM. 1996. Post-bomb radiocarbon records of surface corals from the tropical Atlantic Ocean. *Radiocarbon* **38**, 563-72.
- Druffel, ERM, and Griffin, S. 1995. Regional variability of surface ocean radiocarbon from southern great barrier reef corals. *Radiocarbon* **37**, 517-24.
- Fallon, SJ, Guilderson, TP, and Caldeira, K. 2003. Carbon isotope constraints on vertical mixing and air-sea CO_2 exchange. *Geophysical Research Letters* **30**, doi:10.1029/2003/GL018049.
- Geyh, MA. 2001. Bomb radiocarbon dating of animal tissues and hair. *Radiocarbon* **43**, 723-30.
- Goslar, T, van der Knaap, WO, Hicks, S, Rasanen, S, Andric, M, Czernik, J, and Goslar, E. 2003. ^{14}C dating of modern peat profiles: pre- and post-bomb ^{14}C variations in the construction of age-depth models. In "18th International Radiocarbon Conference." Wellington, New Zealand.
- Grootes, PM, Farwell, GW, Schmidt, FH, Leach, DD, and Stuiver, M. 1989. Rapid response of tree cellulose radiocarbon content to changes in atmospheric $^{14}\text{CO}_2$ concentration. *Tellus* **41B**, 134-48.
- Guilderson, TP, Schrag, DP, Goddard, E, Kashgarian, M, Wellington, GM, and Linsley, BK. 2000. Southwest subtropical pacific surface water radiocarbon in a high-resolution coral record. *Radiocarbon* **42**, 249-56.
- Harkness, DD, and Walton, A. 1972. Further investigations of the transfer of bomb ^{14}C to man. *Nature* **240**, 302-03.

- Hua, Q, and Barbetti, M. this issue. Compilation of tropospheric bomb ^{14}C data for carbon cycle modeling and age calibration purposes. *Radiocarbon* **46**.
- Kaplan, IR. 2003. Age dating of environmental organic residues. *Environmental Forensics* **4**, 95-141.
- Kirner, DL, Burky, R, Taylor, RE, and Southon, JR. 1997. Radiocarbon dating organic residues at the microgram level. *Nuclear Instruments & Methods in Physics Research Section B- Beam Interactions with Materials and Atoms* **123**, 214-17.
- Levin, I, and Hesshaimer, V. 2000. Radiocarbon - A unique tracer of global carbon cycle dynamics. *Radiocarbon* **42**, 69-80.
- Levin, I, and Kromer, B. 1997. Twenty years of atmospheric (CO_2)-C-14 observations at Schauinsland station, Germany. *Radiocarbon* **39**, 205-18.
- Levin, I, and Kromer, B. this issue. The tropospheric $^{14}\text{CO}_2$ level in mid latitudes of the Northern Hemisphere (1959-2003). *Radiocarbon* **46**.
- Levin, I, Kromer, B, Schmidt, M, and Sartorius, H. 2003. A novel approach for independent budgeting of fossil fuel CO_2 over Europe by $^{14}\text{CO}_2$ observations. *Geophysical Research Letters* **30** Art. No. 2194, Art. No. 2194.
- Lovell, MA, Robertson, JD, Buchholz, BA, Xie, C, and Markesbery, WR. 2002. Use of bomb pulse carbon-14 to age senile plaques and neurofibrillary tangles in Alzheimer's disease. *Neurobiology of Aging* **23**, 179-86.
- Manning, MR, and Melhuish, WH. 1994. Atmospheric $\Delta^{14}\text{C}$ record from Wellington. In "Trends: A Compendium of Data on Global Change." pp. 173-202. ORNL/CDIAC-65. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge.
- McCormac, FG, Reimer, PJ, Hogg, AG, Higham, TFG, Baillie, MGL, Palmer, J, and Stuiver, M. 2002. Calibration of the radiocarbon time scale for the southern hemisphere: AD 1850-950. *Radiocarbon* **44**, 641-51.
- McGee, EJ, Gallagher, D, Mitchell, PI, Baillie, MGL, Brown, D, and Keogh, SM. 2004. Recent chronologies for tree rings and terrestrial archives using ^{14}C bomb fallout history. *Geochimica et Cosmochimica Acta* **68**, 2509-16.
- Mook, WG, and van der Plicht, J. 1999. Reporting C-14 activities and concentrations. *Radiocarbon* **41**, 227-39.
- Nydal, R, Gulliksen, S, Lovseth, K, and Skogseth, FH. 1984. Bomb C-14 in the Ocean Surface 1966-1981. *Radiocarbon* **26**, 7-45.
- Nydal, R, and Lovseth, K. 1983. Tracing Bomb C-14 in the Atmosphere 1962-1980. *Journal of Geophysical Research-Oceans and Atmospheres* **88**, 3621-42.
- Puchegger, S, Rom, W, and Steier, P. 2000. Automated evaluation of ^{14}C AMS measurements. *Nuclear Instruments & Methods in Physics Research B* **172**, 274-80.
- Randerson, JT, Enting, IG, Schuur, EAG, Caldeira, K, and Fung, IY. 2002. Seasonal and latitudinal variability of troposphere $\Delta^{14}\text{CO}_2$: Post bomb contributions from fossil fuels, oceans, the stratosphere, and the terrestrial biosphere. *Global Biogeochemical Cycles* **16**, Article no. 1112.
- Reddy, CM, Xu, L, and O'Connor, R. 2003. Using radiocarbon to apportion sources of polycyclic aromatic hydrocarbons in household soot. *Environmental Forensics* **4**, 191-97.
- Stenhouse, MJ, and Baxter, MS. 1977. Bomb ^{14}C as a biological tracer. *Nature* **267**, 828-

- Stuiver, M, and Polach, HA. 1977. Discussion: reporting of ^{14}C data. *Radiocarbon* **19**, 355-63.
- Stuiver, M, and Quay, PD. 1981. Atmospheric C-14 Changes Resulting from Fossil-Fuel CO_2 Release and Cosmic-Ray Flux Variability. *Earth and Planetary Science Letters* **53**, 349-62.
- Stuiver, M, Reimer, PJ, and Braziunas, TF. 1998. High-precision radiocarbon age calibration for terrestrial and marine samples. *Radiocarbon* **40**, 1127-51.
- Stuiver, M, and Robinson, SW. 1974. University of Washington Geosecs North Atlantic carbon-14 results. *Earth and Planetary Science Letters* **23**, 87-90.
- Tans, P. 1981. A compilation of bomb ^{14}C data for use in global carbon model calculations. In "Carbon Cycle Modelling (SCOPE 16)." (B Bolin, Ed.), pp. 131-57. John Wiley and Sons, New York.
- Trumbore, S. 2000. Age of soil organic matter and soil respiration: Radiocarbon constraints on belowground C dynamics. *Ecological Applications* **10**, 399-411.
- Trumbore, S, Gaudinski, JB, Hanson, PJ, and Southon, JR. 2002. Quantifying ecosystem-atmosphere carbon exchange with a ^{14}C label. *EOS, Transactions, AGU* **83**, 267-68.
- Weidman, CR, and Jones, GA. 1993. A Shell-Derived Time History of Bomb C-14 on Georges Bank and Its Labrador Sea Implications. *Journal of Geophysical Research-Oceans* **98**, 14577-88.
- Wild, E, Golser, R, Hille, P, Kutschera, W, Priller, A, Puchegger, S, Rom, W, Steier, P, and Vycudilik, V. 1998. First C-14 results from archaeological and forensic studies at the Vienna Environmental Research Accelerator. *Radiocarbon* **40**, 273-81.

Figure captions

Figure 1. Output from the program CaliBomb for the calibration of a hypothetical sample with $F^{14}C = 1.220 \pm .005$. 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.

