14C in aerosols:what F14C value to use for contemporary carbon

Stenström, Kristina; Genberg, Johan

2009

Kristina Stenström and Johan Genberg
Lund University, Sweden
Report nr: LUNFD6(NFFR-3106)/1-4/2009

^{14}C in aerosols: what F^{14}C value to use for contemporary carbon

Kristina Stenström and Johan Genberg
Lund University, Department of Physics, Divsion of Nuclear Physics, Box 118, SE-221 00 Lund, Sweden

INTRODUCTION

Information about the source of carbonaceous aerosols can be obtained by measuring the $^{14}C/^{12}C$ ratio (leading to a F^{14}C value) in aerosol samples. ^{14}C is a naturally occurring radionuclide with a half-life of 5730 years. Thus carbon in aerosols from sources of fossil origin (such as coal and oil, which are several million years old) will be free from ^{14}C, since all ^{14}C has decayed. On the other hand, contemporary carbon (found in aerosols originating e.g. from combustion of biomass) has a concentration of ^{14}C which may be estimated from the atmospheric concentration. Measurement of the $^{14}C/^{12}C$ ratio is performed by accelerator mass spectrometry (AMS). What F^{14}C value to use for contemporary carbon is complicated by at least two factors, both related to human activities: bomb-^{14}C and ^{14}C from the nuclear industry.

BOMB-^{14}C

The first factor that needs to be considered concerns the circumstance that atmospheric nuclear weapons testing in the late 1950s and early 1960s temporarily almost doubled the amount of ^{14}C in atmospheric CO_2. Since the test ban in 1963, the atmospheric ^{14}C specific activity has decreased due to the uptake of CO_2 in the oceans and the biosphere and due to fossil fuel ^{14}C-free CO_2 input. The bomb-pulse in atmosphere at "clean-air" sites at different latitudes has been extensively studied and monitored (see e.g. Levin (2004); Levin et al. (2008)) as shown in Fig.1.

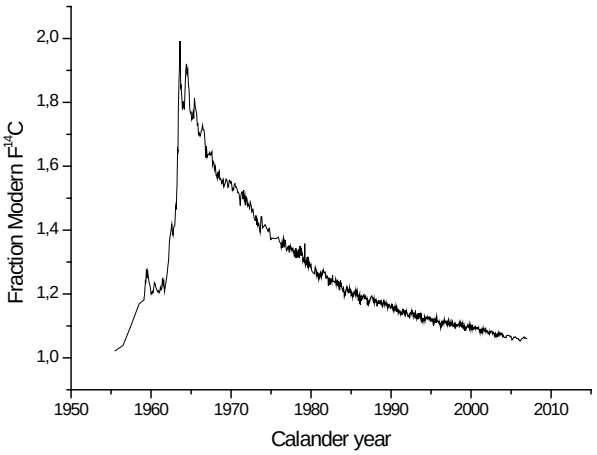

Figure 1. Atmospheric concentration of ^{14}C in CO_2 in the northern hemisphere. Atmospheric testing of nuclear weapons during the late 1950s to early 1960s produced large amounts of ^{14}C. The highest level was observed in 1963, when the specific activity of ^{14}C in atmospheric CO_2 was about twice the natural level (expressed in units of *fraction modern*, F^{14}C; F^{14}C is approximately equal to 1.0 before the nuclear weapons test (Reimer et al, 2004)). Since the test ban in 1963, atmospheric ^{14}C specific activity has decreased due to uptake of CO_2 in oceans and biosphere and due to fossil fuel ^{14}C-free CO_2 input. Data are taken from Levin and Kromer (2004), Levin et al (2008), Hua and Barbetti (2004) and http://intcal.qub.ac.uk/CALIBomb/frameset.html.

Because of the tremendous increase in ^{14}C in the atmosphere due to the nuclear weapon tests in the 1950s and 60s, the amount of ^{14}C in biomass varies with the age of the tree. Lewis et al (2004) has combined the atmospheric ^{14}C values with a growth function of a tree to calculate how the ^{14}C concentration varies with the age of the tree. In their paper the calculation is done for trees harvested in 1999.

The equation they used is: $F^{14}C_{biomass} = \int_{t1}^{t2} F^{14}C_A(t)\ w(t)dt / \int_{t1}^{t2} w(t)dt$

where $F^{14}C_A$ is the fraction modern carbon in the atmospheric CO_2 at time *t*. *w* is a weighting function to determine the increase of carbon in the biomass as a function to time.

w is calculated using the Chapman-Richards growth model presented in Lewis et al. (2004):

$$V = A * (1 - e^{-(t-t_0)/\tau})^m$$

In the equation *V* is the volume of the tree or the biomass of a community. *t* is the year of measurement while t_0 is year when the tree starts to grow. *V* is derived in order to acquire *w(t)* for the equation above. *A*, τ and *m* are selected to fit the growth. Lewis et al. (2004) used $\tau = 50$ and $m = 3$.

We have used the same equation and parameters to calculate $F^{14}C_{biomass}$ using values from atmospheric measurements by Levin et al. (2008 and personal communication) and Stuiver and Quay (1981) depending on age of the tree harvested in 2007. The results are shown in Fig. 2, which can be used to estimate $F^{14}C_{biomass}$ depending on the age of the combusted wood.

The effect is most important for trees which are between 60 and 70 years old. A "standard tree" has to be assumed in order to derive the $F^{14}C$ content in biomass burning. In Sweden this "standard" age is between 60 and 80 years, i.e. $F^{14}C_{biomass}$ = 1.21 – 1.23. For more recent wood or for combustion of seasonal vegetation a more recent $F^{14}C$ needs to be used.

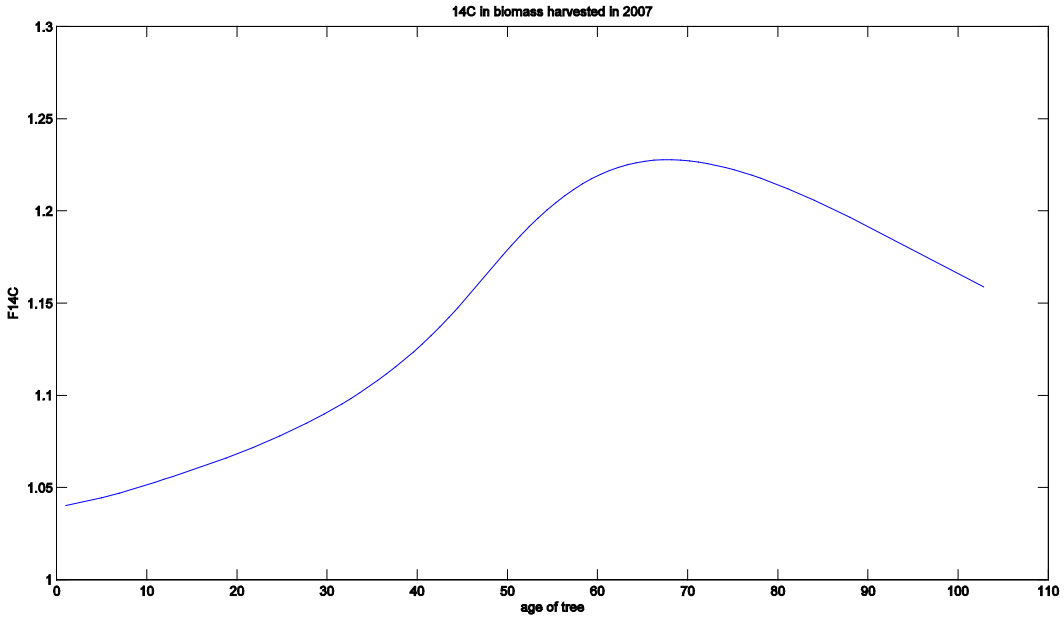

Figure 2. $F^{14}C$ in biomass harvested in 2007. The graph can be used to estimate $F^{14}C_{biomass}$ depending on the age of the combusted wood.

^{14}C FROM THE NUCLEAR INDUSTRY

The second factor that can influence ^{14}C on aerosol filters relates to the production of ^{14}C nuclear power reactors and the use of ^{14}C as a tracer in research and industry.

In nuclear power reactors, part of the ^{14}C produced is continuously released as airborne effluents, mainly as CO_2 and hydrocarbons. ^{14}C can be traced in the local environment of nuclear installations, see e.g. Stenström et al. (2009a). The maximum excess in vegetation is often found within a few km of nuclear power plants (depending e.g. on reactor type and operation, stack height and weather conditions). A typical value for light-water reactors is a maximum excess of 10% within a few km of the power plant. Heavy-water reactors (HWRs) may have a greater effect, since these are known to produce more ^{14}C than most other types of reactors (Magnusson 2007). Milton et al. (1995) reported values of up to 58 times the contemporary level in fruit (year 1992) close to HWRs in Canada ($F^{14}C \approx 57.6$). We have previously presented values for greater distances from these Canadian HWR: $F^{14}C=6.84\pm0.15$ at 2 km from the facility in 1998, and even at a distance of 100 km an excess of 7% was found ($F^{14}C=1.20\pm0.03$) (Stenström et al, 2009a). ^{14}C levels in the vicinity of the Lithuanian nuclear power plant Ignalina (graphite-moderated) have also been investigated (Magnusson et al. 2004; Magnusson et al. 2007; Adliene et al. 2006). Soil samples close to the power plant showed highly elevated ^{14}C levels (up to 20 times the contemporary background), which may indicate releases of particulate material. Very little is known about how potential, particulate ^{14}C releases from nuclear facilities influence ^{14}C aerosol source apportionment measurements.

Another example of anthropogenic ^{14}C sources concerns the use as ^{14}C as a tracer in medicine and industry, e.g. the pharmaceutical industry. In one study (Stenström 2009b) it was seen that human hair can be contaminated by airborne ^{14}C-labelled compounds in ^{14}C-using laboratories, and that this contamination is very difficult to remove from the hair samples. There is also a risk that quartz filters for aerosol sampling can be contaminated by ^{14}C-labelled compounds. However, this is probably only an issue in the local environment of such facilities.

CONCLUSIONS

The $F^{14}C$ value for biomass can be estimated from Fig. 2 using information about the average age of the biomass combusted in the area in question.

Aerosol sampling at urban sites for subsequent ^{14}C analysis needs to address if there are any laboratories using ^{14}C in the vicinity. Sampling at rural sites needs to consider the potential influence of ^{14}C released from nuclear installations located nearby.

REFERENCES

Adliene D, Rääf C, Magnusson Å, Behring J, Zakaria M, Adlys G, Skog G, Stenström K, Mattsson S. Assessment of the environmental contamination with long-lived radionuclides around an operating RBMK reactor station. *Journal of Environmental Radioactivity* 90 (2006) 68-77.

Hua Q, Barbetti M. 2004. Review of tropospheric bomb ^{14}C data for carbon cycle modeling and age calibration purposes. *Radiocarbon* 46(3):1273-1298.

Levin I, Kromer B. 2004. The tropospheric $^{14}CO_2$ level in mid-latitudes of the northern hemisphere (1919-2003). *Radiocarbon* 46(3) 1261-1272.

Levin I, Hammer S, Kromer B, Meinhardt F. 2008. Radiocarbon observations in atmospheric CO_2: Determining fossil fuel CO_2 over Europe using Jungfraujoch observations as background. *Science of the Total Environment* 391:211-216.

Levin I, personal communication.

Lewis CW, Klouda GA, Ellenson WD. 2004. Radiocarbon measurement of the biogenic contribution to summertime PM-2.5 ambient aerosol in Nashville, TN. *Atmospheric Environment* 38, 6053-6061.

Milton GM, Kramer SJ, Brown RM, Repta CJW, King KJ, Rao RR. 1995. Radiocarbon dispersion around Canadian nuclear facilities. *Radiocarbon* 37(2) 485-496.

Magnusson Å, Stenström K, Skog G, Adliene D, Adlys G, Hellborg R, Olariu O, Zakaria M, Rääf C, Mattsson S. 2004. ^{14}C levels in the terrestrial environment in the vicinity of two European nuclear power plants. *Radiocarbon* 46(2) 863-868.

Magnusson Å. 2007. ^{14}C produced by nuclear power reactors – generation and characterization of gaseous, liquid and solid waste. Doctoral Dissertation, Lund University.

Magnusson Å, Stenström K, Adliene D, Adlys G, Dias C, Rääf C, Skog G, Zakari M, Mattsson S. 2007. Carbon-14 levels in the vicinity of the Lithuanian nuclear power plant Ignalina. *Nuclear Instruments and Methods* B259, 530-535.

http://intcal.qub.ac.uk/CALIBomb/frameset.html

Reimer PJ, Brown TA, Reimer RW. 2004. Discussion: Reporting and calibration of post-bomb ^{14}C data. *Radiocarbon* 46:1299-1304.

Stenström K, Unkel I, Nilsson CM, Rääf C, Mattsson S. 2009a. The use of hair as an indicator of occupational ^{14}C contamination. *Journal of Radiation and Environmental Biophysics* in press. DOI 10.1007/s00411-009-0245-9.

Stenström K, Skog G, Nilsson CM, Hellborg R, Leide Svegborn S, Georgiadou E, Mattsson S. 2009b. Local variations in ^{14}C – how is bomb-pulse dating of human cells and tissues affected? *Nuclear Instruments and Methods B* in press. doi:10.1016/j.nimb.2009.10.157

Stuiver M, Quay PD. 1981. Atmospheric ^{14}C changes resulting from fossil fuel CO_2 release and cosmic ray flux variability. *Earth and Planetary Science letters* 53:349-362.