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# SUBTLE $^{14}C$ SIGNALS: THE INFLUENCE OF ATMOSPHERIC MIXING, GROWING SEASON AND IN-SITU PRODUCTION

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**ABSTRACT.** Atmospheric $^{14}C$ concentrations vary with time and latitude. These variations, measured directly on atmospheric samples, or in independently-dated organic material such as tree rings, supply data essential for the calibration of dynamic models of the global carbon cycle. Short variations in the production rate of atmospheric $^{14}C$ are strongly attenuated in the relatively large atmospheric $CO_2$ reservoir. *In-situ* production of $^{14}C$ should be negligible for ages up to 80 ka BP. Background problems in AMS dating are more likely attributable to contamination of very small samples.

## INTRODUCTION

A major factor in the success of radiocarbon as a dating tool is that its atmospheric residence time is long enough for atmospheric mixing to produce nearly uniform $^{14}C$ concentrations throughout the troposphere. Deviations from this uniformity, which can be measured directly on atmospheric samples or on organic materials, are small. These subtle deviations are, however, of considerable interest because they provide details of the cycling of carbon within a reservoir and between reservoirs, as well as of the spatial and temporal variability of the production of cosmogenic isotopes and their injection into the troposphere. Because all processes act simultaneously and semi-independently on $^{14}C$ concentrations, a dynamic model of the interacting atmosphere-ocean-biosphere system is needed to fully interpret observed $^{14}C$ concentration changes.

## SPATIAL AND TEMPORAL VARIABILITY

Spatial and temporal variability in atmospheric $^{14}C$ concentrations have been made quite "visible" by the nuclear bomb tests of the early 1960s. The familiar increase in atmospheric $^{14}CO_2$ in the northern hemisphere shows a clear latitude dependence (Fig. 1). The maximum $\Delta^{14}C$ value obtained decreases with latitude and is delayed by one year for a lower latitude station, such as Trapani (38°02′N). The $\Delta^{14}C$ values for Nordkapp (71°06′N) and Vermunt (46°55′N) show a cross-over, with values at Vermunt being higher than those at Nordkapp in spring (20‰ May 1962, 70‰ in June 1963), but lower in fall (−50‰ in September 1962, −110‰ in October-November 1963). This dynamic behavior cannot be obtained from atmospheric box models. However, several features have already been reproduced in runs of a three-dimensional global tracer transport model (Braziunas, personal communication 1991). Detailed time series of atmospheric $^{14}CO_2$ concentrations at different locations are needed to fine-tune the model.

During the early 1960s, only a few stations collected atmospheric $CO_2$ samples. Consequently, we have relied on tree rings and other dated organic materials for additional information on the atmospheric response to the bomb $^{14}C$ input (Fig. 2). A proper interpretation of such proxy data for this period of rapidly changing $^{14}CO_2$ concentrations obviously requires that the time period when the carbon was taken up by the plant is known (see *e.g.*, the $^{14}C$ concentration profile of the 1963 ring of a Sitka spruce in Grootes *et al.* (1989), Fig. 1). A one-month delay in the start of the growing season, related, for example, to high elevation or latitude, increased in 1963 the average ring $\Delta^{14}C$ value by ~35‰. Samples containing carbon photo-assimilated over several months show an average $\Delta^{14}C$ value that was highest in 1964. I plotted the $\Delta^{14}C$ values for July 1, the midpoint of the growing season. The growth-weighted average time may be slightly different.

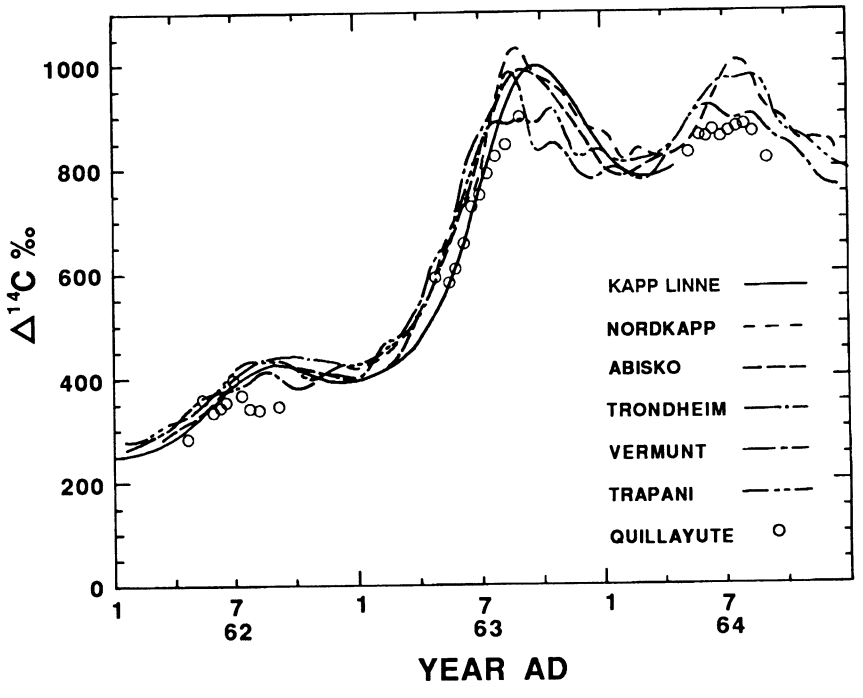


Fig. 1. $^{14}C$ concentrations of atmospheric $CO_2$ following the 1961/1962 nuclear bomb tests: Kapp Linne (78°04′N, 13°38′E) (Olsson & Karlén 1965); Nordkapp (71°06′N, 23°59′E) (Nydal & Lövseth 1983); Abisko (68°20′N, 18°49′E) (Olsson & Karlén 1965); Trondheim (63°N, 10°E) (Nydal & Lövseth 1983); Vermunt (46°55′N, 10°03′E) (Levin *et al.* 1985); Trapani (38°02′N, 12°31′E) (Münnich & Roether 1967); Quillayute Sitka spruce (47°57′N, 124°33′W) (Grootes *et al.* 1989).

The $^{14}C$ concentration of tree rings and other organic materials generally matches the tropospheric $^{14}C$ concentration quite well. Higher-latitude materials contain higher $\Delta^{14}C$, just as in the atmosphere. The Mingyin spruce (27°13′N, 100°20′E) samples the air of the 0–30° N tropical Hadley circulation cell, and, consequently, shows considerably lower $\Delta^{14}C$ values in 1963 and 1964. The Mackenzie delta white spruce shows high $\Delta^{14}C$ values for 1962 and 1963. The results from this tree have been used to argue that slow meridional mixing in the troposphere can result in significant latitudinal concentration gradients in the northern hemisphere troposphere (Dai & Fan 1986). This conclusion then supported the large (10‰) amplitude $\Delta^{14}C$ signal attributed to the 11-year sunspot cycle deduced from ring measurements on this tree (Fan *et al.* 1983, 1986). Comparison with atmospheric $\Delta^{14}C$ values at high northern-latitude stations (Trondheim, 63°N, Abisko, 68°20′N, Nordkapp, 71°06′N, and Kapp Linne, 78°04′N) shows that the $^{14}C$ value of 606‰ of the Mackenzie spruce (68°N) is ~150‰ higher than values observed in the atmosphere in 1962. This indicates that the sample dated as the 1962 ring included lignin and/or oils and resins of 1963 or later. Similarly, some 1964 material may contribute to the high 1963 $\Delta^{14}C$ value. Although direct atmospheric $CO_2$ measurements show that latitudinal $\Delta^{14}C$ gradients exist, they are significantly smaller and more variable than indicated by Dai and Fan (1986).

We may conclude that tree rings and short-lived organic materials closely monitor atmospheric $^{14}CO_2$ concentrations. The pattern of spatial and temporal variability in $\Delta^{14}C$ observed for the bomb $^{14}C$ in the northern hemisphere also applies to the injection into the troposphere of natural, *i.e.*, cosmogenic stratospheric $^{14}C$. Provided the exact period of uptake of the measured carbon is known, and the measuring precision is adequate, tree rings can be used to determine natural latitudinal and short-term temporal variations of $^{14}C$ in the atmosphere.

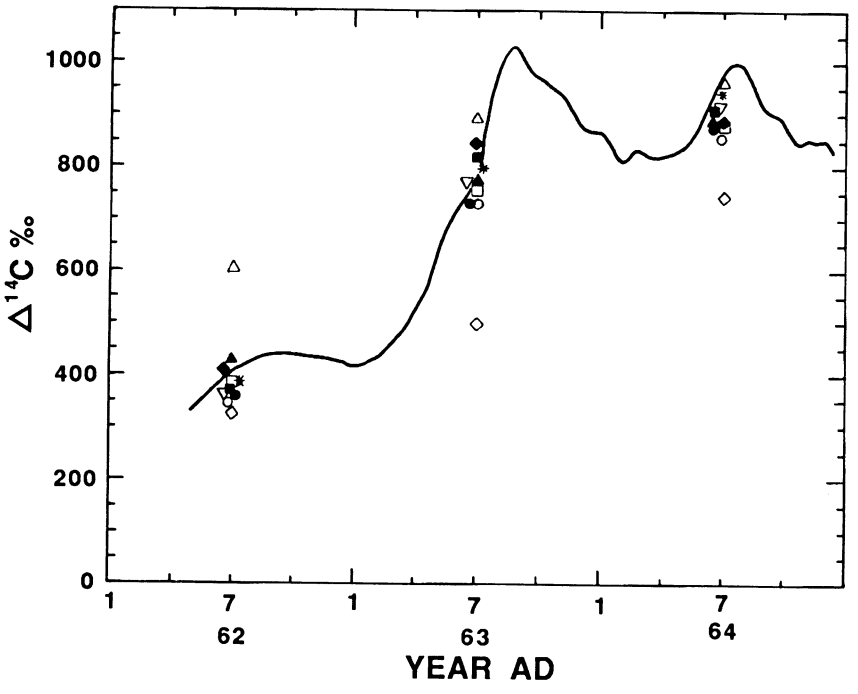


Fig. 2. Atmospheric $^{14}CO_2$ concentrations as measured at Nordkapp (71°06'N, 23°59'E) and Trondheim (63°N, 10°E) (Nydal & Lövseth 1983) and $^{14}C$ concentration in organic samples from various latitudes. ▵ = Mackenzie delta (68°N, 130°W) (Dai & Fan 1986); □ = Lowland USSR (60°N, 31°E) (Kolesnikov, Gorshkova & Biryulin 1970); ◇ = Mingyin, China (27°N, 100°E) (Dai & Fan 1986); ▲ = Treeless Height, USSR (Kolesnikov, Gorshkova & Biryulin 1970); ✳ = Copenhagen, Denmark (56°N, 13°E) (Tauber 1967); ♦ = Kiel, Germany (54°N, 10°E) (Willkomm & Erlenkeuser 1968); ○ = Quillayute, USA (48°N, 125°W) (Grootes *et al.* 1989); ▽ = Dailing, China (48°N, 129°E) (Dai & Fan 1986); • = New York, USA (41°N, 74°W) (Cain & Suess 1976); ■ = Georgia, USSR (38°N, 45°E) (Burchuladze *et al.* 1989).

Temporal variations of $^{14}C$ concentration in the atmosphere have been known since the 1950s. The advent of high-precision $^{14}C$ dating has led to a detailed calibration of atmospheric $^{14}C$ variability against tree-ring chronology throughout the Holocene. This record of $^{14}C$ variability has been used not only to calibrate $^{14}C$ dates, but also to derive the periodicity of factors influencing the $^{14}C$ production rate. Stuiver & Quay (1980) established two different modes by which the sun modulates cosmogenic isotope production on a century time scale. Several papers devoted to the shorter Schwabe or "sunspot" cycle, have, however, yielded conflicting conclusions about its magnitude (Baxter & Farmer 1973; Damon, Long & Wallick 1973; Burchuladze *et al.* 1980; Fan *et al.* 1986; Stuiver & Quay 1981). Using a three-dimensional global tracer transport model, Braziunas (personal communication 1991) obtains peak-to-peak values of 2–4‰ for the 11-year sunspot cycle (dependent on the strength of the solar forcing). This agrees with results of earlier box-model calculations (Damon, Long & Wallick 1973) and high-precision tree-ring measurements (Stuiver & Quay 1981). Because of its small size, the sunspot cycle in $\Delta^{14}C$ is very hard to detect, even with high-precision (≤2‰) counters. For $^{10}Be$, which does not have a significant atmospheric reservoir, concentration variations corresponding to the 11-year sunspot cycle have been observed in the Dye-3 Greenland ice core (Beer *et al.* 1988).

When the tracer-transport model is used with a seasonally varying injection of stratospheric $^{14}C$ into the troposphere, as well as a wind-dependent ocean-atmosphere exchange, it produces a small seasonal $^{14}C$ concentration cycle and a latitudinal $^{14}C$ concentration gradient (Braziunas, personal

communication 1991). Levin *et al.* (1989) estimate 3–5‰ for the seasonal $\Delta^{14}C$ cycle in Europe. A question is whether the seasonal cycle can amplify the 11-year sunspot cycle at high latitudes. The nuclear bomb-test $^{14}C$ has shown little delay between the production of stratospheric $^{14}C$ and its injection into the troposphere. Increased $^{14}C$ production related to lower solar activity will be followed by increased injection of stratospheric $^{14}C$ into the troposphere the next year. The maximum/minimum seasonal $\Delta^{14}C$ cycles will be found when the 11-year $^{14}C$ concentration cycle passes through its average value. At the extremes of the $^{14}C$ sunspot cycle, the seasonal $\Delta^{14}C$ cycle will be of average amplitude. Thus, the seasonal $\Delta^{14}C$ cycle does not contribute to the amplitude of the 11-year sunspot cycle, and we expect an effect of the order of 2–4‰. Fan *et al.* (1983, 1986) reported larger values for a white spruce in the Mackenzie delta (10‰ peak-to-peak) as did Burchuladze *et al.* (1980) for Georgian wines (4.3 ± 1.1‰ amplitude). The highest correlation between $\Delta^{14}C$ of the white spruce and sunspot number, obtained when $\Delta^{14}C$ values were delayed by 4 yr, was −0.38 (−0.48 when the AD 1823–1833 cycle was excluded) (Fan *et al.* 1986). This correlation explains only 15% (23% with exclusion) of the observed variance, and the observed variance in the Mackenzie delta spruce is not incompatible with a much smaller true sunspot $\Delta^{14}C$ cycle in tropospheric $CO_2$ (See Damon *et al.* 1992).

## IN-SITU $^{14}C$ PRODUCTION AND $^{14}C$ BACKGROUND

Production of $^{14}C$ occurs not only in the atmosphere, but also at the Earth's surface and inside the Earth by cosmic radiation and radiation from natural radioactive elements. This so-called "*in-situ*" production is the basis of exposure age dating and measurements of accumulation and erosion/ablation rates. $^{14}C$ production *in situ* in a sample that is to be dated or used as a background material is, however, a continuing worry for $^{14}C$ dating. As part of a $^{14}C$ enrichment and dating project, I checked the likely *in-situ* production of $^{14}C$ for a buried peat sample and for the anthracite used as background standard in the Groningen $^{14}C$ laboratory (Table 1). For these materials, the most important reaction for *in-situ* production is $^{14}N(n, p)^{14}C$ induced by low-energy (<1000 eV) neutrons. The contribution of fast neutrons to this reaction and that of reactions of the type $^{13}C(n, \gamma)^{14}C$ is small (~10%, Sawelski 1968). The neutron flux through a sample buried underground is produced by cosmic radiation (at greater depth almost exclusively muons) and by spontaneous fission reactions of uranium in the soil. We can calculate the activity of the sample due to the *in-situ* production of $^{14}C$ from

$$A_{is} = P(1-e^{-\lambda T}) \text{ (dpm g}^{-1} \text{ carbon)} \tag{1}$$

where P is the production rate of $^{14}C$

$$P = \frac{60\phi\sigma_N N f_N}{f_C M} \text{ ($^{14}$C atoms min}^{-1} \text{ g}^{-1} \text{ carbon)} \tag{2}$$

$\lambda$ is the $^{14}C$ decay constant, T is the time of exposure to neutron irradiation, $60\phi$ is the neutron flux in neutrons per minute per $cm^2$, $\sigma_N$ is the effective cross section of nitrogen for thermal neutrons, N is Avogadro's number, $f_N$ and $f_C$ are the relative nitrogen and carbon content of the sample expressed as weight fraction, and M is the atomic weight. If not only the reaction $^{14}N(n,p)^{14}C$ is considered, but also the combined production rate of all possible reactions, P has to be multiplied by a factor, R~1.1. The neutron flux at the position of the sample was calculated as the sum of the cosmic neutron flux and that due to uranium fission. Table 2 lists parameters used in the calculation. For the calculation of the neutron flux from $^{238}U$ fission, I assumed that the attenuation length of fission neutrons underground is comparable to that for cosmic-ray neutrons in water. Because the neutron capture cross-section for hydrogen is of the same order as for several of the

TABLE 1 . Description of Samples and Estimated *In-situ* Production of $^{14}C$

| Sample | Anthracite | Amersfoort III |
|---|---|---|
| Depth below surface | | 8.40 m |
| Thickness of layer | | 0.35 m |
| Sample composition | | |
| Total dry organic material* | 97% | 86.5% |
| Carbon content | 88.5% | 54.2% |
| Hydrogen content | 3.8% | 5.6% |
| Nitrogen content | 0.83% | 1.1% |
| Uranium concentration, ash** | 26 ± 5 ppm | 18 ± 5 ppm |
| Neutron flux $\phi$ ($ncm^{-2}\ s^{-1}$) | | |
| In thick sample layer† | $1.9 \times 10^{-5}$ | $1.6 \times 10^{-5}$ |
| In embedded layer‡ | $6.3 \times 10^{-4}$ | $4.43 \times 10^{-4}$ |
| Due to cosmic rays | | $6.6 \times 10^{-5}$ |
| Produced activity (dpm $g^{-1}$ C) | | |
| Thick layer | $8.3 \times 10^{-7}$ | $7.8 \times 10^{-6}$ |
| Embedded layer | $2.8 \times 10^{-5}$ | $4.7 \times 10^{-5}$ |
| Apparent age (yr BP)§ | | |
| Thick layer | $1.34 \times 10^{5}$ | $1.15 \times 10^{5}$ |
| Embedded layer | $1.05 \times 10^{5}$ | $1.01 \times 10^{5}$ |

*Analyses carried out by J. Ebels of the Analytical Department, Chemical Laboratory, University of Groningen; C, H, and N content based on total sample. The water content of the samples can be estimated based on the fact that moist peat contains about three times more water than dry organic matter.

**X-ray fluorescence spectrometry analysis by E. A. Th. Verdurmen of Zuiver Wetenschappelijk Onderzoek Laboratory for Isotope Geology, Amsterdam.

†"Thick" means several attenuation lengths (170 g $cm^{-2}$) *i.e.*, several meters; the neutron production over one attenuation length is assumed to contribute to the flux.

‡Composition of the surrounding layers assumed to be equal to that of the ash

§Calculated using a recent activity of 13.56 dpm $g^{-1}$ C (Karlén *et al.* 1964).

TABLE 2. Neutron flux parameters

| | | Reference* |
|---|---|---|
| Neutron flux $\phi$ at air-land interface 0.05–2.0 MeV** | $2.3 \times 10^{-3}$ $ncm^{-2}\ s^{-1}$ | 1 |
| Relative $\mu$ meson contribution to $\phi$ | 4% | 2 |
| Decay constant for spontaneous fission of $^{238}U$, $\lambda_{sf}^{238}$ | $2.7 \times 10^{-24}\ s^{-1}$ | 3 |
| Decay constant for spontaneous fission of $^{235}U$, $\lambda_{sf}^{235}$ | $(1.1 \pm 0.7) \times 10^{-25}\ s^{-1}$ | 3 |
| Average number of neutrons from $^{238}U$ fission | 2.2 ± 0.3 | 3 |
| Attenuation length of neutrons in air | ~120 g $cm^{-2}$ | 4 |
| Attenuation length of neutrons in water | 169 +19/−16 g $cm^{-2}$ | 5 |
| Attenuation length of nucleons in water | ~160 g $cm^{-2}$ | 2 |
| Attenuation length of $\mu$ mesons in water | ~4000 g $cm^{-2}$ | 2 |
| Non-cosmic neutron flux in rock | $(0.14–1.44) \times 10^{-3}$ $ncm^{-2}\ s^{-1}$ | 6 |
| Thermal neutron capture cross section $\sigma_H$ | $(332 \pm 2) \times 10^{-27}$ $cm^2$ | 7 |
| $\sigma_N$ | $(1.8 \pm 0.1) \times 10^{-24}$ $cm^2$ | 7 |

*1. Gold (1968); 2. Cocconi & Cocconi Tongiorgi (1951) and estimate; 3. Segré (1952); 4. Harkness & Burleigh (1974); 5. Bagge & Skorka (1958); 6. Sawelski (1968); 7. Goldman *et al.* (1972).

**Measured at 200 m altitude at 42°N, 88°W; geomagnetic latitude 53°N.

more important rock elements, this assumption seems reasonable. For muons and nucleons, I also assumed that the attenuation length underground is approximately equal to that in water.

I calculated the *in-situ* production (T~infinite) for two cases, one in which the sample came from a thick layer of the same composition, and the other in which the sample was embedded as a thin layer in sediments with a composition equal to that of the sample ash. The assumption that the uranium content of the overlying and underlying inorganic layers is equal to that of the ash of the organic material provides an upper limit for the fission-produced flux, since uranium tends to be enriched in organic material relative to its inorganic surroundings (Felber & Hernegger 1971). The neutron fluxes calculated for thin sample layers agree well with measured neutron fluxes in boreholes (Sawelski 1968). The apparent ages in Table 1 clearly show that *in-situ* $^{14}C$ production in these materials has a negligible influence for samples up to 80 ka. This is even more so because probably a fraction of the $^{14}C$ produced is removed during sample pretreatment (*c.f.*, Harkness & Burleigh 1974). Thus, it seems likely that the general difficulty that accelerator mass spectrometry (AMS) laboratories have with obtaining background samples without $^{14}C$ results from contamination of the very small samples used with $^{14}C$ during sample preparation, rather than from *in-situ* production of $^{14}C$. Improved sample preparation should hold the key to lower backgrounds and a higher AMS $^{14}C$ age range.

## CONCLUSIONS

Subtle variations in $^{14}C$ concentrations related to latitude, season and solar modulation may be revealed by high-precision measurements, and can provide important information on atmospheric mixing and the dynamics of the global carbon cycle when interpreted with dynamic 3-D atmospheric models. The magnitude of these cycles is of the order of 3–5‰. *In-situ* production of $^{14}C$ in carbon-rich samples is still below the current detection limit, and background problems encountered in AMS measurements must be attributed to contamination.

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