HIGH-RESOLUTION AMS ^{14}C DATING OF POST-BOMB PEAT ARCHIVES OF ATMOSPHERIC POLLUTANTS

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ABSTRACT. Peat deposits in Greenland and Denmark were investigated to show that high-resolution dating of these archives of atmospheric deposition can be provided for the last 50 years by radiocarbon dating using the atmospheric bomb pulse. ^{14}C was determined in macrofossils from sequential one cm slices using accelerator mass spectrometry (AMS). Values were calibrated with a general-purpose curve derived from annually averaged atmospheric $^{14}CO_2$ values in the northernmost northern hemisphere (NNH, 30°–90°N). We present a thorough review of ^{14}C bomb-pulse data from the NNH including our own measurements made in tree rings and seeds from Arizona as well as other previously published data. We show that our general-purpose calibration curve is valid for the whole NNH producing accurate dates within 1–2 years. In consequence, ^{14}C AMS can precisely date individual points in recent peat deposits within the range of the bomb-pulse (from the mid-1950s on). Comparing the ^{14}C AMS results with the customary dating method for recent peat profiles by ^{210}Pb, we show that the use of ^{137}Cs to validate and correct ^{210}Pb dates proves to be more problematic than previously supposed.

As a unique example of our technique, we show how this chronometer can be applied to identify temporal changes in Hg concentrations from Danish and Greenland peat cores.

INTRODUCTION

Recent scientific work has demonstrated the feasibility of using peat sediments as a global atmospheric archive for heavy metal and organic contaminants. Thus, peat has been shown to be a reliable archive of atmospheric Pb (Shotyk et al. 1998), and there is evidence that Hg is also effectively immobile in peat, though the question of how faithful an archive peat is for the volatile element Hg is still under investigation (see Benoit et al. 1998). Peat has also yielded a long-term climatic record (Cortizas et al. 1999) and has provided a high-resolution record of atmospheric CO_2 content (White et al. 1994). A high-resolution time series during the last 50 years is urgently needed for pollutants such as Hg to evaluate the effects of emission controls, and to help calibrate atmospheric transport models. Such time series are especially needed from the Arctic, as the most significant gap at the present time in Arctic contaminant research is the "lack of temporal trend information for most contaminants" (Braune et al. 1999).

Although there have been studies where the ^{14}C from the atmospheric bomb pulse has been used to date the top layers of a peat profile (see Gedyé 1998; Arslanov et al. 1999), typically in peat studies the upper layers are dated with radiometric methods, customarily ^{210}Pb (see Appleby et al. 1997).

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In the present study we investigate the feasibility of using the bomb-pulse ^{14}C content to date peat cores from Denmark and Greenland for the period of 1950 to the present. For comparison, the cores were also dated using the customary ^{210}Pb method.

It is the first time that peat from Greenland is used in a high-resolution contaminant study (Goodsite 2000). Peat provided the opportunity to obtain a relatively inexpensive terrestrial archive from the Arctic. The use of accelerator mass spectrometry (AMS) allows dating of single year growth increments in individual plant macrofossils. We have used the dating results to compare the concentration profiles of Hg in Denmark and Greenland to the published North American Hg emission records (Pirrone et al. 1998).

The details of the concentration profiles of Hg and other metal contaminants in the peat cores have been treated elsewhere (Goodsite 2000; Shotyk et al. 2001).

METHODS

Two distinctly geochemically and trophically different peat lands, one in Denmark and one in Greenland, were selected for study. In Denmark, we selected the raised bog at Storelung, Staaby, Funen, Denmark (55°15.5′N, 10°15.5′E)**.** This is an ombrotrophic bog, nourished by the atmosphere since it is raised above the water table. Such bogs are well established as archives of atmospheric deposition. Three cores of predominantly *Sphagnum* peat were sampled in October 1999 and processed.

To the best of our knowledge, no one has located an ombrotrophic peat bog in Greenland. Therefore, it was decided to find and investigate a suitable minerotrophic (groundwater nourished) fen. Small mires (a generic term for unclassified peat lands) were located and sampled in September 1999 on the Narsaq peninsula, southern Greenland (Tasiusaq, Narsaq: 61°08.3′N, 45°33.7′W) (Goodsite 2000). The mires had *Carex* peat accumulation ranging from 20 cm to approximately 100 cm deep. Since they received at least some of their water supply and nutrients from the mineral groundwater table surrounding their landscape, they are classified as fens.

One-meter long cores (monoliths) of peat spanning approximately three thousand years of deposition were taken from each location. As the peat deposits were similarly sampled, and the cores were processed in the same way, only the Greenland cores will be described in some detail. Three (15 cm × 15 cm by approximately 100 cm) replicate monoliths of peat from each of two sites in Greenland (only one site in Denmark) were cored using a Ti Wardenaar peat sampler (Wardenaar 1987). At each site the three replicate cores were taken at a distance of approximately 1.5 m from each other. Further analysis was carried out on cores from only one site, with cores from the other site being frozen and stored. The choice of Greenland site to be analyzed immediately was based on pH profiles of the peat pore water measured in the field. The site chosen had a higher acidity in the upper 20 cm than the other site, which was near neutral pH throughout. Since pH is a typical indicator of trophic status, with ombrotrophic bogs typically having pH of 3 to 4, it was hoped that this upper region might prove to be ombrotrophic, though later analyses showed it was not.

The three cores (labelled A, B, C) from each site were frozen soon thereafter and shipped to the Trace Metals Lab, Geological Institute, University of Berne for further processing and analysis. The zero point on the depth scale is defined by visual inspection as the point where it appears that the living (green material) stops. Hg and metals analyses are as described in Shotyk et al. (2001).

Core A was sliced into 3 cm slices by hand using a stainless steel knife prior to freezing. Pore water was manually squeezed out of the slices, filtered and then stored cool. Portions of the slices were

dried overnight at 105 °C in a drying oven and milled in a Ti mill. The milled powder from pieces of each centimeter slice was then manually homogenized prior to using the powder for further analysis. Lead and 19 other elements were then determined using X-ray fluorescence spectrometry (XRFS) at EMMA Analytical, Canada, by Dr Andriy Cheburkin (see Shotyk et al. 2001).

Core B was cut while frozen into 1 cm slices using a stainless steel band saw, and selected portions of the slices were then dried and milled as above. Samples were then analyzed as before using XRFS. Powders were selected for stable lead isotope analysis using thermal ionization mass spectrometry, based on the Pb concentration profile obtained using XRFS. Plant macrofossils were removed from the centers of the slices from Cores B at the Institute of Plant Science, University of Berne, where they were cleaned and dried at 60 °C. Within one week from selection they were processed at the AMS ^{14}C Dating Laboratory, University of Aarhus, for ^{14}C dating with AMS using a standard procedure for plant material (washed, acid-base-acid treatment). AMS was run on the samples to reproduce the atmospheric bomb pulse curve and to date peaks in the profile. We would like to stress that we use well-defined and carefully selected macrofossils for our study, so all the well-known effects of getting too old or highly varying ages from dating peat water, bulk material or humic acid, humin and fulvic acid fractions (see Olsson 1986; Shore et al. 1995) do not apply.

For the construction of a terrestrial bomb-pulse calibration curve, we used two different materials from southern Arizona (USA), Douglas fir and cottonseeds, measured at the NSF-Arizona AMS Facility. A cross-section of Douglas fir was cut, smoothed with sandpaper and individual rings were then sampled. The cottonseeds were harvested in the year they were produced and archived for later use. Both sample types received the following acid-base-acid pretreatment: They were soaked in 3N HCl overnight to remove inorganic carbon, afterwards they were rinsed to neutral pH with ASTM Type I water, soaked in 2% NaOH overnight to remove mobile carbon (i.e. humic or fulvic acids), rinsed to neutral pH with Type I water, soaked in 3N HCl to neutralize any remaining NaOH, and finally rinsed to neutral pH with Type I water.

Conversion of all the ^{14}C ages to calendar ages was performed via a Bayesian calibration program (Puchegger et al. 2000) using cubic spline interpolation for the calibration curve. Our calibration curve (see solid line in Figure 1) was constructed as follows: For the period before 1956 we used the tree-ring data from the INTCAL98 ^{14}C calibration curve (Stuiver et al. 1998). For the period from 1956 till now we used annually averaged atmospheric $^{14}CO_2$ data for the latitude band 30°–90° N provided by I Levin (personal communication): for the period of 1955 to 1959 data compiled by Tans (1981), from 1959 to 1984 data from Vermunt from Levin et al. (1985); after 1988 the arithmetic mean values from the three northern hemispheric stations Izaña (1985–1997), Jungfraujoch (1987–1997) and Alert (1989–1997) were taken. Values for 1998 and 1999 were obtained by extrapolating the almost exactly exponential global decrease in $^{14}CO_2$ since 1982 (Levin and Hesshaimer 2000).

Core B was also dated with ^{210}Pb and ^{137}Cs at the Liverpool University Environmental Radiometric Laboratory. Dried and ground samples from the profile were analyzed for ^{210}Pb, ^{226}Ra, ^{137}Cs, and ^{241}Am by direct gamma assay using Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detectors (Appleby et al. 1986). Corrections were made for the effect of self-absorption of low-energy gamma ray within the sample (Appleby et al. 1992). ^{210}Pb dates were calculated using the CRS (constant rate of supply) dating model (Appleby and Oldfield 1978) and corrected to agree with the ^{137}Cs signal using the methodology described in Appleby (1998).

Core C remains frozen as an archive at the Trace Metals Lab, Geological Institute, University of Berne.

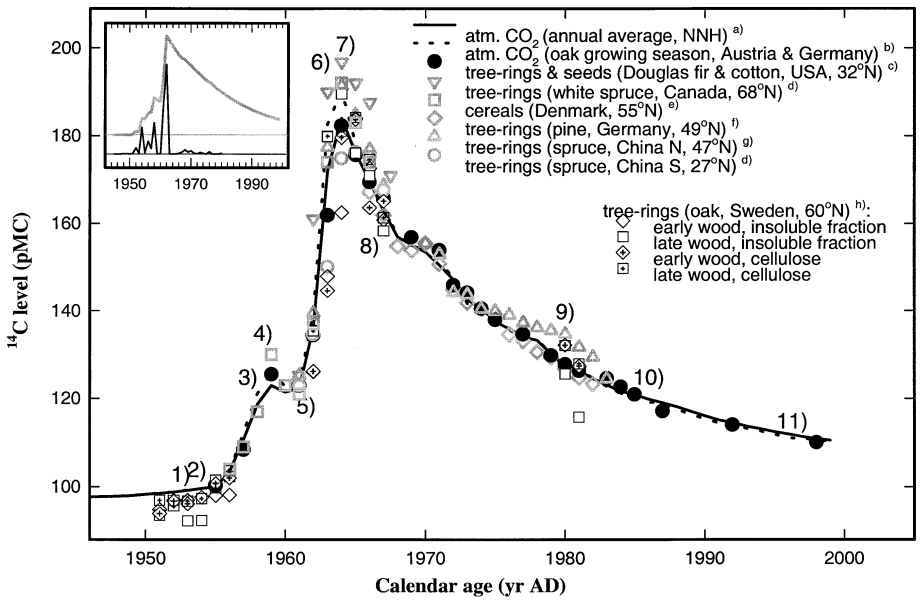

Figure 1 The bomb-pulse curve. a) The annually averaged atmospheric $^{14}CO_2$ curve for the 30°–90°N latitude band provided by I Levin (personal communication). b) Atmospheric $^{14}CO_2$ data from Vermunt, Austria (47°N) and Schauinsland, Germany (48°N) (Levin et al. 1997) but only averaged for April to August, the growing season of oaks (Rom 2000). c) See Table 1. d) Trees from the Mackenzie Delta, Canada, and Mingyin, Yunnan Province, China (Dai and Fan 1986). e) Mainly ears (but also grains or green parts) of barley, wheat, rye and oats from the Copenhagen area (Tauber 1967). Note that the (annually averaged) values given are only approximate values since the original data had been published as D using a physical ^{14}C half-life of 5570 yr and 1958 as the reference year. Regarding the rather high value for 1959 Tauber (1967) mentions that in that year the polar front had an extreme northern position at European latitudes during the whole summer. f) Tree from Obrigheim, Germany (Levin et al. 1985). g) Tree from Dailing, Heilongjiang Province, China (Dai et al. 1992). h) Different sections and different chemical fractions in tree-rings from an oak from Uppsala, Sweden (Olsson and Possnert 1992).

Inset: Global input by the atmospheric nuclear weapons' tests measured as TNT energy equivalent (thin black line) and cumulative input using an exponential decay time of 18.70 ± 0.15 yr corresponding to the uptake by the biosphere and the oceans (see Levin and Hesshaimer 2000). Values are given in arbitrary units.

The "History" of the Bomb-Pulse (see Bennett et al. 2000; USNT 2000), a report on the US bomb tests published by the US Department of Energy, in a few cases states slightly different dates). 1) May 1952: The first major thermonuclear device is successfully tested by the USA (Eniwetok Atoll). Aug 1953: The first major thermonuclear device is successfully tested by the USSR (Semipalatinsk). 2) Feb. 1954: The USA test their first thermonuclear device at the Bikini Atoll, the highest-yield test site of the USA (yield maximum in 1954/6/8). Further relevant test sites with high yields for the USA: 1952 Eniwetok Atoll, 1958/62 Johnston Island, 1962 Christmas Islands. 3) Sept. 1957: The USSR test their first thermonuclear device at the Novaya Zemlya, the highest-yield test site of the USSR (yield maxima in 1958 and 1961/2). 4) Oct/Nov 1958: Due to the "Geneva convention of Experts for the Discontinuance of Nuclear Weapons" the USA and the USSR stop their atmospheric tests. There is a general moratorium till France starts its atmospheric tests in Algeria in Feb 1960. 5) 1961–62: The USSR and the USA resume testing in September 1961 and April 1962, respectively. 6) Nov/Dec 1962 Due to the "Partial Test Ban Treaty" the USA and the USSR stop their atmospheric tests forever. Only underground tests are allowed, and only if no radioactive debris is spread beyond territorial limits. 7) Oct 1964: China starts its atmospheric testing series at Lob Nor (yield maxima in 1967–70 and 1973/6). 8) Jul 1966: France starts its atmospheric testing series at Muroroa and Fangataufa (yield maxima in 1968 and 1970–72, stopped in Sep 1974). 9) Oct 1980: China performs the last atmospheric test ever. 10) April 1986: The nuclear power plant at Chernobyl, USSR explodes. 11) Sept 1996: The Comprehensive Test Ban Treaty is opened for signature, but so far (Nov 2000) has not been ratified by the USA or India.

The decline of the bomb ^{14}C after stopping the atmospheric nuclear weapons tests is mainly driven by uptake into the oceans and the biosphere. In addition, emissions of fossil fuel CO_2, emissions of ^{14}C by nuclear power plants, and possibly nuclear underground tests contribute (Levin and Hesshaimer 2000).

RESULTS AND DISCUSSION

The Bomb-Pulse Curve for the Northern Hemisphere

General Considerations

Figure 1 shows several ^{14}C records covering the bomb-pulse period, which were obtained from either atmospheric $^{14}CO_2$ measurements or from tree-rings and seeds in the northernmost part of the northern hemisphere (NNH), i.e. the region north of about 30°N excluding the tropical (Hadley) convection cell. Now a crucial question arises in connection with any ^{14}C calibration curve: is it only of local validity or does it apply to the entire NNH. In the following we address this question and demonstrate that it is possible to construct a calibration curve, which is valid for the whole NNH producing accurate dates within 1–2 years.

Nydal and Lövseth (1983) investigated atmospheric $^{14}CO_2$ concentration patterns for the period 1962–1980 showing that these patterns are basically the same for the southern tip of Norway (58°N) and Spitsbergen (78°N), and also $^{14}CO_2$ concentrations on the Canary Islands (27–28°N) show similar results. Similarly, for the period 1980–1992 Nydal and Gislefoss (1996) find no significant deviation between the Nordkapp (71°N) and the Canary Islands. From these data there is no evidence for a significant latitude dependence of atmospheric $^{14}CO_2$ concentrations for regions ranging from subtropical to subpolar/polar. However, a decrease of several percent in atmospheric $^{14}CO_2$ concentrations (around the bomb-pulse maximum) when going to northern hemisphere tropical regions (9–15°N) is clearly documented in Nydal and Lövseth (1983). The same effect is also reflected in trees from the tropics (see Murphy et al. 1997).

Dai and Fan (1986) and Dai et al. (1992) compared ^{14}C concentrations in tree-rings from spruce trees grown at different longitudes and latitudes in the northern hemisphere. They see good agreement for tree-rings grown at the same latitude, which reflects the rapid zonal mixing of the troposphere within about 1 month (e.g. Ehhalt 1999). On the other hand, these authors claim a clearly visible latitude dependence of ^{14}C in trees grown in 1961–1967 at 27°N, 47°N, and 68°N (see Figure 1). This may be explained by the fact that meridional atmospheric mixing takes several months (Ehhalt 1999), and injection of air from the stratosphere, enriched in $^{14}CO_2$ compared to the troposphere, is not equally distributed in time and latitude but mainly takes place during spring/early summer at mid-latitudes (Levin et al. 1985; Dai and Fan 1986). Subsequently, the injected $^{14}CO_2$ "diffuses" north and south. Since trees also mainly grow in the spring/summer season, this together with the sufficiently slow meridional mixing (compared to zonal mixing) may lead to a significantly higher ^{14}C concentration taken up into tree-rings relative to the annual atmospheric average, and also may generate a latitudinal gradient in atmospheric $^{14}CO_2$ concentrations. However, Olsson and Possnert (1992) regard some of the values measured by Dai and Fan (1986) in a white spruce from Canada (68°N) (see Figure 1) as unexpectedly high compared to atmospheric measurements from Spitsbergen (78°N) and Abisko, Sweden (68°N), and they point out that finer details of the sample pretreatment are missing (see also discussion below). They therefore advise to treat these results with caution.

For the rising part of the bomb pulse Tauber (1967) compared numerous ^{14}C data in both atmospheric CO_2 and plants at mid-latitude regions all over the world and found no clear latitude dependence. He supposed that the amplitude and extent of a possible latitude effect may depend on annually varying meteorological factors (see also caption of Figure 1). Grass samples taken within one week in June 1963, the year with the highest input of bomb-produced ^{14}C, also did not reveal any significant latitudinal gradient all over Scandinavia (56–70°N). However, for the same month he

observed a clear difference in grass samples from Greenland relative to Scandinavia of more than 14% in ^{14}C level, corresponding to a delay of somewhat more than one month. Furthermore, a clear gradient with high values in the south was observed for Greenland. This general difference between Scandinavia and Greenland has been explained by the location of the polar front: In summer times the polar front is over southern Scandinavia with frequent shift toward north and south, but over the Atlantic it is generally far south of Greenland. Therefore stratospheric $^{14}CO_2$ injected at mid-latitudes and diffusing toward north and south is distributed differently.

Note that the production of ^{14}C by the atmospheric nuclear weapons tests was maximum in 1962 (see Bennett et al. 2000), whereas the maximum ^{14}C concentration in tropospheric measurements show up in August/September 1963 and in June 1964 for the extratropical and the tropical NH, respectively (see Nydal and Lövseth 1983, and Nydal and Lövseth 1996 with slightly corrected and also updated data; see also inset in Figure 1). This clearly shows a) the (seasonally dependent) injection of $^{14}CO_2$ from the stratosphere, and b) a delay due to the different transport patterns between tropics (Hadley cells) and extratropics (Ferrel and polar cells).

Several decades after the bomb-pulse maximum Olsson (1989) finds a still slightly lower ^{14}C level in plants from subarctic and arctic areas (including Iceland, the Faroe Islands, Greenland or Spitsbergen) compared to Sweden (e.g. about 2% in Spitsbergen for 1980). Also atmospheric $^{14}CO_2$ measurements in Abisko, Sweden (68°N) and Kapp Linné, Spitsbergen (78°N) during the 1980s show consistently lower values for Spitsbergen, however in the range of less than 1.5% (Olsson 1993).

Another factor influencing the ^{14}C level is the dilution with fossil fuel derived CO_2: For the above-mentioned oak tree from Sweden studied in Olsson and Possnert (1992) (see Figure 1) a local industrial effect in the range of 2% was claimed. Also for the pine tree from Germany studied in I. Levin et al. (1985) (see Figure 1) a ^{14}C depression of 1.5% by fossil fuel contamination has been ascribed, although in this case not as a local but as a general ground level effect. Olsson (1989) states, that clean air does not exist any longer, and therefore e.g. "clean air" ^{14}C values from Germany, which were generally lower than in Sweden (about 2–3% for the late 1970s/early 1980s), should reflect a higher fossil fuel contamination. However, this difference is gone during the 1980s.

For the present-day northern hemisphere the maximum of fossil fuel CO_2 emissions at mid-latitudes leads to a corresponding minimum in atmospheric ^{14}C activity. E.g. measurements at the Alpine station Jungfraujoch, Switzerland (47°N) from 1993–4 show ^{14}C values, which are about 1‰ and 1.5‰ lower than values obtained at the remote stations Alert, Nunavut, Canada (82°N) and Izaña, Canary Islands (28°N) (Levin and Hesshaimer 2000). Values in the tropics (Llano del Hato, Venezuela, 8°N) are even higher (2‰), which may be due to reemission of bomb-pulse ^{14}C from the highly active biosphere, which has a carbon turnover time of about 30 years (Levin and Hesshaimer 2000).

To study the influence of the sample selection and pretreatment regarding tree-rings Olsson and Possnert (1992) investigated different sections (early wood vs. late wood) and different chemical fractions in tree-rings from an oak tree from Uppsala, Sweden (60°N). After removal of the soluble fraction, which corresponds roughly to applying the commonly used acid-base-acid method, they find no clear evidence for a delay between the atmospheric and tree-ring ^{14}C values on a timescale of weeks. For the years 1962–63, i.e. the steepest rise in the bomb pulse, the difference between early-wood and late-wood values is evident (see Figure 1). Olsson and Possnert (1992) do not ascribe this to the temporally dependent diffusion of injected stratospheric $^{14}CO_2$ but to nutrients from the respective preceding year, which are stored in the roots and are used for the formation of the early wood of the following year.

Figure 1 summarizes a large part of the data discussed above. When comparing the curve of ^{14}C concentrations in plants and atmospheric $^{14}CO_2$ averaged over the growing season of oaks (dashed line) with the curve of the respective annually averaged atmospheric concentrations (solid line), the two curves coincide except for a few years around the bomb-pulse maximum. This difference can be ascribed to the great seasonal oscillations caused by stratospheric injection of excess (bomb) ^{14}C leading to regional differences within the extratropical northern hemisphere (NNH). However, these differences are insignificant after about 1971 (Nydal and Lövseth 1983) (after about 1978 according to Olsson 1993), where basically equilibrium between the stratosphere and the troposphere has been reached. (The deviation of the spruce tree from China from atmospheric concentrations between 1976 and 1982 shown in Figure 1 clearly reflects the Chinese atmospheric atomic bomb tests during that time (Dai et al. 1992).)

Finally we want to emphasize that—even including all the effects discussed above (except for the period 1976–82 in China as discussed above)—any of the bomb-pulse records shown in Figure 1 when used for calibration from the mid 1950s on will generally give the same calibrated age within 1–2 years.

A Complete Tree-Ring/Seed Bomb-Pulse Record and an Atmospheric Bomb-Pulse Calibration Curve for the Extratropical Northern Hemisphere

None of the so far published bomb-pulse records from tree-rings shown in Figure 1 covers the whole second half of the 20th century. We present a first (although not annual) record from plant material covering the whole bomb-peak period till now. The ^{14}C data were obtained from Douglas fir tree-rings and cottonseeds from southern Arizona (32°N), which are summarized in Table 1 and shown in Figure 1. As can be seen from Figure 1, the values closely follow the annually averaged atmospheric $^{14}CO_2$ concentration curve for the NNH (solid line) provided by I Levin (personal communication). It is interesting to note that the data from Arizona do not show significantly higher ^{14}C concentration values for the period around the bomb-pulse maximum (see also the previous section), i.e. they do not follow the seasonal variations as expected for plants compared to annually averaged atmospheric values. For comparison, we constructed a calibration curve using atmospheric $^{14}CO_2$ data from Vermunt, Austria and Schauinsland, Germany (Levin et al. 1997) but only averaged for April to August, the growing season of oak trees (see Wrobel and Eckstein 1992), to simulate the relevant annual portion of $^{14}CO_2$ that is taken up by the plant. Such an "atmospheric" calibration curve (Rom 2000, see dashed line in Figure 1) clearly shows the features expected from the stratospheric injection of excess ^{14}C in spring, which is surprisingly well matched by the cereals data from Denmark (Tauber 1967; see Figure 1). From this, the Arizona data, when compared to the annually averaged atmospheric $^{14}CO_2$ data, may either point towards a latitudinal dependence (see preceding section) or rather seem to be unaffected by the seasonal variations due to the injection of stratospheric $^{14}CO_2$. We checked the gap between 1992 and 1998 in our tree-ring/seed record and measured green leaves of an (unspecified) tree collected at the Brorfelde Observatory, Zealand, Denmark (56°N) resulting in (110.5 ± 0.5) pMC (AAR-3339). Similar to the extrapolation of the atmospheric calibration curve by I. Levin (as described in the Methods section) we interpolated the 1996 value for our Arizona data using an exponential fit to the data points from 1983–1998. The corresponding error was obtained by averaging the variances for the given period. The interpolated value of (110.8 ± 0.6) pMC perfectly matches the measured value.

We finally decided to use the annually averaged atmospheric $^{14}CO_2$ curve for the 30–90°N latitude band provided by I Levin (personal communication) (solid line in Figure 1) as our general purpose bomb-pulse calibration curve since a) the Arizona data closely follow this curve but do not provide

Table 1 ^{14}C determination in plants from Arizona, USA to establish a terrestrial bomb-pulse calibration curve[a]

Lab nr	Sample species[b]	Specification	Sample yr	Sample location	$\delta^{13}C$ (‰)	pMC[c]
AA6665	Douglas Fir	Tree ring	1955	Santa Catalina Mountains, Arizona, USA	–25.0[d]	100.2 ± 0.6
AA6667	Douglas Fir	Tree ring	1957	Santa Catalina Mountains, Arizona, USA	–25.0[d]	108.4 ± 0.6
AA6668	Douglas Fir	Tree ring	1959	Santa Catalina Mountains, Arizona, USA	–25.0[d]	125.5 ± 0.7
AA6670	Douglas Fir	Tree ring	1961	Santa Catalina Mountains, Arizona, USA	–25.0[d]	122.9 ± 0.7
AA6671	Douglas Fir	Tree ring	1962	Santa Catalina Mountains, Arizona, USA	–25.0[d]	134.4 ± 1.2
AA6672	Douglas Fir	Tree ring	1963	Santa Catalina Mountains, Arizona, USA	–25.0[d]	161.8 ± 0.9
AA6673	Douglas Fir	Tree ring	1964	Santa Catalina Mountains, Arizona, USA	–25.0[d]	182.3 ± 1.1
AA6674	Douglas Fir	Tree ring	1965	Santa Catalina Mountains, Arizona, USA	–25.0[d]	175.6 ± 0.9
AA6675	Douglas Fir	Tree ring	1966	Santa Catalina Mountains, Arizona, USA	–25.0[d]	169.4 ± 1.0
AA6676	Douglas Fir	Tree ring	1967	Santa Catalina Mountains, Arizona, USA	–25.0[d]	165.7 ± 0.9
AA6678	Douglas Fir	Tree ring	1969	Santa Catalina Mountains, Arizona, USA	–25.0[d]	156.8 ± 0.9
AA6680	Douglas Fir	Tree ring	1971	Santa Catalina Mountains, Arizona, USA	–25.0[d]	153.8 ± 0.9
AA11846	Cotton	Seed	1972	Southern Arizona, USA	–27.0	145.8 ± 0.6
AA11847	Cotton	Seed	1973	Southern Arizona, USA	–26.4	143.9 ± 0.4
AA6682	Douglas Fir	Tree ring	1973	Santa Catalina Mountains, Arizona, USA	–25.0[d]	145.0 ± 0.8
			1973 (weighted average)			144.1 ± 0.4
AA11848	Cotton	Seed	1974	Southern Arizona, USA	–26.0	140.9 ± 0.4
AA11849	Cotton	Seed	1974	Southern Arizona, USA	–29.3	139.5 ± 0.6
			1974 (weighted average)			140.5 ± 0.3
AA11850	Cotton	Seed	1975	Southern Arizona, USA	–27.1	137.6 ± 0.7
AA11851	Cotton	Seed	1975	Southern Arizona, USA	–27.8	137.8 ± 0.5
AA6684	Douglas Fir	Tree ring	1975	Santa Catalina Mountains, Arizona, USA	–25.0[d]	138.7 ± 0.8
			1975 (weighted average)			137.9 ± 0.4
AA6686	Douglas Fir	Tree ring	1977	Santa Catalina Mountains, Arizona, USA	–25.0[d]	134.7 ± 0.7
AA6688	Douglas Fir	Tree ring	1979	Santa Catalina Mountains, Arizona, USA	–25.0[d]	129.9 ± 0.7
AA11839	Cotton	Seed	1980	Southern Arizona, USA	–28.2	127.7 ± 0.7
AA11840	Cotton	Seed	1981	Southern Arizona, USA	–28.7	126.1 ± 0.4
AA11841	Cotton	Seed	1981	Southern Arizona, USA	–27.3	126.3 ± 0.6
AA6690	Douglas Fir	Tree ring	1981	Santa Catalina Mountains, Arizona, USA	–25.0[d]	127.1 ± 0.7

Table 1 ^{14}C determination in plants from Arizona, USA to establish a terrestrial bomb-pulse calibration curve[a] (*Continued*)

Lab nr	Sample species[b]	Specification	Sample yr	Sample location	$\delta^{13}C$ (‰)	pMC[c]
		1981 (weighted average)				126.3 ± 0.3
AA6692	Douglas Fir	Tree ring	1983	Santa Catalina Mountains, Arizona, USA	–25.0[d]	124.5 ± 0.7
AA11843	Cotton	Seed	1984	Southern Arizona, USA	–26.9	122.6 ± 0.4
AA11844	Cotton	Seed	1985	Southern Arizona, USA	–26.1	120.2 ± 0.4
AA6694	Douglas Fir	Tree ring	1985	Santa Catalina Mountains, Arizona, USA	–25.0[d]	123.8 ± 0.8
		1985 (weighted average)				120.9 ± 0.4
AA6696	Douglas Fir	Tree ring	1987	Santa Catalina Mountains, Arizona, USA	–25.0[d]	119.6 ± 0.8
AA11845	Cotton	Seed	1992	Southern Arizona, USA	–27.2	114.0 ± 0.6
—[e]		Tree ring	1998	Arizona, USA		110.0 ± 0.7

[a]Whenever there is more than one sample in an individual year, first a weighted average for samples from the same sample material (Douglas fir or cottonseeds) was made. These sample-material averages were then combined in a weighted average to give the final result for the respective year. Since most calibration programs use conventional ^{14}C ages as input, for the convenience of reader we give the formula to convert pMC values into ^{14}C age *T*:

$$T = -8033 \cdot Ln\left(\frac{pMC}{100}\right)$$

All the bomb-pulse data will then result in negative ages. We know that there is some reluctance in the ^{14}C community to use negative ^{14}C ages, but we think they might be a suited extension of the pre-bomb calibration curve into the bomb and post-bomb period without changing units. Note that the conventional positive ^{14}C ages are also not "true" ages but only a quite arbitrary though unambiguous mathematical transformation of the ^{14}C activity of the sample, which is still kept for historical reasons. Furthermore using 1950 as a reference year is not based on physical/natural parameters but also just a convention. By using negative ^{14}C ages one would avoid to publish ^{14}C datings before 1950 in yrs BP, whereas they have to be given as pMC or ^{14}C afterwards.

[b]Douglas Fir *(Pseudotsuga menziesii);* Cotton (*Gossypium* species)

[c]pMC (percent Modern Carbon) = (^{14}C concentration of the sample)/(0.95 × Oxalic Acid I ^{14}C concentration) × 100. Both ^{14}C concentrations are $\delta^{13}C$ corrected and refer to the same year.

[d]A $\delta^{13}C$ of –25.0 was assumed for all Douglas Fir samples

[e]Private communication by D Donahue

annual resolution, b) the curve provides consistent, carefully checked data covering the whole bomb pulse till now, c) most of the data used for constructing this curve are widely used among the ^{14}C community (see Levin et al. 1997 for the $^{14}CO_2$ data from Vermunt, Austria), and d) differences to the other extratropical northern hemisphere curves shown in Figure 1 generally result only in calibrated age differences of 1–2 years as stated in the previous section.

For calibration no uncertainties were assumed for our NNH curve. If not otherwise stated, all the calibrated ^{14}C ages in the Tables and Figures of this paper are given as 95%-confidence intervals (often denoted as "2-σ intervals").

The Peat Cores from Greenland and Denmark

Figures 2 and 3 and Table 2 show our results on plant material from the two peat cores Storelungmose in Denmark and Tasiusaq in Greenland. Both ^{14}C and modelled ^{210}Pb results are given.

^{14}C Dating

Regarding the ^{14}C measurements in the Greenland core (see Figure 2 and Table 2), one can see two important features: first, dating of samples from the peat surface gives results that are consistent with the year of the sampling, and second, the sample with the maximum ^{14}C content (AAR-5626) of 179.1 ± 0.8 pMC shows rather good agreement with the maximum of the calibration curve of 184.0 pMC. The peak value in our data from the Greenland core corresponds also well to the $\Delta^{14}C$ value of (776 ± 8)‰, i.e. (178.2 ± 0.8) pMC, measured in a grass sample from Narsaq, the very same peninsula where our peat samples come from, in July 1963 (Tauber 1967). Since no significant natural enrichment process for ^{14}C is known, this agreement between the peak value in our Greenland core and the atmospheric record cannot be just coincidence but ensures that we have no significant dampening for the Greenland peat core.

This is in contrast to results of Jungner et al. (1995) who measured ^{14}C in peat hummocks, i.e. raised surfaces on the peat land as opposed to "hollows", from central and eastern Finland (61°N and 62°N, respectively) using AMS on well defined stems of *Sphagnum fuscum* moss. For the peat core from central Finland the maximum ^{14}C value in the cellulose fraction of a moss sample representing a single-year fraction shows a $\Delta^{14}C$ of (660 ± 13)‰, i.e. (166.4 ± 1.3) pMC, a moss sample from the eastern Finland peat core representing a 3–5 yr average yielded a value as low as (564 ± 11)‰, i.e. (156.8 ± 1.1) pMC. Jungner et al. (1995) inferred that this effect was due to CO_2 emitted from decaying layers below the surface (i.e. older layers). An alternative explanation might be that the samples showing maximum ^{14}C content do not represent the bomb-pulse maximum.

Regarding the ^{14}C measurements in the Denmark core (see Figure 2 and Table 2), one also can see two important features: First, dating of samples from the peat surface gives results that are consistent with the year of the sampling, which is similar to the Greenland core, and second, the sample with the maximum ^{14}C content (AAR-5614) of 152.7 ± 0.8 pMC clearly does not agree with the maximum of the calibration curve of 184.0 pMC. More samples between 2.5 and 8.5 cm depth have to be dated in the near future to find out whether we so far just missed the peak of the bomb-pulse or whether there is significant dampening in the Denmark core, which we cannot exclude so far.

Preliminary results of additional determinations are now available for Denmark and Greenland. A macrofossil from 9.5 cm from the Danish core (AAR-6860) has a pMC of 176.8 ± 0.7, which approaches the expected maximum and additional samples above and below the maximum support a well developed curve. Therefore we originally just missed the peak of the bomb-pulse and no significant dampening is seen in the Danish core.

Table 2a Sample specifications, ^{13}C values and ^{14}C AMS datings of carefully selected and processed macrofossils from the peat cores from the Storelungmose, Denmark, and Tasiusaq, Greenland—Macrofossils from the Storelung Mose, Ståby, Fyn, Denmark (55°15.384′N, 10°15.336′ E)

Lab nr (AAR–)	Sample species	Specification	Depth (cm)	$\delta^{13}C$ (‰)[a]	pMC[b]	Calibrated age ranges (yr AD) (95% confidence intervals)
5611	*Andromeda* sp.	Fresh leaves	0[c]	–27.5	111.36 ± 0.54	1957 1995–1999
5612	*Leucobryum* sp.	Branches and leaves	0.5	–26.1	111.31 ± 0.61	1957 1995–1999
5613	*Leucobryum* sp.	Branches and leaves	2.5	–26.8	115.84 ± 0.65	1958 1989–1992
5614	*Leucobryum* sp.	Branches and leaves	8.5	–24.3	152.68 ± 0.76	1963 1970–1971
6612	*Sphagnum* sp.	Leaves	10.5	–23.0	136.99 ± 0.74	1962 1975–1976
6613	*Sphagnum* sp.	Leaves	11.5	–24.0	123.68 ± 0.65	1959–1961 1982–1984
6614	*Sphagnum* sp.	Leaves	12.5	–23.6	127.09 ± 0.67	1962 1980–1981
6615	*Sphagnum* sp.	Leaves and stems	13.5	–24.3	109.74 ± 0.65	1957 1995–1999
5615	*Leucobryum* sp.	Branches and leaves	14.5	–24.7	120.19 ± 0.55	1958; 1960 1984–1987
5616	*Leucobryum* sp.	Branches and leaves	15.5	–25.7	120.58 ± 0.56	1958–1961 1984–1987
5617	*Cyperaceae*	Leaves	16.5	–27.0	100.12 ± 0.53	1693–1726; 1813–1850 1862–1918; 1951–1956
5618	*Sphagnum* sp.	Branches and leaves	18.5	–24.2	99.43 ± 0.49	1687–1729; 1810–1923 1950–1955
5619	*Sphagnum* sp.	Branches (?) and leaves	78.5	–24.5	68.39 ± 0.38	1425–1424 BC; 1412–1211 BC 1201–1192 BC; 1179–1163 BC 1141–1132 BC

[a] $\delta^{13}C$ values have been measured by Árny E Sveimsbjörnsdóttir, Science Institute, the University of Iceland

[b] See Table 1 for definition

[c] These samples were taken from slightly above ground and correspond to the Hg concentrations values at negative depths shown in Figure 4

Table 2b Sample specifications, ^{13}C values and ^{14}C AMS datings of carefully selected and processed macrofossils from the peat cores from the Storelungmose, Denmark, and Tasiusaq, Greenland—Macrofossils from Tasiusaq, Narssaq, Greenland (61°08.314′N, 45°33.703′E).

Lab nr (AAR–)	Sample species	Specification	Depth (cm)	$\delta^{13}C$ (‰)[a]	pMC[b]	Calibrated age ranges (yr AD) (95% confidence intervals)
5620	*Sphagnum* sp.	Branches and leaves	0[c]	–26.2	110.99 ± 0.57	1957 1996–1999
5621	*Sphagnum* sp.	Branches and leaves	0	–28.3	111.88 ± 0.62	1957 1994–1999
5622	*Sphagnum* sp.	Branches and leaves	0.5	–26.8	114.34 ± 0.57	1957–1958 1991–1994
5623	*Sphagnum* sp.	Branches and leaves	5.5	–26.3	140.33 ± 0.61	1962 1973–1974
5624	*Sphagnum* sp.	Branches and leaves	7.5	–25.8	143.13 ± 0.69	1962 1973
5625	*Sphagnum* sp.	Branches and leaves	9.5	–25.9	162.32 ± 0.72	1963 1967
5626	*Sphagnum* sp.	Branches and leaves	12.5	–27.6	179.13 ± 0.83	1963–1965
5627	*Sphagnum* sp.	Branches and leaves	14.5	–26.7	126.62 ± 0.59	1961–1962 1980–1981
5628	*Sphagnum* sp.	Branches and leaves	16.5	–27.2	121.07 ± 0.54	1958–1961
5629	*Dwarf bush*	Twigs	18.5	–27.1	102.52 ± 0.54	1956
5630	*Vascular plant*	—	87.5	–26.7	69.52 ± 0.44	1292–1280 BC 1263–973 BC 959-941 BC

[a] $\delta^{13}C$ values have been measured by Árny E Sveimsbjörnsdóttir, Science Institute, the University of Iceland

[b] See Table 1 for definition

[c] These samples were taken from slightly above ground and correspond to the Hg concentrations values at negative depths shown in Figure 4

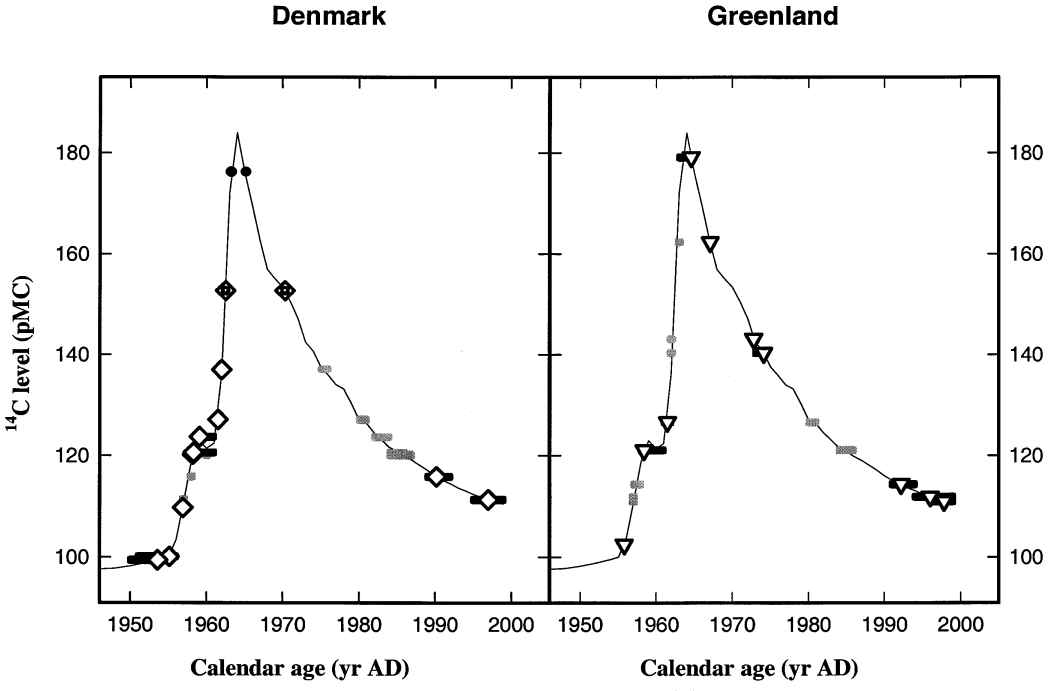

Figure 2 ^{14}C AMS dating results for the Storelungmose (Denmark) and the Tasiusaq (Greenland) peat cores. The solid line represents the annually averaged atmospheric $^{14}CO_2$ curve for the northernmost northern hemisphere provided by I Levin, Heidelberg. This curve was used for calibrating all our data from the peat cores. Symbols denote the maximum-probability age for the respective sample. Dotted symbols mark the two possible solutions for a sample (AAR-5614, see Table 2a) for which it was impossible to decide on one of these solutions from stratigraphic evidence. Horizontal bars denote 95%-confidence intervals, coherent portions in agreement with stratigraphic order are shown in black, portions contradicting stratigraphic order that therefore were rejected are shown in gray. The enclosed circles at 176.8 ± 0.7 pMC show in fact that no dampening occurred in the Danish bog.

In the following we just speculate about the possible consequences of the dampening seen in the Danish core prior to additional dating. Since we measure living moss samples from the surface to be in agreement with atmosphere values this case would then be similar to Jungner et al. (1995), and similarly we may conclude that the mean ^{14}C activity of the CO_2 emitted from decaying sub-surface peat layers is not significantly below 100 pMC. Diluting the atmospheric values with about 35% of emitted CO_2 at 100 pMC (which gives the maximum measured in the Denmark core) may produce a significant shift of up to four years towards older ages regarding the maximum-probability age for the samples AAR-5615, 5616 and 6613 (see Table 2), which are centered around the wiggle in atmospheric $^{14}CO_2$ activity at the end of the 1950s/beginning of 1960s. However, these shifted ages lie only one year outside the confidence intervals (marked in black in Figures 2 and 3) of the respective samples. The calibrated ages of all the other samples on the rising part of the bomb pulse would be much less affected by such a dampening effect.

Emanation of CO_2 from decomposition of layers close to the surface might lead to an apparent time lag for the bomb-pulse peak value in the peat, but in any conceivable scenario pre-peak layers will be dated too old, when calibrated with our general-purpose calibration curve, not too young. For comparison with ^{210}Pb results see later.

Going from 11.5 cm to 15.5 cm there is a significant dip in the ^{14}C content of the respective moss plants (see sample AAR-6615 at 13.5 cm, Table 2a). The order of this excursion (more than 10 percent relative to natural level) corresponds to measurements in cereals from Denmark for the period 1959–62 (Tauber 1967; see Figure 1), which also show a pronounced wiggle (about 130 pMC in 1959, 123 and 121 pMC in 1960 and 1961, respectively, and 137 pMC in 1962). However, the absolute values in our peat core are about 10 percent lower. Such a depletion cannot be explained by a simple model of admixture of 35% peat-derived CO_2 with an activity of 100 pMC as discussed above, but a more elaborate model would be required to cover all the features of a possibly "dampened" curve for a core.

The ^{14}C data from both the Denmark and the Greenland peat core suggest a significantly different accumulation rate between the topmost layers and lower layers (see Figure 3) A linear regression for all ^{14}C data in the Greenland and the Denmark core (omitting only the unresolved two-fold solution for AAR-5614, see Table 2) gives an average accumulation rate of 4.3 and 3.7 mm yr^{-1}, respectively, whereas for the lower layers as shown in Figure 3 one gets 6.9 and 8.2 mm yr^{-1}, respectively. So the peat layers close to the top seem to comprise more years per cm than the lower layers. This is just opposite to what one expects if gravitational compression takes place. One might assume that increased decomposition in the peat layers close to the surface compared to the lower layers is responsible for this effect. However, dry bulk density (DBD) vs. depth profiles, which show a high degree of similarity in both cores and which are also highly correlated with Hg concentration profiles down to about 18 cm (see Goodsite 2000), do not consistently support this assumption. To clarify this question it will be necessary to measure more samples from different depths close to the surface.

Since we have taken 1 cm slices from the individual peat cores, it is evident from the accumulation-rate data given above that on average a single slice contains more than two years. So to get annual resolution for the Hg profiles it therefore might be preferable to measure both ^{14}C and Hg in the annual growth increment of the very same moss plant.

^{14}C dating of macrofossils in a peat core makes it possible to selectively date objects from any depth of the profile in the bomb-pulse period (and possibly also before; see below). (This, of course, is the case only if significant dampening can be excluded, which clearly is the case for both of our cores.) ^{14}C therefore is able to pick up details of the peat evolution, e.g. changes of the accumulation rate, which may serve as an important input for the ^{210}Pb modelling. Moreover, the immediate need for a continuous chronology, i.e. the need to date an entire column with other radiometric means such as ^{210}Pb, to get a date for a certain peat layer is eliminated. However, for flux calculations of e.g. Hg in the associated layers a continuous chronology is still required. Flux calculations derived from ^{14}C measurements should be more precise though account may need to be taken of possible migration of the pollutant relative to the peat matrix. (Regarding a possible different basic trend between ^{14}C and ^{210}Pb—especially in the data for the Denmark core—see the discussion of the ^{210}Pb data below.)

For a given ^{14}C concentration in a sample there is always an (at least) twofold solution in the bomb-pulse period regarding calibrated-age ranges. Therefore it is necessary to measure at least two points of a profile from the bomb-peak period, which are close to each other in depth, in order to determine which side of the bomb-pulse one's points are on. Then—by assuming an undisturbed stratigraphic order of the peat layers—it is generally possible to discard one of the solutions for each sample.

Although ^{14}C dating is commonly regarded as impossible after 1650 (and before the bomb-peak era), we think this is not completely true. Especially the period from 1900 to 1950 shows an almost perfect monotonic decrease in ^{14}C. Single-year data from tree-rings from Douglas firs (grown on the

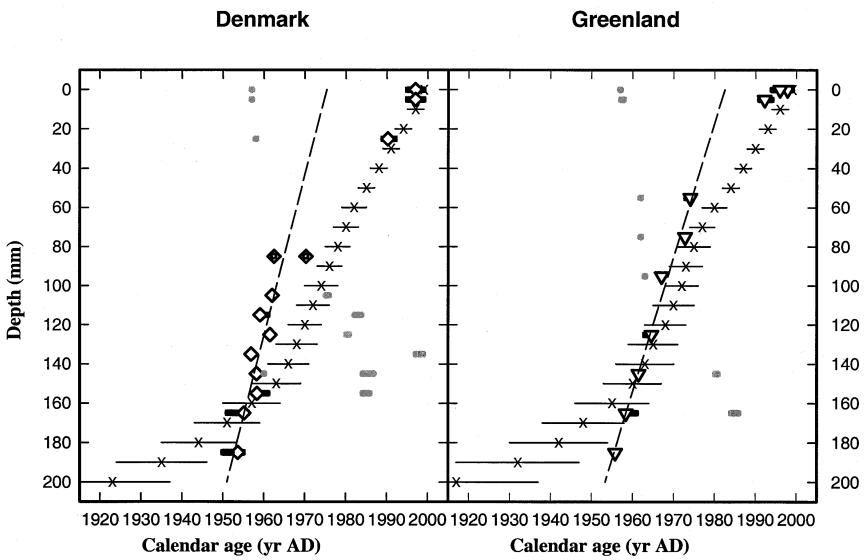

Figure 3 Age-depth profiles for the Storelungmose (Denmark) and the Tasiusaq (Greenland) peat cores. Both ^{14}C AMS dating results and data from ^{210}Pb modelling (corrected according to ^{137}Cs) are shown. ^{14}C results correspond to those shown in Figure 2 (for symbols and bars see caption of Figure 2). A linear regression has been applied to the ages of highest probability at 10.5–18.5 cm depth in the Denmark core and at 5.5–18.5 cm depth in the Greenland core, showing a high linear correlation between age and depth (especially for the Greenland core with a correlation of more than 99%), although one has to be aware that in general peat accumulation rates may deviate a lot from linearity. Data at the top clearly deviate from this regression lines indicating a slower accumulation (see text). ^{210}Pb values and the respective uncertainties (1-σ) are shown as crosses and thin lines. Note the significant deviations in the Denmark core between the ^{14}C data and the respective ^{210}Pb data. Better agreement is found for the Greenland core, though in both cases there are significant discrepancies in the pre-1963 sections.

Olympic Peninsula, 47°N) show a decrease from about −2‰ to −25‰ (Stuiver and Quay 1981) corresponding to a change of about 120 ^{14}C years within 50 cal yr. If therefore another method (such as ^{210}Pb dating) may provide evidence that a peat sample stems from the first half of the 20th century, high-resolution dating with ^{14}C can be performed for this period.

^{210}Pb Dating

^{210}Pb is a naturally occurring fallout radionuclide that is deposited on the bog surface from the atmosphere and incorporated in the bog archive, along with records of atmospherically delivered pollutants such as stable Pb and Hg. Concentrations of ^{210}Pb at different depths in the bog depend on the atmospheric flux at the time the layer was at the bog surface, the net peat accumulation rate (original growth rate minus subsequent losses by organic decay), and the age of the layer. Although globally the atmospheric ^{210}Pb flux may vary spatially by up to an order of magnitude (Appleby and Oldfield 1992), its value at a given location is generally considered to be fairly constant, at least on an annually averaged basis. This assumption is supported by measurements comparing the contemporary flux via rainfall with the long-term flux via soil cores (unpublished data). Because of the effects of varying net peat accumulation rates, the unsupported (atmospherically deposited) ^{210}Pb activity does not usually follow a simple exponential reduction with depth, even when plotted against cumulative

dry mass. The most widely used method for calculating dates in cores with non-exponential records is the CRS (constant rate of ^{210}Pb supply) model (Appleby and Oldfield 1978). This involves measuring ^{210}Pb at regular intervals down to the depth at which ^{210}Pb reaches equilibrium with the supporting ^{226}Ra (ca. 130 years). The results are presented as a continuous set of dates spanning this period. Because of small-scale irregularities over the surface of the bog, the efficiency with which fallout ^{210}Pb is trapped at a given site in the bog may vary through time, causing errors in the CRS model dates. In such cases, independently dated horizons, usually based on records of the artificial fallout radionuclides ^{137}Cs and ^{241}Am from the atmospheric testing of nuclear weapons (peaking in 1963) or the Chernobyl accident (1986), are used to make corrections to the ^{210}Pb dates (Appleby 1998).

The Danish (Storelungmose) and Greenland (Tasiusaq) peat cores had relatively similar ^{210}Pb records. In both cores, ^{210}Pb/^{226}Ra equilibrium was reached at depths of between 24–26 cm. The unsupported ^{210}Pb activity-versus-depth profiles were approximately exponential though with a shallower gradient in the upper part of the core. The profiles from both cores had small non-monotonic features in the top 15 cm. ^{210}Pb dates calculated using the CRS model indicated episodes of rapid peat growth during the past 40 years, though the general trend was one of declining net accumulation rates in the older sections reflecting losses from the peat matrix.

Results for the Denmark and Greenland peat cores revealed significant differences between the ^{210}Pb dates and the 1963 depths indicated by the ^{137}Cs record. In both cores the uncorrected ^{210}Pb dates placed 1963 at a depth of 12.5 cm. The ^{137}Cs stratigraphy suggested that it was significantly deeper in the core, at 14–16 cm in Storelungmose and 15.5 cm in Tasiusaq. (The latter also has a second more recent peak at 3.5 cm that may record fallout from the 1986 Chernobyl accident.) Figure 3 shows the corrected ^{210}Pb dates for each core using the 1963 ^{137}Cs date as a reference point (Appleby 1998). Uncertainties are given as 1-σ intervals, corresponding to 68%-confidence intervals.

Note that shifting the reference point influences basically all the modelled ^{210}Pb dates. E.g. placing the 1963 peak to 12.5 cm instead of 15 cm means that all the dates above this level are also shifted in age although the shift gradually decreases when approaching the surface, which is used as another reference point. Dates below the 1963 level are all shifted by a constant value (6 yr in the case of the Denmark core).

The ^{14}C measurements now offer a means for testing the validity of the ^{210}Pb dating procedure, and in particular the use of ^{137}Cs as an independent time marker to correct the ^{210}Pb results. Since ^{137}Cs fallout from atmospheric weapons tests peaked in 1963, the same year as the NNH atmospheric ^{14}C concentration reached its maximum value, it might be expected that bog records of these two radionuclides should have peaks at similar depths. This, however, is not the case. The ^{14}C peaks occur at 9.5 cm in Storelungmose, and at 12.5 cm in Tasiusaq. The uncorrected ^{210}Pb dates are thus in better agreement with the ^{14}C dates than those corrected by ^{137}Cs.

Although pore-water diffusion of ^{137}Cs in peat and sediment cores is well documented, this is normally assumed to mainly affect the tail of the ^{137}Cs profile. Until now there has been very little direct evidence for a significant displacement of the peak. Two possible causes of this are an initial advective displacement through partially saturated surface vegetation at the time of deposition on the bog surface, or downward diffusion and preferential adsorption onto a layer containing higher concentrations of clay minerals. The apparently larger displacement in the Storelungmose core may be due to its lower mineral content, evidenced by very low ^{226}Ra concentrations (<10 Bq kg^{-1}). Higher

^{226}Ra concentrations in the Tasiusaq core (mean value 21 Bq kg^{-1}) suggest a significant minerogenic input, possibly as wind-blown dust.

A further contributory factor that cannot be discounted is distortion of the ^{137}Cs record by changing peat accumulation rates. Since this also affects ^{210}Pb, the local maximum value of the $^{137}Cs/^{210}Pb$ activity ratio may be a better indicator of the depth of the 1963 fallout maximum than the ^{137}Cs peak itself. In both cores the $^{137}Cs/^{210}Pb$ ratio has a maximum value at 12.5 cm. Regardless, the results presented here show that the use of ^{137}Cs records to validate and correct ^{210}Pb dates is more problematic than previously supposed. Since ^{210}Pb records, and also those of other trace metal pollutants, might be similarly affected, the interpretation of pollutant records in peat bogs, and in particular their relationship to the peat matrix, is an issue that needs to be addressed in greater detail. Bomb ^{14}C, which offers an accurate means for dating the matrix itself, will be an invaluable tool for investigating this relationship.

One problem that needs further investigation is the decrepancy between ^{14}C and ^{210}Pb in pre-1963 sections of the core (see Figure 3) where ^{14}C dates get progressively younger than ^{210}Pb dates (corrected or uncorrected). This cannot be explained either by downward migration of ^{210}Pb (since this would produce younger ^{210}Pb dates) or by emanation of CO_2 from decay of sub-surface layers (see Jungner et al. 1995). As mentioned above, this would lead to a shift towards higher ages.

Mercury

Figure 4 shows an application of the ^{14}C bomb-pulse dating method to Hg concentration profiles measured in the peat cores from Denmark and Greenland. The chronology of concentration changes at the two sites is similar. The similarity of the two curves from two geochemically different mires and different climate regimes is consistent with current views, which suggest global transport of Hg. Our profiles also show a fair correlation with Hg emissions in the northern hemisphere (Pirrone et al. 1998), and this similarity also suggests that peat may be a suitable archive for recording atmospheric Hg, though no conclusions can be drawn about how faithfully the archive preserves Hg concentrations. E.g. the peaks observed in the mid 1960s and the late 1950s for the Greenland core appear to correspond with maxima observed in the ice record (Boutron et al. 1998), though concentrations in the peat are approximately 10,000 times higher. Declines in North American emissions after 1989 have been reported (Pirrone et al. 1998) and declines seen in Hg archives representing the last decade may be related to the closing of major former East German chlor alkali plants and coal-fired power plants (Gerhard Petersen, pers. comm.) At this time we cannot exclude that the declines seen in our peat profiles may be artifacts due to some combination of physical, chemical, and biological processes, but the possibility remains that the declining Hg concentrations since the late 1940s and more sharply during the last decade, reflect a real decline in atmospheric Hg concentrations. The same Hg concentration shape, with the same fall in Hg concentration in the approximate top ten cm has been seen in many other studies including a recent study at the far south latitude location of Patagonia (Biester et al. 2000).

We do not present Hg flux calculations in this paper since more ^{14}C dates are required for samples from a) depths between the surface layers and lower layers to obtain the proper change of the accumulation rate in this region, and b) depths around the 1963 bomb-pulse peak to find out whether dampening takes place in the Denmark core.

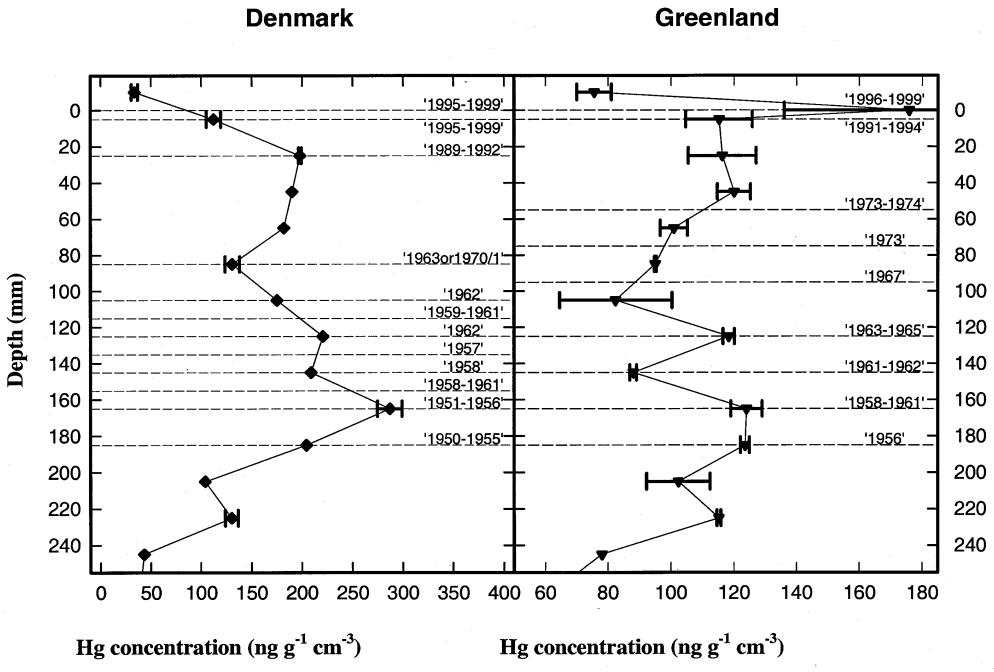

Figure 4 Hg concentration profiles for the Storelungmose (Denmark) and the Tasiusaq (Greenland) peat cores. Hg concentrations are normalized to dry bulk density. Horizontal dashed lines indicate the depth where the macrofossils used for ^{14}C dating were taken. The corresponding calibrated ages are given as numbers close to the lines. Where two Hg concentration measurements were performed unweighted mean values and error bars corresponding to the uncertainty of the mean are shown.

CONCLUSION

By comparing data sets from all over the extratropical northern hemisphere, the present paper shows that ^{14}C dating of plants during the bomb-pulse period is generally possible at a precision of 1–2 years. For the first time, we were able to clearly reproduce the whole atmospheric ^{14}C bomb-pulse curve in peat cores by measuring ^{14}C in macrofossils in peat from Greenland and Denmark, and we could exclude any significant dampening effect for these cores. We compared the ^{14}C bomb-pulse dating method, which allows precise dating of single points in the peat matrix, with the more familiar techniques based on records of fallout radionuclides. ^{14}C is actively taken up into the living material from the surrounding atmosphere and gets fixed via photosynthetic activity along with the stable isotopes ^{12}C and ^{13}C, which provide normalization of the ^{14}C concentrations, allowing direct dating of the material. In contrast, records of fallout ^{210}Pb, together with those of Pb and Hg, may be subject to small displacements at the time of deposition on the bog surface. Comparisons between ^{14}C and ^{210}Pb offer a means for determining a more precise interpretation of pollution records in bog archives.

As an example of the usefulness of the ^{14}C bomb-pulse dating method combined with peat core analysis, we applied the ^{14}C bomb-pulse dating method to two peat cores from Greenland and Denmark to obtain high-resolution dates for Hg concentration profiles for the second half of the 20th century.

The ^{14}C bomb-pulse method is currently being evaluated in peat from the Faroe Islands and another site (Store Vildmose) in Denmark. Aside from the above sites, it will be used to obtain a high-resolution profile of contaminants through time in peat from locations such as Bathurst Island (Canada), and Carey Øerne (the Carey Islands, Greenland) in the high Arctic.

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