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Rapid response of tree cellulose radiocarbon content to changes in atmospheric $^{14}CO_2$ concentration

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ABSTRACT

A detailed radial profile for the ^{14}C concentration in tree stem cellulose, covering growth rings for the years 1962–1964, was obtained for a Sitka spruce of the US Pacific Coast using accelerator mass spectrometry. The tree cellulose ^{14}C closely follows atmospheric $^{14}CO_2$ concentrations, responding to changes with an apparent delay of 5 to 6 weeks. The delay in response is mostly due to the addition of between 13% and 28% of biospheric CO_2 to the canopy-air CO_2 used by the tree for stem cellulose. Delayed incorporation and the use of stored photosynthate of the previous fall appear less important.

1. Introduction

The large, rapid increase in atmospheric $^{14}CO_2$ caused by the nuclear weapons tests immediately preceding the Partial Test Ban Treaty of 1963 acted as a global tracer. This tracer has been used in detailed ^{14}C measurements on stem cellulose deposited by trees to determine how quickly changes in atmospheric CO_2 composition are incorporated in stem cellulose.

In conventional radiocarbon dating, the time resolution of tree ring studies is limited to years (e.g., Stuiver and Quay, 1981; de Jong and Mook 1980; Alekseev et al., 1975; Baxter and Farmer, 1973; Damon et al., 1973) or decades. Maximum bomb ^{14}C has been found in the 1964 ring in the Northern Hemisphere (Kai-Mei Dai and Fan, 1986; Cain and Suess, 1976; Kolesnikov et al., 1970), while the highest atmospheric ^{14}C levels occurred in August 1963 (Levin et al., 1985; Nydal and Lövseth, 1983; Münnich and Roether, 1967).

Accelerator mass spectrometry (AMS) uses only milligram quantities of carbon to measure a ^{14}C concentration. This small sample size allows sampling of the radial concentration profile of cellulose ^{14}C in a tree with sufficient resolution to measure the response of the tree cellulose ^{14}C to change in atmospheric $^{14}CO_2$ concentration on a time scale of weeks.

The difference between ^{14}C concentrations of stem cellulose and atmospheric CO_2 during the years 1962–1964 is caused by the difference in ^{14}C concentration between atmospheric CO_2 and CO_2 recycled into the atmosphere from the biosphere and by tree physiological factors such as isotopic fractionation during photosynthesis and delay between photosynthetic uptake of CO_2 and incorporation of the carbon in stem cellulose. Close comparison of atmospheric and tree ring ^{14}C concentrations provides an indication of the relative importance of these processes. Previous isotopic data on CO_2 uptake by conifers were mainly obtained in laboratory experiments on seedling- and sapling-sized trees (e.g. Shiroya et al., 1966; Gordon and Larson, 1968, 1970; Ursino and Paul, 1973; Schier, 1970; Niziolek et al., 1969; Webb, 1977) with short-duration, high-activity ^{14}C spikes. The results of the present 1962 to 1964 bomb tracer "experiment" pertain to a young mature tree growing under natural conditions.

2. Material and methods

We used the annual growth rings of 1962, 1963 and 1964 of the Sitka spruce (*Picea sitchensis*) from the United States Pacific coast near Quillayute, Washington (47°57′N, 124°33′W), elevation 55 m, about 5 km from the coast. In 1962 the tree was 40 years old. The three rings, 10 to 13 mm wide, were each split into ten equal radial growth increments. The wood was ground and subjected to chemical extraction to isolate alpha-cellulose (Stuiver et al., 1984). The alpha-cellulose was charred in an argon atmosphere at 900°C. The charcoal was powdered and prepared for AMS measurement by encapsulating about 3 mg of it in tantalum (Farwell et al., 1984). The ^{14}C concentrations of the samples were measured using the $^{13}C^{+4}$ ion beams for normalization (Farwell et al., 1983, 1984). Measured AMS ^{14}C beam intensity ratios were translated into conventional ^{14}C values (Stuiver and Polach, 1977) by (i) correcting for a ^{14}C background of about 0.2% of modern carbon, (ii) correcting for isotopic fractionation using the measured $\delta^{13}C$, and (iii) converting into $\Delta^{14}C$. Here $\delta^{13}C$ is the relative difference in isotopic abundance ratio $^{13}C/^{12}C$ between a sample and a standard expressed in per mil (‰):

$$\delta^{13}C = [([^{13}C/^{12}C]_{sample} - [^{13}C/^{12}C]_{standard})/[^{13}C/^{12}C]_{standard}] \times 1000‰,$$

$\Delta^{14}C$ is the relative difference in ^{14}C activity (‰) between a sample and 0.95 times the decay-corrected activity of CO_2 prepared from the United States National Bureau of Standards oxalic acid radiocarbon standard (normalized for isotopic fractionation to $\delta^{13}C(PDB) = -25‰$ (sample) and to $\delta^{13}C(PDB) = -19‰$ (oxalic acid CO_2)).

The results shown in Fig. 1 are the respective averages of all measurements made for the individual growth increments. The uncertainty for each is based on the reproducibility of the individual measurements or on their statistical uncertainty, whichever is larger.

3. Timing of radial growth

Radial tree growth data are not available in the Quillayute area for the 1962–1964 period. Therefore we use data for Douglas fir [*Pseudotsuga*

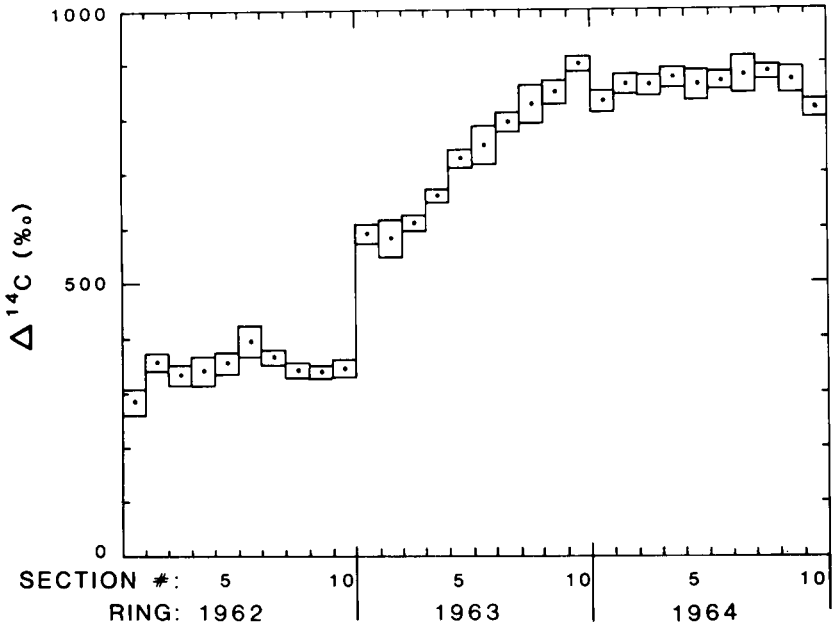

Fig. 1. Radial cellulose $\Delta^{14}C$ profile for the years 1962, 1963, and 1964 of a Sitka spruce (*Picea sitchensis*) grown near Quillayute on the United States Pacific coast (47°57′N, 124°33′W).

menziesii (Mirb.) Franco] trees growing about 200 km away in the University of British Columbia (UBC) Research Forest (49°18′N, 122°34′W) (Griffith, 1968) to estimate the timing of the radial growth of our Sitka spruce (Table 1). This assumes that site location, climate, species, and tree age all combine to give similar growth curves for the UBC Douglas fir and the Quillayute Sitka spruce.

Such an assumption appears to be very reasonable. Both sites are located near the Pacific Ocean in the marine temperate climate zone west of the Cascade Range. Anomalies in mean monthly temperature and monthly precipitation (Climatological Data, 1961–1967) correlate strongly between the 2 sites indicating they are influenced by the same weather patterns. Seasonal radial growth curves may vary considerably from year to year depending on the weather. Yet in a particular year their timing differs little among conifers of different species and different ages, especially for the period of rapid radial growth from 10% to 90% of the ring width. We estimate an uncertainty of plus and minus five days for the timing of the inferred growth pattern of our Sitka spruce from the growth curves reported by Reukema (1965) and Godman and Gregory (1955). This is the uncertainty in the timing of the period of 10 to 20 days (Table 1) represented by the 10% growth increment.

With the inferred growth pattern we have converted the measured ^{14}C concentrations for the radial segments into the ^{14}C concentration of tree cellulose as a function of time (Fig. 2). Most of the 1963 $\Delta^{14}C$ data were presented at the 1984 Zürich AMS symposium (Farwell et al., 1984). There we used a preliminary time scale that was based on constant radial growth throughout a March through October growing season. The more realistic April through September growing season and the s-shaped growth curve (Griffith, 1968) used in timing the ^{14}C change in Fig. 2 lead to a significantly different rate of change of $\Delta^{14}C$.

A wealth of atmospheric $\Delta^{14}C$ data exists for the second part of 1963 and later years, but during 1962 there were only a few stations where atmospheric CO_2 was sampled regularly. The $\Delta^{14}C$ values for the most relevant stations are shown in Fig. 2 for comparison with our tree ring results (Levin et al., 1985; Nydal and Lövseth, 1983; Münnich and Roether, 1967; Berger et al., 1965). $\Delta^{14}C$ values of tree ring cellulose seem to follow those of atmospheric CO_2 with a delay of 5 to 6 weeks (the delay needed to match the Vermunt and the Sitka $\Delta^{14}C$ values during the period of rapid rise in 1963, see also section 4, below).

The highest cellulose $\Delta^{14}C$ value ($+897 \pm 14‰$) occurs at the end of the 1963 growing season when tropospheric $\Delta^{14}C$ levels are also at their maximum (Levin et al., 1985; Nydal and Lövseth, 1983; Münnich and Roether, 1967; Olsson and Karlén, 1965). The 1964 maximum reported for single-year ring studies on Northern Hemisphere trees and for cereals (Table 2) is the result of taking a seasonal average

Table 1. *Estimated growth period for 10% increments of the radial growth of a Sitka spruce on the United States Pacific Coast (47°57′ N, 124°33′ W) during the years 1962, 1963, and 1964 based on Griffith, 1968*

Growth increment %	Growth period		
	1962	1963	1964
0–10	April 10–May 7	April 14–May 13	May 1–May 22
10–20	May 7–May 25	May 13–May 23	May 22–June 1
20–30	May 25–June 6	May 23–June 2	June 1–June 10
30–40	June 6–June 17	June 2–June 16	June 10–June 23
40–50	June 17–June 26	June 16–June 27	June 23–July 4
50–60	June 26–July 7	June 27–July 9	July 4–July 14
60–70	July 7–July 21	July 9–July 20	July 14–July 26
70–80	July 21–Aug. 1	July 20–Aug. 3	July 26–Aug. 7
80–90	Aug. 1–Aug. 18	Aug. 3–Aug. 18	Aug. 7–Aug. 21
90–100	Aug. 18–Sept. 29	Aug. 18–Sept. 14	Aug. 21–Sept. 15

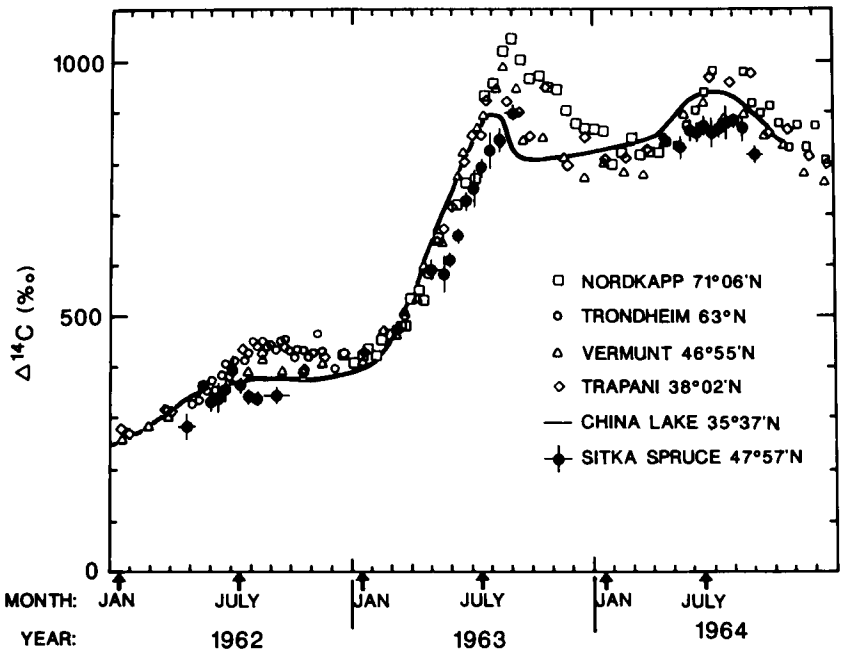

Fig. 2. Radial cellulose $\Delta^{14}C$ values in a Sitka spruce (47°57′N, 124°33′W) for the years 1962, 1963, and 1964, (●) compared with directly measured atmospheric $\Delta^{14}C$ values at Trondheim (○) (63°16′N, 10°22′E) and Nordkapp (□) (71°06′N, 23°59′E), Norway, (Nydal and Lövseth, 1983), at Vermunt (△) (46°55′N, 10°03′E), Austria, (Levin et al., 1985), at Trapani (◇) (38°02′N, 12°31′E), Sicily, Italy (Münnich and Roether, 1967), and at China Lake (−) (35°37′N, 117°41′W), California, U.S.A., (Berger et al., 1965; graph only no data points).

and is reflected by the average $\Delta^{14}C$ values of the 1963 and 1964 rings of our Sitka spruce (726‰ and 858‰, respectively). Weighted average values for atmospheric $\Delta^{14}C$ over the Sitka spruce growing seasons of these years agree well with the tree ring data (Table 2). The apparent one-year delay in tree ring response to atmospheric $\Delta^{14}C$ change observed in the earlier studies is the result of comparing cellulose $\Delta^{14}C$ values averaged over the growing season with a record of weekly atmospheric $\Delta^{14}C$ values.

The delay in Sitka spruce cellulose $\Delta^{14}C$ change relative to the atmosphere can be used to estimate the influence of the local environment and of physiological processes on the incorporation of atmospheric CO_2 in tree rings. Before this can be done, however, we have to decide on the $\Delta^{14}C$ values to be used for the clean, free atmosphere at the growth site of our Sitka spruce.

4. $^{14}CO_2$ in the clean atmosphere

There are 2 probable causes for the differences in atmospheric ^{14}C concentration between various stations, viz. (i) incomplete mixing of the latitudinally unevenly distributed input of fossil fuel CO_2 and bomb ^{14}C into the free troposphere on a time scale of days to weeks, and (ii) contamination by local biospheric CO_2 of the air sampled at a station.

4.1. Incomplete mixing

Nydal and Lövseth (1983) consider the Northern Hemisphere between 30° and 90°N well mixed. Lal and Rama (1966) used the same simplification in their mixing model, specifying a time scale (2 to 3 weeks) and a precision (within a few percent) for which it was valid. Evaluation of the relatively small difference in $\Delta^{14}C$ between tree ring cellulose and atmospheric CO_2 over a growing season requires both a shorter time scale and a higher precision (see Fig. 2). The atmospheric $\Delta^{14}C$ data in Fig. 2 show seasonally-dependent latitudinal differences between the stations during the years of maximum bomb ^{14}C (see Fairhall and Young, 1970). This latitude dependence is also evident in the tree ring results

Table 2. *Northern hemisphere tree ring and atmospheric CO_2 $\Delta^{14}C$ values during the years 1962–1965*

Sample	Location	Ref.	$\Delta^{14}C$ (‰)			
Trees:			1962	1963	1964	1965
Mackenzie Delta, Canada						
White spruce	68°N, 130°W	1	606 ± 5	895 ± 5	964 ± 5	915 ± 5
Lowland, USSR						
Pine	60°25′N, 30°45′E	2	385	751	878	705
Treeless height, USSR						
Pine		2	418	780	888	722
Kiel, Germany						
Ulmus	54°20′N, 10°07′E	3	407 ± 22	844 ± 10	906 ± 15	
Quillayute, Washington, USA,						
Sitka spruce	47°57′N, 124°33′W	4	345 ± 6	726 ± 6	858 ± 6	
Dailing, China						
Spruce	47°30′N, 129°16′E	1	392 ± 5	771 ± 5	909 ± 5	843 ± 5
Bear Mountain, New York, U.S.A.,						
Red Oak	41°20′N, 74°00′W	5	360	732	888	788
Central Park, New York, U.S.A.,						
Oak	40°45′N, 74°00′W	5	320	614	740	654
Mingyin, China						
Spruce	27°13′N, 100°20′E	1	325 ± 5	497 ± 5	743 ± 5	758 ± 5
Atmospheric CO_2:						
Trondheim/Nordkapp, Norway	63°N, 10°E/ 71°06′N, 23°59′E	6	395 ± 14	811 ± 48	944 ± 16	794 ± 6
Vermunt, Austria	46°55′N, 10°03′E	7	380 ± 6	825 ± 36	905 ± 4	782 ± 6
China Lake, California, U.S.A.	35°37′N, 117°41′W	8	364 ± 5	798 ± 28	923 ± 5	776 ± 5
Trapani, Sicily, Italy	38°02′N, 12°31′E	9	399 ± 12	798 ± 36	946 ± 9	798 ± 2
Cereals: (Oats, Barley)						
Copenhagen, Denmark, 1 July	55°50′N, 12°30′E	10	385 ± 8	794 ± 11	945 ± 8	844 ± 11

1. Kai-Mei Dai and Fan (1986)
2. Kolesnikov et al. (1970)
3. Willkomm and Erlenkeuser (1968)
4. This work
5. Cain and Suess (1976)
6. Nydal and Lövseth (1983)
7. Levin et al. (1985)
8. Berger et al. (1965); Berger and Libby (1966)
9. Münnich and Roether (1967)
10. Tauber (1967)

in Table 2 and in detailed measurements of atmospheric CO_2 concentrations and of CO_2 $\delta^{13}C$ values in clean air (Pearman and Hyson, 1980; Mook et al., 1983; Keeling et al., 1984).

The large injection of bomb-produced stratospheric $^{14}CO_2$ into the troposphere at mid-northern latitudes during each spring resulted in ^{14}C concentrations that were higher in the mid-latitudes (Vermunt, 47°N) than at high latitude (Nordkapp, 71°N). Despite the low mixing resistance of the 30° to 90°N reservoir observed maximum differences vary from 20‰ for May 1962 to 70‰ for June 1963. When the rate of injection decreased, southward mixing with air of lower ^{14}C concentration in the 0–30°N and southern hemisphere reservoirs and exchange with the oceans led to declining concentrations in the 30° to 90°N reservoir and lower values at Vermunt than at Nordkapp (observed maximum difference from 50‰ in September 1962 to 110‰ in October–November 1963). The lower $\Delta^{14}C$ values and the 1964 maximum observed at China Lake (Berger et al., 1965) and Trapani (Münnich and Roether, 1967) result from the proximity to the 0° to 30°N reservoir. These stations fall in the transition zone (30° ± 5°N) of Lal and Rama (1966). Because of the latitudinal differences in tropospheric $\Delta^{14}C$ levels the record of Vermunt at 46°55′N is the best that is available to indicate tropospheric $\Delta^{14}C$ values at the site

of our Quillayute Sitka spruce (47°57′N). A complication is the strong western-European fossil fuel CO_2 input which lowered European tropospheric $\Delta^{14}C$ values (European Suess effect). For Vermunt this contribution is estimated at 5‰ during this period (Levin et al., 1985).

4.2. Local biospheric CO_2

Air sampled by the Vermunt station may not be truly representative of the free troposphere because the sampling station is located in a valley. An extra, local addition of biospheric CO_2 is indicated by the difference between $\delta^{13}C$ values at Vermunt and at the mountain-top station Schauinsland (48°N, 8°E, 1205 m elevation; I. Levin, personal communication, 1985). The difference was 0.55 ± 0.16‰ during the 1977 through 1983 growing seasons (based on Levin et al., 1985) indicating on average about 3% CO_2 of biospheric origin in the Vermunt samples. This contamination with biospheric CO_2 resulted in lower $\Delta^{14}C$ values at Vermunt during the period of rapid atmospheric ^{14}C increase. Münnich (1963) calculated from measurements on soil CO_2 efflux during the period 1958–1962 a time lag between soil CO_2 and the atmosphere of two to three years. Soil CO_2 is, however, produced from a mixture of organic materials with residence times varying from less than a year to well over 100 years (e.g, Dörr and Münnich, 1986; O'Brien, 1984; Harkness et al., 1986). Our correction of the Vermunt $\Delta^{14}C$ for a 3.2% biospheric contamination is based on a model of biospheric CO_2 production (Table 3) that considers the complexity of this mixture. The corrections are small, from 5 to 19‰.

The best indicator for the $\Delta^{14}C$ of the Pacific coast clean air sampled by our Sitka spruce is the Vermunt atmospheric $\Delta^{14}C$ record after it has been corrected for the local European Suess effect of 5‰ (Levin et al., 1985) and for the local biospheric contamination.

5. Factors influencing tree-stem cellulose ^{14}C concentration

Our Sitka spruce stem-cellulose $\Delta^{14}C$ values ($\Delta_c{}^{14}C$) are, with one exception, significantly lower than $\Delta^{14}C$ for the Vermunt atmosphere. Thus local conditions and/or tree physiology resulted in a depression of $\Delta_c{}^{14}C$. The $\Delta^{14}C$ differences between clean tropospheric air (the corrected Vermunt data) and Sitka stem-cellulose are shown in Fig. 3. The uncertainty is largely that of the AMS $\Delta^{14}C$ measurement. The differences do not show a clear seasonal trend. The weighted average is 51 ± 5‰, 137 ± 7‰, and 62 ± 6‰ in 1962, 1963, and 1964 respectively.

$\Delta^{14}C$ indicates the relative deviation from the ^{14}C standard, corrected for isotopic fractionation. Therefore, the lower cellulose $\Delta^{14}C$ values reflect one or more of the following:

(i) Input of CO_2 produced by biomass decomposition (biospheric CO_2) into the air sampled by the tree crown. Literature data on the CO_2 flux connected with organic decomposition in and on the ground are scarce and show large differences among ecosystems. No $\Delta^{14}C$ measurements of the canopy air were made at the time of the bomb ^{14}C tracer "experiment". The flux and the $\Delta^{14}C$ value of biospheric CO_2 contributing to the growth of our Sitka spruce during these years must therefore be estimated. We calculate a $\Delta^{14}C$ value for the biospheric CO_2 ($\Delta_b{}^{14}C$) from the model for biospheric CO_2 production summarized in Table 3, and we will use our data to estimate the biospheric contribution to the tree's photosynthate. From Table 3 it is clear that root activity [i.e., turnover of fine roots (<2 mm diameter) and root respiration] is a major factor in determining $\Delta^{14}C$ of soil CO_2. The $\Delta^{14}C$ value of the CO_2 produced by forest litter depends strongly on the relative contributions of long-lived, slowly-decomposing conifer parts and short-lived, rapidly-decomposing epiphytes/vascular plants. Calculated $\Delta_b{}^{14}C$ values, therefore, have a considerable uncertainty resulting not only from the uncertainty in the $\Delta^{14}C$ values of the materials contributing to the biospheric CO_2 flux, but also from the uncertainty in their relative contributions to this flux.

(ii) Delayed incorporation of recent photosynthate into stem cellulose. In Scots pine seedlings (*Pinus silvestris* L.) ^{14}C incorporation into stem cellulose was detected 2 h after exposure of the needles to $^{14}CO_2$, but incorporation still continued after two days (Niziolek et al., 1969). Rangnekar et al. (1969) found in 15-year-old red pine (*Pinus resinosa* Ait.) incorporation of ^{14}C in stem cellulose for three to seven days after a branch was allowed to take up ^{14}C labeled CO_2

Table 3. *Estimating biospheric CO_2 $\Delta^{14}C$ values*

(a) Components of Soil CO_2. Estimated relative contributions and ages for our lowland, evergreen forest, Sitka spruce growth site are based on papers by Ågren and Axelsson, 1980; Ågren et al., 1980; Anderson, 1973; Dörr and Münnich, 1986, Edwards and Harris, 1977; Edmonds, 1980, 1984; Grier and Logan, 1977; Linder and Troeng, 1981; Schulze et al., 1977; Vogt et al., 1983; Vogt K. A., personal communication, 1987

	Fraction (%)	Material	Fraction (%)	Age[1] (yr)	$\Delta^{14}C$ Value (‰)[2] 1962	1963	1964
litter 01	15	needles	50	4	+150	+260	+220
		epiphytes and vascular plants	30	1	+230	+345	+726
		twigs	10	4	+150	+260	+220
		cones	10	1	+230	+345	+726
		total	100				
litter 02	15	needles	55	10	−20	−20	−20
		epiphytes and vascular plants	14	2	+220	+230	+345
		twigs	16	10	−20	−20	−20
		cones	15	10	−20	−20	−20
		total	100				
Soil organics	5			100	−20	−20	−20
pine roots	45			1	+230	+345	+726
root respiration[3]	20			recent[4]	+345	+726	+858
total	100						

[1] Litter production is assumed to occur in fall after the growing season, and fine roots are assumed to live one growing season. Decay of the current year's litter or fine roots would increase $\Delta^{14}C$ in soil CO_2 later in the season.

[2] Estimated $\Delta^{14}C$ values are based on a compilation of atmospheric $\Delta^{14}C$ values by Tans (1981) (for pre-1959 values) and on Münnich and Roether (1967) (for 1959–1961), and on Sitka stem-cellulose $\Delta^{14}C$ for the years 1962–1964. The average atmospheric value over an April to September growing season was reduced by 10‰ to take into account the difference between atmospheric and biospheric $\Delta^{14}C$ values.

[3] The ratio decay-respiration is kept constant in our calculations.

[4] Respiration CO_2 is assumed to come from recent photosynthate. Its $\Delta^{14}C$ value varies over the growing season with that of the Sitka stem cellulose.

(b) Biospheric $\Delta^{14}C$ values calculated from the soil CO_2 breakdown of Table 3(a) and an extra addition by stem and branch respiration of 5% of the soil CO_2. Respiration $\Delta^{14}C$ values correspond to those of Sitka stem cellulose. Needle respiration is assumed to occur mainly near the outside of the crown where air mixes more readily

Growth increment	$\Delta^{14}C$ (‰) 1962	1963	1964
1	194	331	569
2	210	329	576
3	205	336	575
4	207	347	579
5	210	363	575
6	220	369	577
7	213	379	580
8	207	387	581
9	207	392	577
10	208	404	565

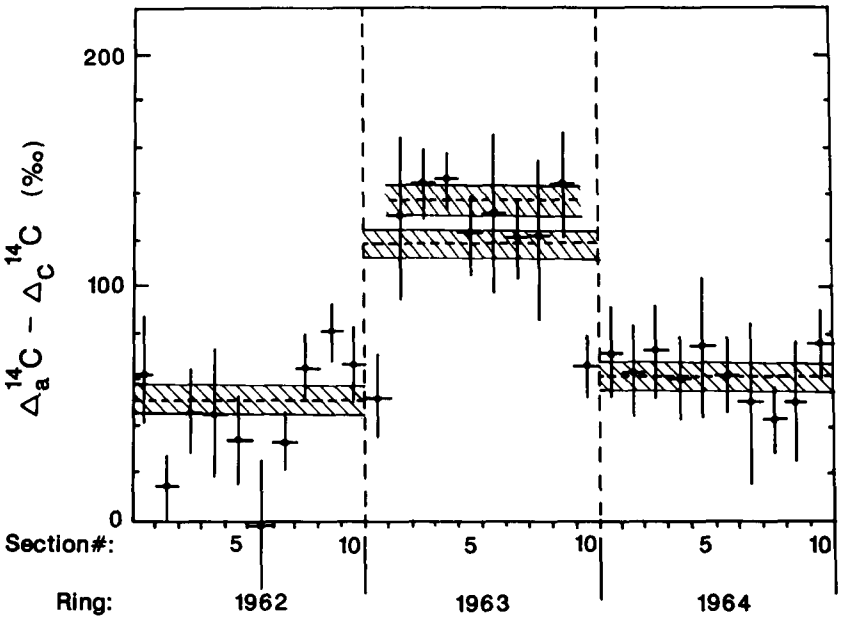

Fig. 3. $\Delta^{14}C$ difference between CO_2 in the clean tropospheric air at 47°–48°N ($\Delta_a{}^{14}C$) and Sitka spruce stem-cellulose ($\Delta_c{}^{14}C$). The air values are those of Vermunt (47°57′N, Levin et al., 1985) corrected for a 3.2% biospheric CO_2 addition and for an extra European Suess effect. Weighted average differences are 51 ± 5‰, 118 ± 6‰ (137 ± 7‰ without increments #1 and 10), and 62 ± 6‰ in 1962, 1963, and 1964 respectively. The uncertainty is largely that of the AMS $\Delta^{14}C$.

during its period of rapid growth (June). Balatinecz et al. (1966) similarly obtained 3 to 4 days for actively growing 8-month-old jack pine (*Pinus banksiana* Lamb). During 1963, when $\Delta^{14}C$ in the air ($\Delta_a{}^{14}C$) increased by about 3.5‰ per day in April, May, and June, delayed incorporation could make $\Delta^{14}C$ values in cellulose significantly lower than those in contemporary air. Internal recycling of CO_2 (Zelawski et al., 1970) as well as renewed uptake of respiration CO_2 contribute to the average delay between initial photofixation and incorporation.

(iii) The use of stored photosynthate from the previous fall. This is a form of long-delayed incorporation, important in our case because of the large difference in atmospheric $\Delta^{14}C$ values between fall 1961/spring 1962 and fall 1962/spring 1963. As with the biospheric CO_2 we will estimate the $\Delta^{14}C$ value of the stored photosynthate ($\Delta_s{}^{14}C$) and use our data to estimate its contribution x_s to Sitka spruce stem growth. A problem in estimating $\Delta_s{}^{14}C$ is that net photosynthesis in conifers continues after radial growth ends. For the Washington Pacific coast where the Sitka grew net photosynthesis may occur during most of the fall and winter (Neilson et al., 1972; Linder and Troeng, 1980; R. B. Walker, pers. comm., 1987). We, arbitrarily, restrict our discussion of stored photosynthate to that produced during the preceding calendar year in order to have an analogy with deciduous trees and avoid a continuum of (ii) and (iii). During the fall and winter of 1961 and 1962 the changes in $\Delta^{14}C$ in the air were minor while the increase during the subsequent 1962 and 1963 growing seasons was rapid and large (c.f. Fig. 2). Under these conditions the uncertainty in the estimated $\Delta_s{}^{14}C$ does not seriously influence our calculation of x_s. The decrease in $\Delta_a{}^{14}C$ from about 990‰ in August 1963 to about 800‰ by early 1964 exceeds the range of tropospheric $\Delta^{14}C$ values during the 1964 growing season and leads to an uncertainty in $\Delta_s{}^{14}C$ too large to estimate a stored photosynthate contribution for 1964.

6. Numerical estimates of the use of biospheric CO_2 and stored photosynthate

As discussed above the cellulose $\Delta^{14}C$ value $\Delta_c{}^{14}C$ is the result of contributions by clean air CO_2 ($\Delta_a{}^{14}C$), biospheric CO_2 ($\Delta_b{}^{14}C$) and stored photosynthate ($\Delta_s{}^{14}C$) and an incorporation delay (Δt).

$$\Delta_c{}^{14}C = (1 - x_b - x_s)[\Delta_a{}^{14}C - (\partial\Delta_a{}^{14}C/\partial t)\Delta t] + x_b[\Delta_b{}^{14}C - (\partial\Delta_b{}^{14}C/\partial t)\Delta t] + x_s\Delta_s{}^{14}C.$$

Here x_b and x_s are the fraction of the total carbon contributed by biospheric CO_2 and by stored photosynthate, and $\partial\Delta/\partial t$ is the rate of increase in $\Delta^{14}C$. Only $\Delta_c{}^{14}C$ has been measured directly; $\Delta_a{}^{14}C$ has been deduced from the Vermunt data. $\Delta_b{}^{14}C$ and $\Delta_s{}^{14}C$ values can be estimated using our model in Table 3 and published $\Delta_a{}^{14}C$ data.

The measured and estimated $\Delta^{14}C$ values for 1962, 1963, and 1964 allow us to determine the range of possible biospheric CO_2 and stored photosynthate contributions to the Sitka stem cellulose. In 1962 and 1963 $\Delta_b{}^{14}C$ and $\Delta_s{}^{14}C$ values were both below those of cellulose. A *maximum* value for one is then obtained by assuming the other to be zero and allowing no delay in incorporation. In 1964 $\Delta_s{}^{14}C$ of stored photosynthate, assumed to have been produced in the fall of 1963, is higher than that of the 1964 cellulose and $\partial\Delta/\partial t$ is small. Then only the addition of a certain minimum amount of biospheric CO_2 can produce the observed lowering of $\Delta_c{}^{14}C$ relative to $\Delta_a{}^{14}C$ and $\Delta_s{}^{14}C$. The combination of 1963 and 1964 defines the range of possible biospheric CO_2 incorporation x_b in the Sitka stem-growth. As stated above, no x_s can be estimated for 1964.

6.1. *Biospheric CO_2*

Biospheric CO_2 contributions x_b calculated using Table 3 are shown in Fig. 4. The uncertainty intervals are based on the uncertainty in the measured $\Delta_c{}^{14}C$ values. A similar calculation was made considering only decay CO_2 (no respiration in Table 3). The weighted average maxima are for biospheric CO_2 $27.9 \pm 2.8\%$ and $27.6 \pm 1.3\%$ and for decay CO_2 $22.4 \pm 2.3\%$ and $22.4 \pm 1.1\%$ in 1962 and 1963 respectively. The anomalously low x_b value of increment #10 in 1963 has been excluded from the average to obtain a conservative estimate for the upper limit of x_b. Taking x_s and Δt zero in 1964 gives an x_b of $18.0 \pm 1.7\%$ and $14.4 \pm 1.4\%$ for biospheric CO_2 (Fig. 4) and decay CO_2 respectively. The minimum biospheric CO_2 contribution for 1964 can be somewhat less than calculated if we include stored photosynthate. Production during fall and early winter of 1963 must have led to an average $\Delta_s{}^{14}C$ between the 897‰ of 1963 increment #10 and the 828‰ of 1964 #1. Replacing air CO_2 by this stored photosynthate, however, lowers the amount of biospheric CO_2 needed only slightly because the $\Delta^{14}C$ values are close to each other. From Fig. 4 it is clear that values of $x_b > 28\%$ exceed most of the uncertainty intervals for 1963. Similarly values of $x_b < 13\%$ will be below the majority of the 1964 uncertainty intervals. An upper estimate for the range of possible contributions of biospheric CO_2 to our Sitka spruce stem cellulose is thus from 13 to 28%. The contribution of decay CO_2 (from 10 to 23%) is better defined than that of the total biospheric CO_2 since it is independent of the relative contribution of respiration (root, stem/branch, and some of the crown) to the subcanopy CO_2 production.

The x_b range can be compared with conclusions drawn from the concentration and $\delta^{13}C$ values of atmospheric CO_2 and from the $\delta^{13}C$ values of leaves growing at different height above the ground.

(i) Concentration and $\delta^{13}C$ of atmospheric CO_2 in the temperate rain forest of the Washington Pacific coast were measured by Keeling (1958, 1961) and Burk (R. L. Burk, personal communication, 1985). Keeling (1958) found in 1955 at 1 m above ground under a dense Washington rainforest canopy extreme values of 410 ppm and 312.6 ppm CO_2. The $\delta^{13}C$ values of the CO_2 varied from -10.79‰ to -6.66‰ relative to the PDB standard. A value of 314 ppm with $\delta^{13}C = -6.87$‰ obtained at the Washington coast was presumed to represent "background" air. Relevant for a biospheric CO_2 contribution to photosynthate are the CO_2 concentrations during the day. At 06.15, 09.15 hrs, and 13.00 hrs, these were respectively, 398.2 ppm ($\delta^{13}C = -10.54$‰), 387.9 ppm ($\delta^{13}C = -9.77$‰), and 352.5 ppm ($\delta^{13}C = -8.45$‰) on September 6, 312.6 ppm ($\delta^{13}C = -6.66$‰) on September 7. During the early part of the

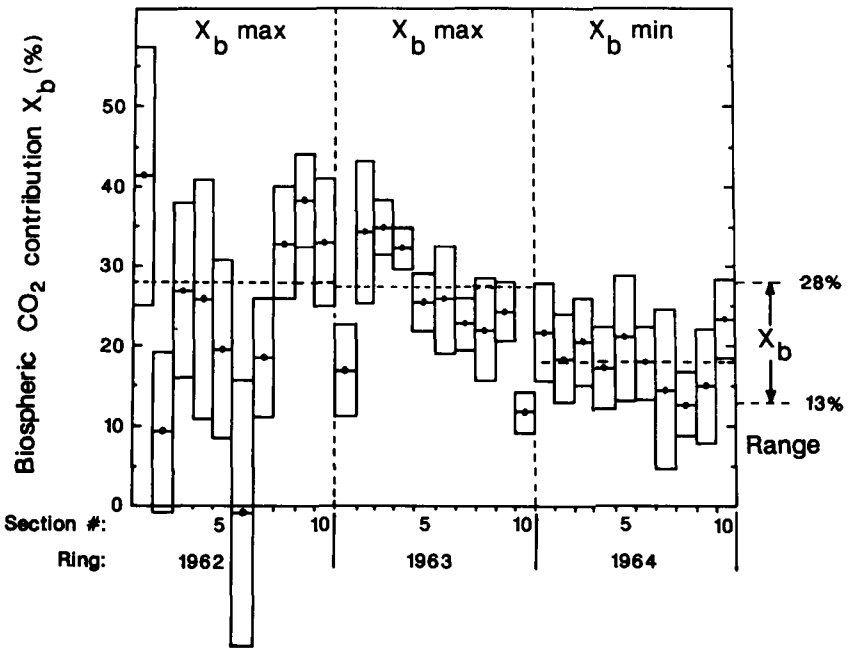

Fig. 4. Biospheric CO_2 contribution x_b to the Sitka stem-cellulose. For 1962 and 1963 a maximum value for x_b is obtained by assuming zero contribution of stored photosynthate (x_s) and zero delay in incorporation. For 1964, x_b calculated with the same assumptions represents a minimum amount of biospheric CO_2 needed to produce the observed lowering of cellulose $\Delta^{14}C$ relative to atmospheric $\Delta^{14}C$ since the stored photosynthate $\Delta^{14}C$ of fall 1963 is above that of the 1964 cellulose. The uncertainty intervals are based on the AMS $\Delta^{14}C$ uncertainty.

morning biospheric CO_2 made up from 21 to 19% of the total canopy CO_2 near the ground, dropping to 11% and 0% by 13.00 hrs on September 6 and 7, respectively. The difference between the 2 readings at 13.00 hrs probably is due to a 0.5 – 1 m/s wind on the 2nd day. Burk observed near the coast a $\delta^{13}C$ depression by 2.5‰ at 5 a.m. (under dense canopy, no wind) that decreased to 0.8‰ by 8 a.m. (3 m/s wind) and to 0.4‰ under less dense canopy 75 m from the beach. This indicates admixture of 13.9%, 4.4%, and 2.2% of CO_2 of biospheric origin with $\delta^{13}C = -25‰$. An average $\delta^{13}C$ value of $-24.0‰$ for our three rings and a $\delta^{13}C$ value of $-22.3‰$ for 48°N in 1965 (Stuiver and Braziunas, 1987 and unpublished data) indicate that $\delta^{13}C$ of the biospheric CO_2 may locally have been less negative than $-25‰$ and the required biospheric contribution consequently larger. These local observations agree with the general diurnal biospheric modulation of the atmospheric CO_2 concentration both in a forest environment and over wheat/crop fields (Lieth, 1963).

The return of the canopy air to "clean air" values observed by Keeling and Burk can be the result of mixing with the air above the canopy which would minimize biospheric CO_2 influence. It can, however, also be accomplished by photosynthetic uptake of an amount of CO_2 equal to the excess CO_2 plus any biospheric CO_2 produced during the day. Return of the concentration and $\delta^{13}C$ of subcanopy CO_2 to clean air values then simply reflects no net flux of carbon between biosphere and atmosphere. Under these conditions $\Delta_a{}^{14}C$ under the canopy can differ from that of the free air since it depends on the total flux of biospheric decay CO_2 into the air due to the large difference in $\Delta^{14}C$ values between decaying organic matter and the clean troposphere during 1962 to 1964. Both mixing and uptake will

occur simultaneously. Their relative importance will be determined by local wind and local conditions for photosynthesis. Consequently, the biospheric CO_2 contribution to the tree's photosynthate may have been quite variable. The concentration and $\delta^{13}C$ values of atmospheric CO_2 discussed above thus show that a considerable amount of biospheric CO_2 was produced, but they are inconclusive as to the effect this had on $\Delta^{14}C$ values of the air under the canopy. Uptake of biospheric CO_2 is favored by the fact that photosynthesis in spruce and fir is most rapid around mid morning (Lieth, 1963) when the CO_2 excess is still relatively high.

(ii) $\delta^{13}C$ values of leaves growing at different heights above the ground have been interpreted as showing a biospheric CO_2 input that decreases with increasing height (Vogel, 1978; Medina and Minchin 1980; Schleser and Jayasekera, 1985). The calculated biospheric CO_2 contribution is 15% for leaves 2 m above the ground in a Bavarian forest (Vogel, 1978) and 20% for the shade flora in Amazonian rain forests (Medina and Minchin, 1980) if the total $\delta^{13}C$ change with height is attributed to biospheric CO_2. Schleser and Jayasekera (1985) argue that the physiological state of the leaves and assimilatory performance of the tree contribute to the observed differences. This reduces their estimate of the contribution of soil respired CO_2 to the total assimilation by almost 65%. For the lower part (1–5 m) of the canopy the reduction is less leaving an estimated 22% contribution in a German forest. These values agree with the range estimated from our $\Delta^{14}C$ results.

Similar $\delta^{13}C$ differences have been reported for a Tasmanian rain forest (Francey et al., 1985; Francey, 1986) and a subtropical Chinese monsoon forest (Ehleringer et al., 1986, 1987). These authors favor a decreasing light intensity under the canopy as the cause of the observed $\delta^{13}C$ changes and consider the influence of biospheric CO_2 minor. Light certainly is an important factor determining photosynthesis and isotope fractionation as proposed by Francey et al. (1985). Yet increased CO_2 levels, 19 ppm ~ 5.3% and 7 ppm ~ 2% of biospheric CO_2 in the subcanopy CO_2 at 1 and 5 m above the ground between 9.00 hrs and 17.00 hrs local time (Francey et al., 1985), show the influence of biospheric CO_2 in the Tasmanian rainforest. Considering the general diurnal CO_2 concentration cycle (Keeling, 1958, 1961; Lieth, 1963) significantly higher CO_2 levels may have existed under the canopy during the early morning hours before 9.00 hrs. The predominance of morning net photosynthesis (Lieth, 1963; also Francey et al., 1985) thus favors incorporation of CO_2 of biospheric origin. A combination of a biospheric CO_2 vertical mixing gradient with physiological influences, both genetic and environmental (such as light intensity and humidity), as proposed by Schleser and Jayasekera (1985), is likely to offer the best description of the complex ecosystem interactions that determine the composition of the photosynthate.

A 40-year old Sitka spruce certainly reaches higher than 5 meters so it is important where the photosynthate used in radial stem growth was taken up. ^{14}C tracer experiments by Ross (1972) on source-sink relationships in the branch of 10-year old Douglas firs showed that photosynthate translocated to the stem of the tree comes only from the older needles, closer to the stem, during its period of rapid diameter growth. Such older needles still take up a considerable fraction of the CO_2 (Schulze et al., 1977). Rangnekar et al. (1969) showed that in 15-year-old red pine (*Pinus resinosa* Ait) photosynthate exported by a branch moved predominantly upwards from the second whorl, but downwards from lower whorls. Movement occurred in a direct line above and below the labeled branch, with generally decreasing activities away from the branch in both directions. Therefore, the air CO_2 monitored by cellulose in the lower part of the Sitka stem is that in the lower, interior part of the crown, which is least accessible to mixing with the clean air above the canopy. The 13 to 28% range of x_b determined from our $\Delta_c{}^{14}C$ data is therefore compatible with the $\delta^{13}C$ results, especially when one considers that $\delta^{13}C$ is sensitive to the net biospheric carbon flux and $\Delta^{14}C$ to the total flux of decay CO_2.

6.2. *Stored photosynthate*

Maximum values for stored photosynthate x_s during 1962 and 1963 were calculated similarly to those for x_b. Weighted averages (again without 1963 increment #10) are $33.4 \pm 2.9\%$ and $31.8 \pm 1.3\%$, respectively. The 1964 data that indicate a minimum biospheric contribution

of 13% lower the weighted average x_s to $17.9 \pm 2.8\%$ and $14.2 \pm 1.3\%$ for 1962 and 1963 respectively. Consideration of the 1σ uncertainty intervals gives about 15% as the maximum x_s. Thus stored photosynthate of the previous year may make a 0 to 15% contribution to radial stem growth in our Sitka spruce. The calculated x_s values do not show a consistent trend over the growing season. Variations in x_s are most likely the result of the uncertainty in the data.

Our estimated contribution by stored photosynthate is much lower than the 80% to 90% estimated for a Western red cedar (*Thuja plicata* Donn.) by Fairhall and Young (1970). The good agreement between results listed in Table 2 suggests that either *Thuja plicata* has a different physiology, or that the ring dating of Fairhall and Young was off by one year. In the latter case, the Fairhall and Young $\Delta^{14}C$ values of $382 \pm 12‰$, $715 \pm 11‰$ and $895 \pm 12‰$ agree well with other results for 1962, 1963, and 1964 tree rings. The low contribution of stored photosynthate to radial stem growth in our 40-year old Sitka spruce confirms and expands the results of Schier (1970) for red pine seedlings. These showed little or no incorporation of ^{14}C into the cell walls of stem and branches during the early part of the growing season when the ^{14}C had been taken up in October of the previous year (Schier, 1970). Only after the initial phase of shoot and needle growth was completed was ^{14}C from earlier photosynthate incorporated into the stem.

6.3. Delayed incorporation

No data are available for the average delay in incorporation of photosynthate in stem cellulose for a 40-year old Sitka spruce. To study the importance of delayed incorporation we assumed an average delay by ten days. (This delay is probably unrealistically long judging from the results of Rangnekar et al. (1969) who found that during rapid growth incorporation of ^{14}C labeled carbon into stem tracheids of 15-year-old red pine continued for about 7 days after exposure.) For the x_b calculation we assumed canopy air followed the clean troposphere with about 0.69‰ per day increase during the 1962 growing season, 3.3‰ per day in 1963, and 1‰ per day increase at the beginning of the 1964 growing season changing to 1.3‰ per day decrease by its end. The new weighted averages calculated for the biospheric CO_2 contribution x_b are: $25.1 \pm 2.8\%$ and $22.2 \pm 1.3\%$ for the 1962 and 1963 maximum respectively, and $18.1 \pm 1.7\%$ for 1964. The range of possible biospheric CO_2 use with a 10-day delay in incorporation is thus about 13% to 23% compared with 13 to 28% without delay. Thus, the effect of a shorter, more realistic delay in incorporation would be an order of magnitude smaller than the effect of the use of biospheric CO_2. In view of the uncertainty in the data delayed incorporation may be neglected.

7. Conclusions

^{14}C concentrations in Sitka spruce stem-cellulose respond to changes in the atmospheric $^{14}CO_2$ concentration within 5 to 6 weeks. An important fraction of this delay seems attributable to the influence of biospheric decay CO_2. Thus trees, especially when growing in open conditions, are excellent monitors of the ^{14}C content of atmospheric CO_2 even on a sub-annual time scale. They preserve a detailed record of atmospheric $\Delta^{14}C$ during their growing season (April to September in coastal Washington).

Our cellulose $\Delta^{14}C$ is consistently lower than the corresponding $\Delta^{14}C$ of the clean troposphere. This is due mainly to the large difference in $\Delta^{14}C$ between "clean" (but bomb contaminated) air CO_2 and biospheric decay CO_2 during the period 1962–1964. Under natural conditions differences of only a few per mil are to be expected. The fraction of biospheric CO_2 incorporated in our Sitka spruce stem-celluse was in the range of 13% to 28%.

The use of stored photosynthate from the previous fall for radial stem growth is less than 15% (for 13% biospheric CO_2) and may be zero. The conclusion that there is little or no contribution of stored photosynthate to radial growth is consistent with ^{14}C relocation experiments on pine seedlings (Schier, 1970).

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