

RESEARCH ARTICLE

# Modeling the contribution of carbon sources using cellulose $^{14}\text{C}$ concentrations in subannual tree ring increments over the 1963 bomb spike

Helene L. Svarva<sup>1</sup> , Pieter M. Grootes<sup>1,2</sup> , Martin Seiler<sup>1</sup>  and Marie-Josée Nadeau<sup>1,3</sup> 

<sup>1</sup>The National Laboratory for Age Determination, NTNU University Museum, Norwegian University of Science and Technology, Sem Sælands vei 5, 7491 Trondheim, Norway, <sup>2</sup>Institute for Ecosystem Research, Christian-Albrechts Universität zu Kiel, Olshausenstrasse 75, D-24118 Kiel, Germany and <sup>3</sup>Institute of Physics – CSE, Silesian University of Technology, Konarskiego 22B, Gliwice 44-100, Poland

**Corresponding author:** Helene L. Svarva; Email: [helene.svarva@ntnu.no](mailto:helene.svarva@ntnu.no)

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## Abstract

Measurements of the radiocarbon ( $^{14}\text{C}$ ) content of subannual wood cellulose samples over the 1963 bomb spike have revealed an apparent delay between the increase in atmospheric radiocarbon content and that of wood cellulose. This delay is apparent in both coniferous and deciduous tree species and is of a magnitude of approximately 4 weeks. The delay in wood cellulose  $^{14}\text{C}$  change as measured in a Sitka spruce from Washington state, USA, was previously used to estimate the relative influence of tree physiological effects contra environmental effects. We repeated the measurements with the increased measurement precision of today's AMS systems and compare the new results to the ones of a Scots pine tree from Trondheim, central Norway and a white oak from Oregon state, USA. The results challenge the assumption that the  $^{14}\text{C}$  tree ring records directly show the atmospheric  $^{14}\text{C}$  concentration of a homogeneous, zonally well-mixed atmosphere. Instead, the apparent 1963 delay reflects local influences of the ecosystem and tree physiology. The 1963/1964 data allows for exploratory modeling of the effects of biospheric decay  $\text{CO}_2$  and local environmental influences assuming the absence of stored photosynthates from the previous year. Compared to the 10–30% contribution from biospheric  $\text{CO}_2$ , the effects of delayed incorporation of carbon into the wood cellulose and the effect of stored photosynthate are small in the conifers. Highly detailed  $^{14}\text{C}$  records of stem cellulose can, in combination with stable isotope studies, contribute to our understanding of variability of the local carbon cycle, climate, and the environment.

## Introduction

The radiocarbon ( $^{14}\text{C}$ ) content of tree rings is central to the reconstruction of atmospheric  $^{14}\text{C}$  concentrations, reaching from the present (post)-nuclear-testing-era (e.g., Hua et al. 2022) back to New Zealand Kauri trees around 42,000 years ago (Cooper et al. 2021). With the increased availability of high-precision AMS  $^{14}\text{C}$  measurements, the available tree-ring  $^{14}\text{C}$  database is rapidly expanding and the question regarding the relationship between the  $^{14}\text{C}$  content archived in the cellulose of tree rings and the  $^{14}\text{CO}_2$  content of the free atmosphere thus becomes increasingly important. A better understanding of how environmental information is recorded and archived by trees is also important for the interpretation of fluctuations in the relative abundances of stable isotopes ( $^2\text{H}/\text{H}$ ,  $^{13}\text{C}/^{12}\text{C}$ , and  $^{18}\text{O}/^{16}\text{O}$ ) and in width or density of tree rings in paleoclimatic studies (e.g., Cernusak and Ubierna 2022; McCarroll and Loader 2004; Zhang et al. 2016). The atmospheric testing of nuclear weapons in the 1950s and early 1960s has created a unique atmospheric  $^{14}\text{C}$  tracer signal that was archived globally in

trees growing under natural conditions and that now can be used to study the properties of tree rings as recorders of atmospheric and environmental information.

Grootes et al. (1989) used the large, rapid increase in atmospheric  $^{14}\text{CO}_2$  of 1963, caused by atmospheric testing of nuclear weapons, to show that  $^{14}\text{C}$  of stem cellulose of a Sitka spruce (*Picea sitchensis* [Bong.] Carr.) from the US Pacific coast followed the atmospheric  $^{14}\text{C}$  increase with an apparent delay of only 5 to 6 weeks. A rapid, but somewhat delayed, response was also apparent in a White oak (*Quercus alba* L.) from Oregon, USA (Cain et al. 2018), and in a Scots pine (*Pinus sylvestris* L.) from central Norway (Svarva et al. 2019). The difference between the increase in  $^{14}\text{C}$  content in the atmosphere and that in subannual wood cellulose samples, apparent in both evergreen (Grootes et al. 1989; Svarva et al. 2019) and deciduous (Cain et al. 2018) tree species, may have several causes: (1) a difference between the local free atmosphere and the zonal atmospheric reference record, (2) the use of older non-structural carbon (NSC) generated by earlier photosynthesis, and (3) photosynthesis (partly) taking place in the atmospheric boundary layer, which is influenced by photosynthesis, plant respiration, and by soil  $\text{CO}_2$  from the decay of soil organic matter, instead of in the free atmosphere. The latter two causes create a “memory” of prior changes in atmospheric  $^{14}\text{C}$  concentration in the tree ring cellulose that is also important for paleo-environmental studies, because it involves the tree and its local ecosystem.

The measurement of radiocarbon content in the rings of years 1952–1965 of a Trondheim Scots Pine at subannual resolution has been reported in Svarva et al. (2019). We repeated with improved precision the 1989 measurements of the Washington Sitka spruce on the original sample material at the National Laboratory for Age Determination, Trondheim, Norway. Our aim is to reanalyze the discrepancies between the atmospheric  $^{14}\text{C}$  increase for the years 1962–1964 and that of cellulose of subannual increments of the coniferous Sitka spruce and Scots pine as well as of the Oregon oak of Cain et al. (2018).

## Material and methods

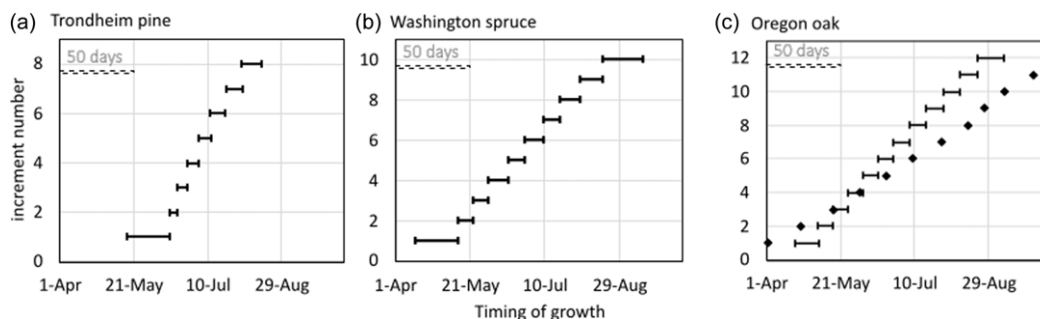
### Sample description

The samples used are from subannual sections of tree rings. For each tree, the rings from 1962, 1963, and 1964 were sliced into the largest practical number of subannual sections, depending on the width of the ring. The cutting precision is important for the results. Svarva et al. (2019) compared two cores of subannual increments from the same tree and found that difficulties in following the curved ring boundaries could lead to some material of the previous or next year being incorporated in the first or last increment. Each increment is then assigned to a growing period using a model of the local ring width growth curve.

**Trondheim pine:** The tree is a Scots pine (*Pinus sylvestris*) from Trøndelag, central Norway at 63°16'N, 10°27'E and at an elevation of 134 m a.s.l. The tree started its growth in the 1920s in relatively open canopy conditions. Eight increments were sampled from each ring and holocellulose was prepared from each increment. The sampling, assignment of increments to a growing period, pretreatment, and AMS measurements are detailed in Svarva et al. (2019).

**Washington spruce:** This Sitka spruce (*Picea sitchensis*) grew in Quillayute, Washington state, USA (47°57'N, 124°33'W). Ten increments were sampled from each ring and the sampling and assignment of increments to a growth period are described in Grootes et al. (1989). Surplus material from these ring increments has been stored since the 1980s. To reduce the measurement uncertainty, all increments, except the last increment of 1964 for which no material was left, were prepared to holocellulose and remeasured at the National Laboratory for Age Determination using methods described in Seiler et al. (2019) and Nadeau et al. (2015).

**Oregon oak:** The sampling, pretreatment, and AMS measurement of the Oregon white oak (*Quercus alba*) are described in Cain et al. (2018). Six to 12 increments were sampled from each ring and pretreated to holocellulose. The tree grew in western Oregon, USA (45°N), ca. 300 km south of the



**Figure 1.** Period of cellulose deposition per increment in 1963 tree ring using an S-shaped growth curve for the (a) Trondheim pine, (b) Washington spruce, and (c) Oregon oak, where the diamonds indicate the midpoint of the timing used in Cain et al (2018).

Washington spruce. The timing of the growth of the Oregon oak samples has been re-evaluated in our analysis from that of the original study (Cain et al. 2018). Instead of assuming a linear growth throughout the season based on ring mass, a polynomial, S-curve, growth function based on ring width was fitted to the estimates that were made for the Washington spruce. This changes the timing of the Oregon oak growth (Figure 1), especially in 1963, when ring growth now ends before the maximum of the bomb spike. Although this new timing estimate does not take into account the exact climate where the white oak grew, nor the species of tree, it does capture the changing rate of tree growth that occurs through a season.

### Sample preparation and AMS $^{14}\text{C}$ analysis of the Washington spruce remeasurements

Each subannual sample was pretreated with an adaptation of the BABAB (base-acid-base-acid-bleach) protocol (Khumalo et al. 2024; Němec et al. 2010; Seiler et al. 2019). First, oils, resin and waxes were extracted by treatment in petroleum ether, then the samples were cleaned at 75°C with steps of 4% NaOH, 4% HCl, 4% NaOH, and 4% HCl. A bleaching step with a mixture of 5%  $\text{NaClO}_2$  and 4% HCl at 75°C ( $\text{pH} \leq 4$ ), followed by ultrasonic bath at room temperature was applied for the holocellulose purification. The cellulose was then combusted in an elemental analyzer, and the  $\text{CO}_2$  was reduced to graphite with  $\text{H}_2$  gas over a Fe catalyst in an automated reduction system (Ohneiser 2006; Seiler et al. 2019).

The  $^{14}\text{C}/^{12}\text{C}$  and  $^{13}\text{C}/^{12}\text{C}$  ratios were measured in the HVE 1 MV AMS system at the National Laboratory for Age Determination in Trondheim (Nadeau et al. 2015). Radiocarbon results are reported as  $\Delta^{14}\text{C}$  after correction for the radioactive decay of samples and standard (Stuiver and Polach 1977).

The measurement uncertainties were calculated according to Nadeau and Grootes (2013). The measurements were normalized to the Oxalic Acid II primary standards (NIST SRM-4990C, Mann 1983). The samples were measured in five (5) different wheels containing 22 secondary standards of different  $^{14}\text{C}$  activities ranging from 15 to 104 pMC. The average ( $n=22$ ) of the normalized deviations from the canonical values, Z-scores, is  $-0.04 \sigma$  with a standard deviation of  $0.94 \sigma$  indicating that there is no systematic offset and that the quoted uncertainties are representative of the true uncertainties of the measurements. The average of these should be centred at zero and the width of the distribution should be 1 as it is in units of  $\sigma$ .

### Numerical model to determine different carbon contributions

The radiocarbon concentration of cellulose,  $\Delta_c^{14}\text{C}$ , is the result of contributions from clean air,  $\Delta_a^{14}\text{C}$ , input of  $\text{CO}_2$  produced by plant and soil respiration (biospheric decay  $\text{CO}_2$ ,  $\Delta_b^{14}\text{C}$ ), and stored

photosynthates,  $\Delta_s^{14}\text{C}$ . The contribution of the latter is complicated by a variable delay in the incorporation of stored carbon, present as non-structural carbon (NSC), into the cellulose. A numerical model to estimate the use of biospheric  $\text{CO}_2$  and stored photosynthate in trees is described in Grootes et al. (1989) (Equation 1).

Equation (1),  $^{14}\text{C}$  composition of cellulose from the different carbon sources involved (Grootes et al. (1989):

$$\Delta_c^{14}\text{C} = (1 - X_b - X_s)[\Delta_a^{14}\text{C} + (\delta\Delta_a^{14}\text{C}/\delta t)\Delta t] + X_b[\Delta_b^{14}\text{C} + (\delta\Delta_b^{14}\text{C}/\delta t)\Delta t] + X_s[\Delta_s^{14}\text{C}] \quad (1)$$

Where,

$X_b$  = Fraction of carbon contributed by biospheric decay  $\text{CO}_2$

$X_s$  = Fraction of carbon contributed by stored photosynthate

$\Delta_c^{14}\text{C}$  =  $^{14}\text{C}$  value of cellulose

$\Delta_a^{14}\text{C}$  =  $^{14}\text{C}$  value of atmosphere

$\Delta_b^{14}\text{C}$  =  $^{14}\text{C}$  value of biospheric decay  $\text{CO}_2$

$\Delta_s^{14}\text{C}$  =  $^{14}\text{C}$  value of stored photosynthate

$\Delta t$  = time interval of considered growth

$\delta\Delta/\delta t$  = rate of change in  $\Delta^{14}\text{C}$

While  $\Delta_c^{14}\text{C}$  can be measured directly, the accompanying values for  $\Delta_a^{14}\text{C}$ ,  $\Delta_b^{14}\text{C}$ , and  $\Delta_s^{14}\text{C}$  must be estimated and assumptions have to be made, as one of our goals is to estimate the contribution of biospheric carbon.

Atmospheric and biospheric contributions were evaluated and averaged over each increment. Since each tree-ring increment is evaluated separately, we set  $\delta\Delta/\delta t = 0$  which removes the time dependence, leading to Equation (2).

$$\Delta_c^{14}\text{C} = (1 - X_b - X_s)[\Delta_a^{14}\text{C}] + X_b[\Delta_b^{14}\text{C}] + X_s[\Delta_s^{14}\text{C}] \quad (2)$$

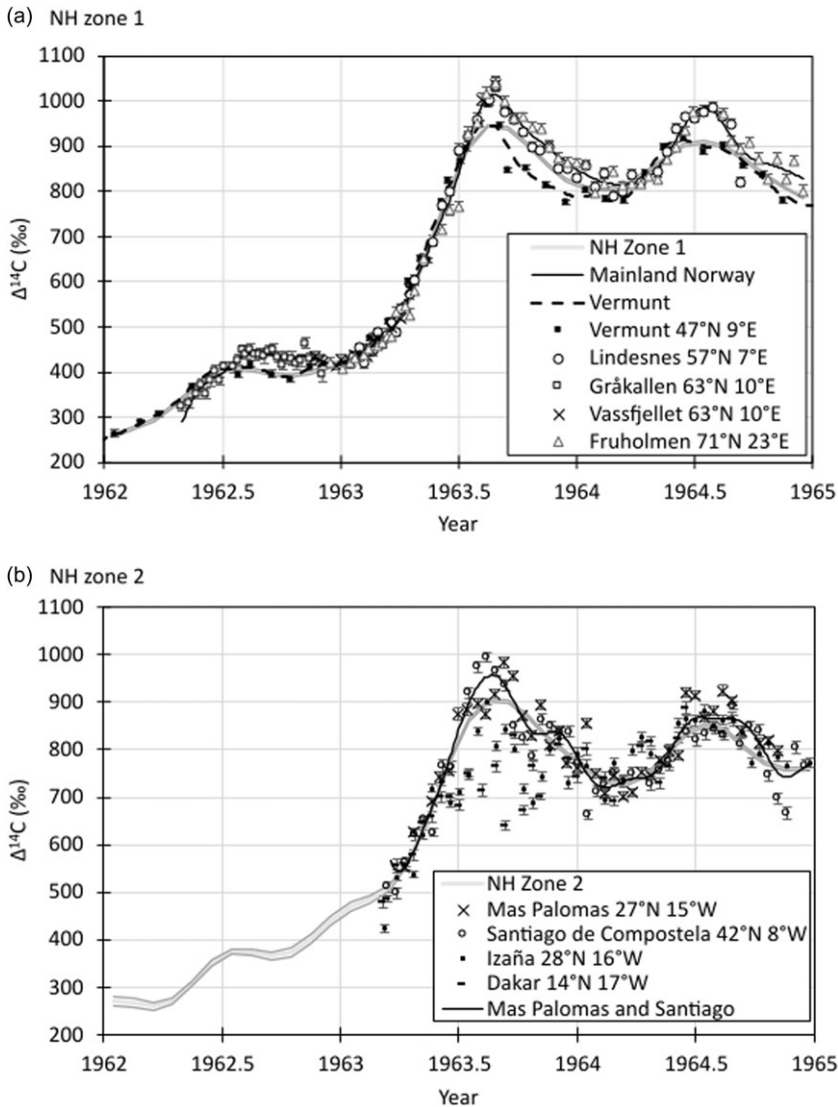
### Radiocarbon values of clean air: $\Delta_a^{14}\text{C}$

The Trondheim pine and the Washington spruce are both located in the Northern Hemisphere Zone 1, while the Oregon oak is in Northern Hemisphere Zone 2 (Hua et al. 2022).

The bomb calibration curves (Hua et al. 2013, 2022) include values from a variety of locations in each zone. Close examination of the records that make up the Northern Hemisphere Zone 1, reveals that atmospheric records from mainland Norway (Nydal and Løvseth 1996) have a higher  $^{14}\text{C}$  concentration during the bomb peak than the NH Zone 1 bomb curve (Figure 2a). This is based on data from Fruholmen (71°N), Gråkallen and Vassfjellet near Trondheim (63°N), and Lindesnes (57°N). In the peak of 1963, this difference is about 70 permille (Figure 2a). This indicates that the NH Zone 1 is not a homogeneous reservoir at the height of the bomb peak, 1962–1964, and that the lower  $^{14}\text{C}$  concentrations at the sites Vermunt (47°N; Levin et al. 1985; Levin and Kromer 2004) and Smilde (52°N; Vogel 1970) indicate incomplete meridional mixing and dilution of the Northern Hemisphere towards the south. To get the most accurate values  $\Delta_a^{14}\text{C}$  for the Trondheim pine at 63°N, we chose to combine the atmospheric records from mainland Norway.

For the Washington spruce (48°N), the Vermunt record (47°N) was deemed most suitable due to the small latitudinal differences (Grootes et al. 1989). This atmospheric record was further corrected for a  $2.9 \pm 2\%$  fossil fuel effect estimated to apply for the summer months (Ingeborg Levin pers. comm.). This correction is based on the added differences between Vermunt/Schauinsland (cf. Levin et al. 1985; Figure 2) and the cleaner site Jungfraujoch of 1.5‰ (May–August AD 1986–2016; Hammer and Levin 2017), and between Jungfraujoch and the even cleaner site Mace Head at the Irish coast, which is about 1.4‰ (May–August AD 2000–2016; Ingeborg Levin pers. comm.).

The Oregon oak (45°N) was placed in NH zone 2, based on the analysis of its subannual  $^{14}\text{C}$  profile by Cain et al. (2018; also Hua et al. 2022). We reanalyze the record with the revised timing of an



**Figure 2.** (a) Atmospheric records from NH zone 1; grey band: The Bomb NH zone 1 (Hua et al. 2022), dashed line: measurements from Vermunt in Austria (Levin et al. 1985; Levin and Kromer 2004) with correction for fossil fuel effect and its smoothed curve, solid line: Records from mainland Norway, i.e. Lindesnes, Fruholmen, Gråkallen and Vassfjellet (Nydal and Løvseth 1996), and their smoothed curve. (b) Atmospheric records from NH zone 2; grey band: The Bomb NH zone 2 (Hua et al. 2022), measurements from Mas Palomas, Santiago de Compostela, and Izaña in Spain and Dakar in Senegal (Nydal and Løvseth 1996), and solid line: the smoothed curve of the Mas Palomas and Santiago de Compostela records.

S-shaped tree-ring growth curve. The atmospheric samples included in the calibration curve compiled by Hua et al (2022) from zone 2 in 1963 and 1964 are from Santiago de Compostela (42°N), Izaña (28°N) and Mas Palomas (27°N) in Spain, and from Dakar (14°N) in Senegal (Nydal and Løvseth 1996). Because atmospheric samples are not available for 1962 from NH Zone 2, the curve is based only on tree-ring measurements in which the Oregon oak dataset is included. We, therefore, excluded 1962 from the analysis of the Oregon oak and use only the Santiago de Compostela and Mas Palomas datasets, which are significantly higher in  $^{14}\text{C}$  in 1963 than the Dakar and Izaña records (Figure 2b).

A smooth curve fit was made for each group of atmospheric records using the CCGCRV fitting procedure (Thoning et al. 1989; [www.esrl.noaa.gov/gmd/ccgg/mb/mb/crvfit/](http://www.esrl.noaa.gov/gmd/ccgg/mb/mb/crvfit/)) and uncertainties to the smoothed curves were assigned using a Monte Carlo technique (Turnbull et al. 2017).

### Radiocarbon values of biospheric decay CO<sub>2</sub>: $\Delta_b^{14}\text{C}$

CO<sub>2</sub> in air in the atmospheric boundary layer, especially air under a forest canopy, contains significant contributions from the decay of litter on the ground and the decay of soil organic matter and hair roots in the ground as well as from (root) respiration. These contributions are highly variable, being climate and ecosystem dependent, and normally difficult to quantify using <sup>14</sup>C, because their <sup>14</sup>C concentrations follow that of the atmosphere with various delays. Grootes et al. (1989) made a model for the lowland evergreen forest site of the Sitka Spruce on the Washington State Pacific coast. This model takes into account the estimated relative contributions and ages of litter such as needles, twigs, and vascular plants, and of root respiration assumed to come from recent photosynthate. Estimated  $\Delta^{14}\text{C}$  values are based on published compilations of atmospheric radiocarbon values. The ratio of decay and respiration is assumed to be constant. For simplicity we use the same values for all three trees as an approximation, the values are listed in Table 3b of Grootes et al. (1989) and in the supplemental data of this paper.

### Radiocarbon values of stored photosynthate (NSC): $\Delta_s^{14}\text{C}$

These values are difficult to quantify because they depend on the delay in fixing NSC carbon from the previous season into stem cellulose which depends on many factors, including tree physiology, phase of the growing season, climate, and ecosystem. The use of stored photosynthate is expected to be most relevant at the start of the growing season (e.g. Kromer et al. 2024 and references therein), especially in deciduous trees as seen in the results. As will be discussed in the next section, by assuming that the contribution from stored photosynthate is zero ( $X_s = 0$ ), we calculate the maximum and minimum possible values for the biospheric decay contribution based on our cellulose values in 1963 and 1964, respectively (Equation 3).

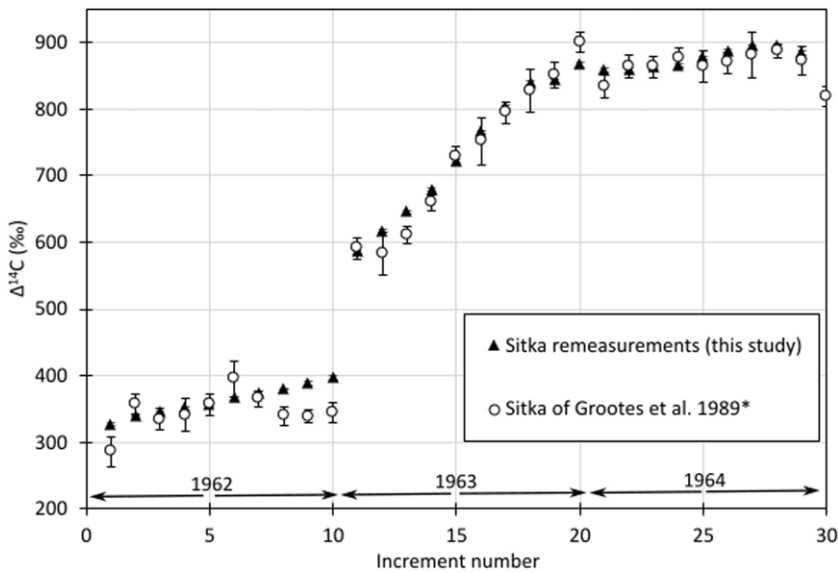
$$\Delta_c^{14}\text{C} = (1 - X_b)[\Delta_a^{14}\text{C}] + X_b[\Delta_b^{14}\text{C}] \quad (3)$$

## Results and discussion

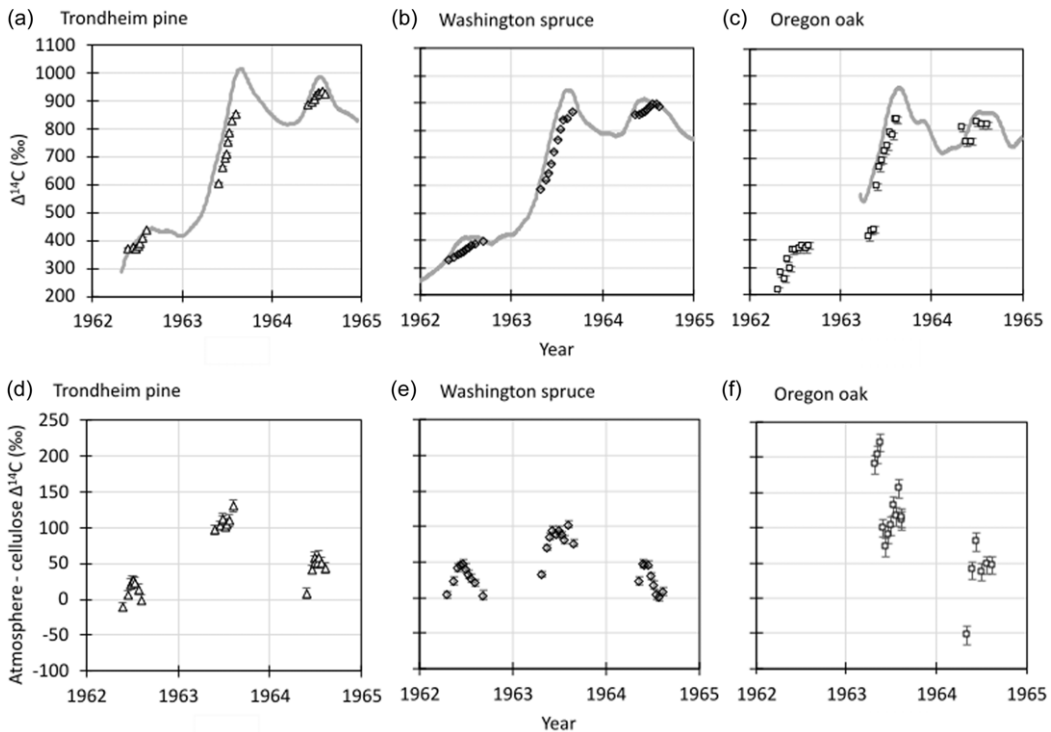
Because the radiocarbon values of 1989 of the Washington spruce were unavailable, an approximation of these values and their uncertainties was made by superimposing a Microsoft Excel graph onto an image of Figure 1 of Grootes et al. (1989). Remeasurement of the <sup>14</sup>C content of the subannual increments of the Washington spruce at NTNU verify the 1989 results with smaller uncertainties. Some differences are observed, especially in 1962 (Figure 3), when the remeasurement values scatter less than the original 1989 values and follow more closely the Vermont atmospheric radiocarbon increase. The differences between the original and repeat values are mostly not statistically significant except for the sections 1962 #8, 9, 10 and 1963 #3 and 10. The reduction in the scatter and uncertainty of the Washington spruce data set allows a closer comparison with the results obtained for the Trondheim pine and the Oregon oak.

Our best estimates of the <sup>14</sup>C level of the local free atmosphere are shown together with the tree ring <sup>14</sup>C values for the Trondheim pine, the Washington spruce and the Oregon oak in Figure 4a–c. The three trees show a striking increase in their stem cellulose <sup>14</sup>C concentration during the 1963 growing season. Plotted on their estimated growth curves, this increase approximately parallels the estimated <sup>14</sup>C increase of the local free atmosphere. An apparent delay of 5–6 weeks in the tree's response to the 1963 atmospheric radiocarbon increase has been described previously by Cain et al. (2018), Grootes et al. (1989), and Svarva et al. (2019). This response is reproduced here after remeasurements of the





**Figure 3.** The remeasured radiocarbon values of the subannual increments Washington Sitka spruce compared to the approximated 1989 values. \*The Washington spruce values from 1989 were obtained visually from Figure 1 of Grootes et al. (1989).



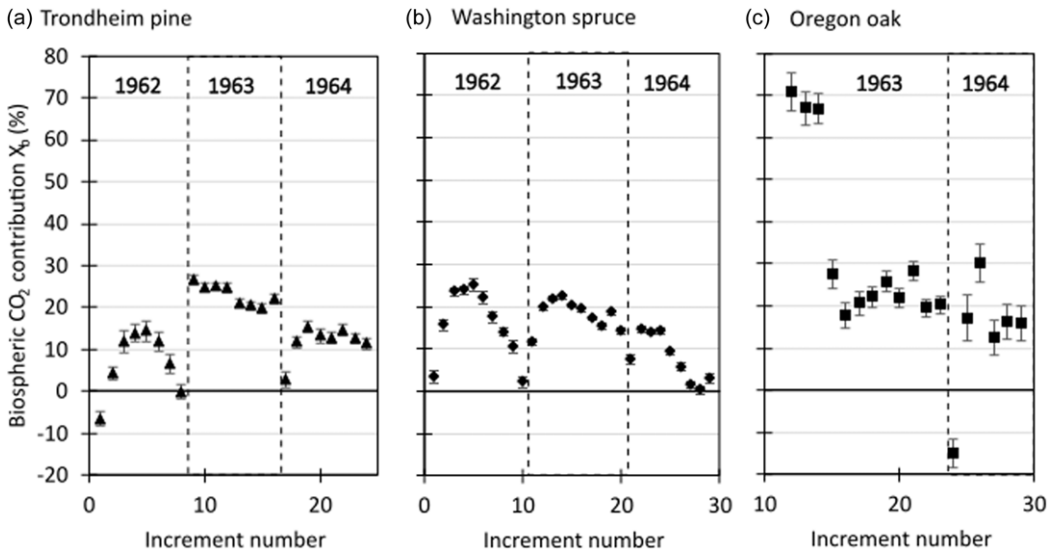
**Figure 4.** Radiocarbon values of tree cellulose and free atmosphere for (a) The Trondheim pine and mainland Norway atmosphere, (b) Washington spruce and Vermont atmosphere, (c) the Oregon oak and Mas Palomas and Santiago de Compostela atmosphere, and the difference between atmosphere and cellulose of the (d) Trondheim pine, (e) Washington spruce, and (f) Oregon oak.

Washington spruce and with a revised timing of the growth for the Oregon oak while using the corresponding local atmospheric  $^{14}\text{C}$  data for comparison (Figure 4a–c). The  $^{14}\text{C}$  increase in the tree cellulose in 1963 appears to be delayed by about 26 days in all three trees. Results of  $^{13}\text{CO}_2$  pulse-labeling experiments on an evergreen conifer branch by Kagawa et al. (2005) showed that the incorporation of photosynthesis carbon into cellulose happens within a few days both in the earlywood and in the latewood. Therefore, this apparent 1963 delay primarily reflects a lower  $^{14}\text{C}$  concentration of boundary layer  $\text{CO}_2$  due to the admixture of biospheric decay  $\text{CO}_2$  and plant respiration  $\text{CO}_2$  during the rapid input of stratospheric bomb  $^{14}\text{C}$  into the troposphere at mid northern latitudes during spring and summer of 1963. The newly obtained response is similar for the different trees and ecosystems and agrees with the values reported earlier (Grootes et al. 1989). For the Oregon oak, which is the only deciduous tree in this study, the first increments of each year deviate from the corresponding atmospheric values; those of 1962 and especially of 1963 being significantly lower in  $^{14}\text{C}$  than the atmosphere and the first increment of 1964 significantly higher than the atmospheric record of Mas Palomas and Santiago de Compostela (Figure 4c). Similar deviations were reported by Kromer et al. (2024) for an oak from the Upper Rhine Valley. We interpret this as the use of NSC from the end of previous growing season at the onset of radial growth, before the time of leaf formation, in this deciduous tree. Olsson and Possnert (1992) observed similar differences between the  $^{14}\text{C}$  concentration of earlywood in an Uppsala oak (60°N) and that of the clean air at Abisko (68°N), northern Sweden, with earlywood  $^{14}\text{C}$  significantly below the atmosphere in 1963 and above in 1964 (c.f. their Fig. 4, our Figure 4c). Yet, they reported no clear evidence for a delay between the cellulose and the atmosphere on a time scale of weeks. This may be due to the large  $^{14}\text{C}$  measurement uncertainty of ca. 20 ‰ and to uncertainty in their timing of the latewood growth increments. A memory effect in the earlywood of the oak was mentioned in Olsson and Possnert (1992). A comparable earlywood effect is not obvious in the evergreen conifers (Figure 4a,b).

The differences between the  $^{14}\text{C}$  concentration of the local free atmosphere and the measured cellulose  $^{14}\text{C}$  concentrations for the years 1962–1964 (Figure 4d–f) are significant compared with data scatter and uncertainty and invite further interpretation. The assumptions of Eq. (1) ascribe these differences to the influence of respiration of the local vegetation and soil,  $\Delta_b^{14}\text{C}$ , on the local boundary layer atmosphere plus the delay between the photosynthetic fixation of carbon from this atmosphere at  $\Delta_s^{14}\text{C}$  and its incorporation into stem cellulose. The differences between the atmosphere and cellulose  $\Delta^{14}\text{C}$  values in 1963 range from 98 to 131‰ for the Trondheim pine, from 33 to 96‰ for the Washington spruce, and from 71 to 220‰ for the Oregon oak (Figure 4d–f). In 1964, the differences range from 9 to 60‰ for the Trondheim pine, from 1 to 48‰ for the Washington spruce, and from –53 to 80‰ for the Oregon oak. In 1962, only the differences for the Trondheim pine, –10 to 26‰, and the Washington spruce, 4 to 48‰, are available. These significant differences reflect carbon from the bomb spike entering the pool of decaying organic matter and changing  $\Delta_b$  and  $\Delta_s$  of Eq. (1). During the rapid increase in atmospheric  $^{14}\text{C}$  of 1962 and 1963, biospheric decay  $\text{CO}_2$  and stored photosynthate contained less  $^{14}\text{C}$  than the atmosphere. By contrast, photosynthate from late in the 1963 growing season was higher in  $^{14}\text{C}$  than in the beginning of the 1964 growing season, as can be seen in the first increment of the Oregon oak.

We estimate from the measured  $\Delta_c^{14}\text{C}$  values of 1962 and 1963 the maximum contribution of biospheric decay  $\text{CO}_2$  to stem cellulose,  $X_b$ , under the simplifying assumption that once the growing season and tree photosynthesis is in full swing, fresh photosynthate will be used for ring growth and the use of older NSC can be neglected ( $X_s = 0$ , Figure 5). The potentially stored photosynthate, produced in the autumn of 1963, has a higher  $\Delta^{14}\text{C}$  value than 1964 cellulose, while the rate of change in  $\Delta^{14}\text{C}$  in 1964 is small. Then, again assuming  $X_s = 0$  yields the minimum amount of biospheric decay  $\text{CO}_2$  that is needed to produce the observed lowering of cellulose  $\Delta^{14}\text{C}$  values compared to atmosphere. Figure 5 indicates a maximum contribution of 25–30% C from biospheric decay carbon to the cellulose of all three trees for most of the 1963 ring and for the Washington spruce for 1962. This agrees with the maximum contribution of  $\approx 28\%$  estimated by Grootes et al (1989). For 1964, all three trees suggest a minimum carbon contribution from biospheric decay of 10–15%.





**Figure 5.** The modeled contribution from biospheric CO<sub>2</sub>,  $X_b$  (%) to each tree-ring increment for (a) the Trondheim pine, (b) Washington spruce, and (c) the Oregon oak. Uncertainties are based on the cellulose radiocarbon measurements. For the calculation, the contribution from stored photosynthate,  $X_s$ , is assumed to be zero.

The Trondheim pine, Figure 5a, indicates a lower value of 10 to 15% decay CO<sub>2</sub> for 1962, possibly reflecting more open growing conditions and colder winters. The mean temperature for April–August was  $\approx 2^\circ\text{C}$  warmer in 1963 than in 1962 and 1964 (Norwegian Meteorological Institute, station code: 68860). This could explain the flatter shape of the  $X_b$  values of the Trondheim pine in 1963 as decay might have started earlier in the year. The Trondheim pine increment #1 of 1962 gives a negative  $X_b$  value. This is likely due to cutting error as described in the supplemental data of Svarva et al. (2019).

The coniferous Scots pine and Sitka spruce show a close  $\Delta^{14}\text{C}$  agreement between atmosphere and cellulose for the first increments of 1962 and 1964 (Figure 4d,e, Figure 5a,b) indicating a small  $X_b$  and  $X_s$ . A low influence of decay CO<sub>2</sub> may reflect low soil temperatures leading to low soil activity early in the season. Negligible use of stored photosynthate of the preceding fall and winter for early stem growth in evergreens agrees with results reported by Schier (1970) for red pine seedlings.

The  $\approx 70\%$  biospheric decay contribution of the Oregon oak, calculated for the first three increments of the 1963, and the negative value for the first increment of 1964, indicate that the assumption  $X_s = 0$  was wrong and stored NSC must have been used. Warmer winter temperatures at this site compared to Trondheim might cause less fluctuations in biospheric decay through the year.

## Conclusions

Remeasurement of the Washington spruce has confirmed the earlier results. The shift between atmospheric and cellulose  $^{14}\text{C}$  contents in 1963 is consistent between the two evergreens and the deciduous oak. This reflects the response of the tree to rapid changes of atmospheric  $^{14}\text{C}$  content. The combined 1963/1964 data indicate a lower tree-ring cellulose  $^{14}\text{C}$  concentration in 1963 due to a biospheric C contribution of the order of 10 to 30 %. The estimated influence of soil CO<sub>2</sub> in 1962 and 1964 is similar. The actual contribution will vary with local conditions like density of tree stand, thickness and quality of forest floor organic cover, local climate, etc. It is likely that this interpretation can be transferred generally to NH sites and evergreen as well as deciduous taxa.

Analyses of this detail require careful consideration of many factors, including (1) the timing of tree growth, (2) the biospheric  $^{14}\text{C}$  values, and (3) the atmospheric  $^{14}\text{C}$  values representing clean air. The detailed analysis reveals that stem cellulose provides an archive of the fluctuations in atmospheric  $^{14}\text{C}$  concentration during the growing season with subannual to weekly resolution, largely independent of tree species. This tree cellulose  $^{14}\text{C}$  record differs, however, from that of the zonally defined free atmosphere due to (1) zonal  $^{14}\text{C}$  mixing inhomogeneities in the atmosphere, (2) modification of the local atmosphere in which photosynthesis takes place by local ecosystem activity, and (3) plant physiology varying the delay between photosynthesis and fixation of the photosynthate carbon in stem cellulose. The  $^{14}\text{C}$  differences, here highlighted by the bomb  $^{14}\text{C}$  spike, are always present, reflecting the flow of stratospheric  $^{14}\text{C}$  through carbon cycle pathways in troposphere, biosphere, and oceans. With increased availability of  $\Delta^{14}\text{C}$  data with 1 to 2‰ uncertainty, the assumption that  $^{14}\text{C}$  tree ring records directly show the atmospheric  $^{14}\text{C}$  concentration of a homogeneous, zonally well-mixed atmosphere, must be reassessed. The information from highly detailed  $^{14}\text{C}$  records on carbon contributions to stem cellulose can, in combination with stable isotope studies, contribute to our understanding of variability of the local carbon cycle, climate, and the environment.

**Supplementary material.** To view supplementary material for this article, please visit <https://doi.org/10.1017/rdc.2025.10102>

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