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IMPORTANCE OF BIOSPHERIC CO_2 IN A SUBCANOPY ATMOSPHERE DEDUCED FROM ^{14}C AMS MEASUREMENTS

P M GROOTES*,**, G W FARWELL*, F H SCHMIDT*, D D LEACH*
and
MINZE STUIVER**

ABSTRACT. ^{14}C concentrations in the stem cellulose of a Sitka spruce from the Pacific coast of Washington respond to changes in atmospheric $^{14}CO_2$ concentration within 5–6 weeks. $\Delta^{14}C$ values for cellulose were consistently lower than those of the corresponding clean troposphere during rapid increase in atmospheric ^{14}C caused by nuclear weapons tests (1962–64). Possible reasons for this include: 1) a delay of days or weeks in incorporation of recent photosynthate, 2) the use of stored photosynthate, and 3) photo-assimilation of biospheric decay CO_2. We estimate that the influence of process 1 is small or negligible. The respective contributions to the total carbon deposited as radial stem growth in our Sitka spruce then are 2) < 15% (possibly 0), and 3) 10%–23% (13%–28% if the possible effect of root respiration is included in the biosphere decay component). We plan to test this concept by looking for a vertical ^{14}C gradient in the 1963 growth ring of a tree located in a dense forest canopy; we do not expect to find such a gradient in a similar tree from a strongly wind-washed location.

INTRODUCTION

We used the University of Washington accelerator mass spectrometry (AMS) facility to obtain a detailed radial profile for the ^{14}C concentration in the growth rings for 1962, 1963, and 1964 of a Sitka spruce tree (*Picea sitchensis*). The tree grew in the rain forest on the Olympic Mountain slopes near the Pacific Ocean, and was a part of a forest canopy (a circumstance crucial to the conclusions we have drawn).

The period 1962–1964 was one of large and rapid increase in atmospheric ^{14}C, due to nuclear weapons tests. During these "bomb spike" years, the ^{14}C content of the atmosphere nearly doubled, and the increase during 1963 was especially dramatic.

By measuring ^{14}C in ten sequential segments for each annual growth ring, we observed a close parallel between the ^{14}C concentration of tree cellulose and that of the atmosphere, with a time resolution of 1–2 weeks (Farwell *et al*, 1984; Grootes *et al*, 1986).

Originally this was a sort of exercise to sharpen our AMS techniques and to add a new dimension, a "fine structure" in time, to tree-ring radiocarbon analysis. In preparing the data for publication, however, one of us (PMG) delved into plant physiology and in doing so drew some interesting conclusions about the important role that biogenic CO_2 plays in the formation of tree-stem cellulose. A brief account is given below; more detailed arguments are presented elsewhere (Grootes *et al,* 1989).

*Nuclear Physics Laboratory and Department of Physics
**Quaternary Isotope Laboratory, Department of Geological Sciences

PROCEDURE AND RESULTS

In 1962, our tree was 40 years old, and it was harvested in January 1977. Our sample was located near the base of the tree, well under the surrounding forest canopy. The three annual rings, each 10–13mm wide, were each split into ten equal radial growth increments; the samples prepared from these were analyzed by AMS by measuring ^{14}C against ^{13}C.

Calendar assignments for the growth segments were derived from what we believe to be a very reasonable translation of data from the measured growth curve for another conifer, a Douglas fir, growing nearby in a similar environment. We estimate an uncertainty of ± 5 days for the timing of the inferred growth pattern.

We converted the measured ^{14}C concentrations into a plot of ^{14}C concentration in tree cellulose as a function of time (Fig 1). Also shown in Figure 1 are atmospheric $\Delta^{14}C$ values at Vermunt, Austria (Levin *et al,* 1985), which are the most relevant available atmospheric ^{14}C data for that period. Vermunt has essentially the same latitude as our tree (ca 47°N *vs* ca

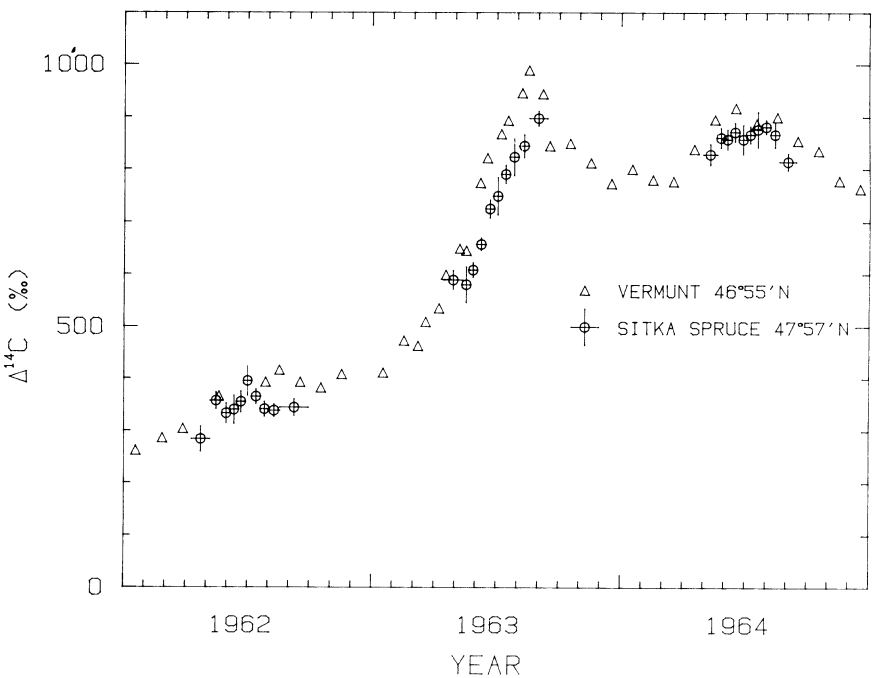

Fig 1. Comparison of $\Delta^{14}C$ in tree-stem cellulose of Sitka spruce (Olympic peninsula, Pacific coast, Washington) with that of the atmosphere (Vermunt, Austria) (Levin *et al*, 1985). The Sitka spruce growth site and the Vermunt location have almost identical latitudes and their atmospheres should have experienced very similar nuclear-weapons-induced increases in $^{14}CO_2$ between 1962–1964.

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48°N) and its atmosphere should have responded similarly each spring to the large injection of bomb-produced ^{14}C from the stratosphere into the troposphere. (A small positive correction (ca 5‰) was made for the local European fossil fuel CO_2 input, and another for biospheric contamination (5–19‰).) Our inferences and conclusions about cellulose formation are based upon a detailed comparison of the ^{14}C data for stem cellulose and for the (corrected) Vermunt atmosphere.

DISCUSSION AND CONCLUSIONS

Since the growing season for Sitka spruce is roughly April through September, our $\Delta^{14}C$ calendar sequence for cellulose shows a significant gap each year. A dramatic increase in $\Delta^{14}C$ is recorded during 1963 for both cellulose and the atmosphere; we note that while $\Delta^{14}C$ peaks in late 1963 and is somewhat lower in 1964, the average over the growing season is lower in 1963 than in 1964. Thus, single whole growth rings for 1964 should (and do) have the higher ^{14}C concentration, as is well known.

During 1963, when $\Delta^{14}C$ was changing most rapidly, the tree-ring values seem to follow those of the corrected Vermunt atmosphere with an apparent delay of 5–6 weeks. For 1962 and 1964, we might better describe the differences as a deficiency in $\Delta^{14}C$ for tree-stem cellulose; this could apply to 1963 as well. So, is it a delay or a deficiency, or both? And, if there is a deficiency, what is the cause?

The deficiencies that we wish to explain ($\Delta^{14}C$ in stem cellulose *vs* $\Delta^{14}C$ in clean tropospheric air) are estimated from our data at 51±5‰ (1962), 137±7‰ (1963) and 62±6‰ (1964). They most likely occurred as the result of the use, for radial stem growth, of carbon that had not been in full exchange with the troposphere for some time. The difference is largest in 1963, when the rapid rise in atmospheric ^{14}C evidently created a strong disequilibrium between the "clean" tropospheric air and the air under the forest canopy.

Several possibilities include: 1) a delay of days or weeks in the incorporation of current photosynthate, 2) the use of stored photosynthate from the previous growing season and 3) the use of CO_2 from biospheric decomposition of material on and in the forest floor.

Delayed Incorporation. We conclude that this can only cause a small fraction of the observed difference. Several investigators, watching for the incorporation in stem cellulose of ^{14}C from enriched CO_2 placed around the needles of young pine trees, have established that, during rapid growth, the incorporation of ^{14}C in stem cellulose is essentially complete after 4–7 days (see eg, Rangnekar, Forward & Nolan, 1969; Balatinecz, Forward & Bidwell, 1966). The apparent 5–6 week delay for 1963 must be due largely to other causes.

Stored Photosynthate and Photosynthate from Biospheric Decomposition CO_2. To determine the contributions of these two carbon sources to the observed deficiency, we need to know their ^{14}C concentrations. Since no direct measurements at the relevant time and place were made, these have to be estimated independently. The estimate for decomposition CO_2 is based on the composition of the forest litter, the age of the different components of the forest floor, including soil organics and fine roots, and the

respective decay rates of these components. We estimate $\Delta^{14}C$ for decomposition CO_2 near our Sitka spruce at ca +165‰ (1962), +251‰ (1963) and +492‰ (1964) (Grootes *et al,* 1989).

We assume that stored photosynthate used in a given growth season is produced during the previous fall and has an appropriate $\Delta^{14}C$ for that period. These assumptions and the $\Delta^{14}C$ data for 1963 and 1964 allow us to estimate respective contributions to the total carbon incorporated as cellulose in radial stem growth.

First, consider cellulose formed in 1963. During 1963, the tropospheric ^{14}C concentration increased rapidly, and both decomposition CO_2 and stored photosynthate from fall 1962 had a ^{14}C concentration below those of the troposphere and the stem cellulose. Thus, for 1963, any combination of these two could give the observed ^{14}C deficiency in cellulose. To get the maximum for one, we assume zero for the other. If, for 1963, no stored photosynthate is used, the amount of decomposition CO_2 required is ca 23% (*ie,* 23% of all CO_2 converted to cellulose in the lower tree stem). This represents the maximum amount for decomposition CO_2. Similarly, assuming that no decomposition CO_2 is used, we find ca 33% for the maximum amount of stored photosynthate that could have been used.

Next, consider cellulose formed in 1964. We recall that the tropospheric $\Delta^{14}C$ values peaked in August 1963. Thus, photosynthate produced in fall 1963, but not converted to cellulose until 1964, will have a high ^{14}C concentration, comparable to that of the troposphere in 1964. Thus, during 1964, only decomposition CO_2 could cause the observed cellulose ^{14}C deficiency, which leads to an estimate of the minimum contribution of biospheric decomposition CO_2: 10% of the total cellulose carbon.

But if we go back to 1963 and assume that the minimum amount of decomposition CO_2 (10%) is used in that year, we find that, at most, 15% of stored photosynthate is required, and the amount could be as low as zero.

We conclude, then, that the observed ^{14}C deficiency in tree-stem cellulose (or the apparent 5 to 6 weeks' delay of incorporation during 1963) is caused 1) *not* by actual delay of incorporation beyond a very few days, 2) possibly, in part, by the use of stored photosynthate, but not above 15% of the total carbon assimilated and 3) by use of CO_2 from biospheric decomposition, forming probably 10–23% of the total cellulose carbon. If the possible effect of root respiration of CO_2 is included in the biospheric component, the same maximum of 15% is placed on stored photosynthate, but the total biospheric CO_2 contribution now ranges from 13%–28%.

IMPLICATIONS; A VERTICAL ^{14}C GRADIENT?

We have postulated (and believe we have evidence to support) the presence of a significant amount of biogenic CO_2 under the forest canopy and surrounding our Sitka spruce. If this is so, our scenario predicts a vertical ^{14}C gradient in the air under the canopy and, if there are branches with photosynthesizing leaves at many heights, in the stem of such a deep-forest tree itself: 1) limited mixing in the biospheric-atmospheric boundary layer creates different CO_2 microclimates at different levels of the tree, 2) limited mixing within the tree stem of photosynthate from different branches then

leads to vertical ^{14}C gradients in the stem cellulose. We should look first for such a vertical gradient in the 1963 ring, where the disequilibrium between tropospheric and biospheric ^{14}C concentrations is most severe.

On the other hand, there should *not* be a vertical ^{14}C gradient in an isolated, wind-washed tree, perhaps located on the Pacific coast or an offshore island, which would experience virtually pure Pacific Ocean air, at all levels, from prevailing westerly winds.

Several investigators (Grootes *et al,* 1989) have observed increases of ^{13}C concentration in tree leaves with increasing height above ground and attribute such gradients, in part, to soil-respired CO_2 (depleted in ^{13}C relative to the atmosphere). Others attribute them primarily or wholly to differences in isotopic fractionation (reduced fractionation near the top, where the sunlight is more intense); if this is correct, we should not observe a vertical ^{14}C gradient in either of the trees we plan to measure, since our $\Delta^{14}C$ results are adjusted for the observed $\delta^{13}C$ and any fractionation effects are thereby eliminated.

These experiments will also elucidate the source of photosynthate incorporated in growth of the lower stem. If, as some believe, carbon incorporated in stem growth represents an assimilation-weighted average over the whole crown, no vertical ^{14}C gradient will be observed.

SIGNIFICANCE

The present study strongly indicates that recycled biogenic CO_2 makes a significant contribution to the carbon in the cellulose of the lower tree stem. The possible uptake of "local" CO_2 and incorporation of resulting "local" photosynthate into tree-stem cellulose without complete mixing within the tree are important complications, especially in paleoenvironmental tree-ring isotope studies.

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Editor's note: Together with the authors, the editor regrets that it is not possible to reproduce here the color photographs of the Sitka spruce in its natural settings.

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