

Annual radiocarbon measurements in a century-old European beech tree (*Fagus sylvatica*) from coastal northeastern North America

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ABSTRACT

Radiocarbon (¹⁴C) concentrations of annual tree rings from an European beech tree (*Fagus sylvatica*) from Woods Hole, MA, USA were analyzed at National Ocean Sciences Accelerator Mass Spectrometry facility (NOSAMS) to construct a ¹⁴C bomb curve record from northeastern North America. The ¹⁴C concentration rises from a pre-bomb (1895–1955) average of Fraction Modern (F¹⁴C) = 0.9764 to a peak of 1.8639 in 1964. After 1964, F¹⁴C gradually decreases to 1.0611 in 2014. The annual tree-ring radiocarbon content agrees with the atmospheric radiocarbon content of the Northern Hemisphere and is very similar to the radiocarbon concentration of a red oak (*Quercus rubra*) tree located in Bear Mountain State Park in New York, USA. Intra-annual tree-ring pairs did not produce evidence of a seasonal effect on the radiocarbon concentration, but there were few samples and more study is warranted.

1. Introduction

Atmospheric testing of nuclear weapons during the 1950s and early 1960s nearly doubled the concentration of ¹⁴C in the atmosphere between 1955 and 1963. Since the enactment of the Nuclear Test Ban treaty banning atmospheric testing of nuclear weapons in 1963, the excess of atmospheric ¹⁴C has decreased due to oceanic and terrestrial uptake, fossil-fuel dilution, atmospheric mixing, and radioactive decay [39,32,31,20,9,12]. The abrupt rise and steady fall in atmospheric ¹⁴C, known as the radiocarbon “bomb curve”, has been well-documented [38,27,19,30,26,15,2,9,12]. Since 1971, bomb radiocarbon reached equilibrium in the atmosphere [27]. Seasonal cycles are also observed in atmospheric ¹⁴C concentrations. In the Northern Hemisphere, seasonal cycles are thought to be explained by three processes: the transport of fossil fuel emissions to the surface which are most pronounced in winter and spring, the transport of ¹⁴C-enriched air from the stratosphere to the surface in summer and fall, and ¹⁴C enrichment of the atmosphere from the terrestrial biosphere in the summer and fall [32,20,9]. Levin [20] concludes that the strongest ¹⁴C seasonal cycle in the Northern Hemisphere is dominated by the fossil fuel component.

Trees provide excellent records of the history of atmospheric ¹⁴C because they incorporate atmospheric carbon into the cellulose of

annual rings, are often formed annually in multiple global temperate regions, and the rings can be counted directly to determine the year of growth [35]. The atmospheric radiocarbon concentration peak due to nuclear weapons testing occurred in late 1963 [27,12], after the growth cycle of northern hemispheric deciduous trees was complete [14,22]. This resulted in a one year offset in peak northern hemisphere ¹⁴C concentrations recorded in tree rings [4,14,11,22,31]. Annual tree rings can be used to construct calibration curves [12,33]. These calibration curves can be used to convert the measured ¹⁴C age to determine a corresponding range of calendar years [43].

European Beech (*Fagus sylvatica*) are diffuse-porous trees which produce annual tree rings during a spring (post budburst) to late summer/early fall growing season in temperate climates. The average growing season is late April/early May to late August/September depending on climatic conditions [40,17,24,41]. In the spring, the cells of the new growth, or earlywood (EW), have thin walls and appear lighter in color. Growth outside of spring/early summer conditions produces thickened cell walls that are reduced in internal volume and produce a distinct band of darker cells referred to as latewood (LW) [36].

Few annually resolved radiocarbon tree-ring records exist for northeastern North America. Records from two red oak (*Quercus rubra*) trees in New York [4] show radiocarbon concentrations rising in 1955,

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Fig. 1a. Location of sampled *Fagus sylvatica*- Woods Hole, MA, USA.

peaking in 1964, and then falling until the end of the record. As defined before, this decrease is due to ocean and biosphere uptake, fossil fuel dilution, and radioactive decay [39,20,9,10]. The *Quercus rubra* tree from Central Park in New York City, NY shows a larger effect of fossil-fuel burning in a more urban area (larger Suess-effect and less intense radiocarbon concentration peak in the bomb curve) compared to the *Quercus rubra* from Bear Mountain State Park. A radiocarbon record from annual growth bands from a mollusc shell (*Arctica islandica*), from Georges Bank in the Gulf of Maine off the northern United States coast shows a delay in the radiocarbon bomb peak relative to the atmospheric peak due to the air-sea isotope exchange rates and North Atlantic circulation [45]. The first observed increase in radiocarbon concentrations occurs in 1956 and peaks around 1972. The record from this shell is in phase with two low-latitude North Atlantic banded corals from Florida and Bermuda [8], but with a smaller amplitude and more depleted ^{14}C values.

In this study we analyze the ^{14}C concentrations of annual tree rings from a *Fagus sylvatica* tree to provide another radiocarbon bomb curve record from northeastern North America. The data from this study are compared to other radiocarbon tree-ring records in the area. We also investigate if there is any influence of the increased fossil-fuel burning during the winter months and its effects on the radiocarbon content of EW compared to LW.

2. Methods

2.1. Study site

In April of 2015 one of four *Fagus sylvatica* trees was removed from the Woods Hole Oceanographic Institution's Village Campus, Woods Hole, Massachusetts, USA (41.5°70.7' N, 70.7° 40.4' W, Fig. 1a) due to its decline in health from bark disease. It is estimated this tree was

planted around 1870 on the Fay estate after deforestation of the area for farming and shipbuilding [46]. The *F. sylvatica* was an overstory tree in a suburban area with limited light and resource competition (Fig. 1b). This *F. sylvatica* tree grew < 500 m from the shoreline at an elevation of roughly 10 m above sea level. Woods Hole is located in the Northern Hemisphere temperate zone. Regional mean temperature and precipitation are 4.6 °C and 140 mm from October to March and 16 °C and 104 mm from April to September, respectively [25]. The site may have a large marine influence due to its proximity to the ocean. Northwest winds dominate in the winter and Southwest in the summer in this area [34]. Though there is no major industry in Woods Hole, air mass CO_2 content is influenced by urban centers and industrial activities to the West [18].

2.2. Sample preparation

A complete cross-section of the *F. sylvatica* tree was obtained for this study and brought to NOSAMS for an undergraduate student project. The section chosen for this project was 2.5 m above the base of the tree due to disease and rot in the lower trunk. Approximately 2 mm of material was removed from the surface of the cross-section with an electric wood planer to remove contamination. The exposed surface was then sanded with progressively finer grits (from 200 to 600 grit) to better resolve the annual growth rings visually. Annual rings were counted from youngest to oldest growth where ring 1 represents the youngest ring of the *F. sylvatica* tree. We assigned the calendar year 2014 to ring 1 because the tree was cut down prior to budburst. Rings were counted on two additional cleaned radial transects of the trunk to provide an intra-tree check of the ring counts.

A preliminary set of samples (every 10 rings) was collected and analyzed to verify the location of the bomb curve prior to annual sampling. These samples were collected by driving a cleaned 5 mm



Fig. 1b. A view of the European Beech (*Fagus sylvatica*) tree prior to removal from Woods Hole Oceanographic Institution's village campus in 2015.

outside diameter increment borer into the surface of the tree disk. The section found to contain bomb radiocarbon was sampled annually (rings representing years 1952–1974). The annual samples were milled from the cleaned surface of the tree disk using cleaned single edge razor blades and drill bits. Sampling produced fine particles, so further homogenization was not necessary. Sub-annual, or EW and LW, pairs were collected using the same method, and three of these pairs were analyzed (calendar years 1960, 1961, and 1982). Darker LW was differentiated from lighter EW visually (Fig. 2).

All samples were weighed into glass centrifuge tubes and pretreated using a modified Acid-Base-Acid (ABA) method [7,23] to remove non-structural carbon (primarily sugars and starch). All glassware in this study was baked at 550 °C for 1 h prior to use. The samples were first acidified with 10% v/v HCl for 1 h at 60 °C. The samples were rinsed once with Milli-Q water and then soaked in 2% by weight NaOH solution for 1 h at 60 °C. This step was repeated until the NaOH solution was visibly colorless. The samples were rinsed once with Milli-Q water and received a final acidification with 10% v/v HCl for 1 h at 60 °C. Once the final acid step was complete the samples were rinsed with Milli-Q water to pH 7, placed in pre-combusted petri-dishes, and dried overnight at 60 °C in a drying oven.

Approximately 2 mg (~1 mg of carbon) of each treated sample was weighed into a 5x9 mm tin capsule. The samples were combusted using an Elemental vario EL Cube elemental analyzer. The resulting CO₂ gas was cryogenically separated from the helium carrier gas stream [3] and transferred to a graphite reaction tube. Using the zinc reduction method [47] the resultant CO₂ was reduced to graphite in a sealed pyrex tube containing Fe, Zn, and TiH₂ powders. The sealed tubes were then baked

at 500 °C for 3 h and 550 °C for 4 h to promote graphite formation [47,3].

Graphite was stored in the sealed tubes until pressing into aluminum targets and analysis on the USAMS instrument (3 MV Tandemtron) at NOSAMS [44,21]. Samples were normalized to OX-I [28], and radiocarbon-free acetanilide (J.T. Baker, A068-03) was used for blank correction. Corrections for stable isotope fractionation were made using the on-line AMS data. Radiocarbon data are reported as Fraction Modern (F¹⁴C) as defined by Stuiver and Polach [37] and Reimer et al. [33].

3. Results

The samples produced a time series from 1895 to 2014 for the *F. sylvatica* tree from Woods Hole, MA (Table 1). The pre-bomb (1895–1955) average F¹⁴C is 0.9793. All subsequent errors are given as one-sigma analytical error. Samples were not run in duplicate, but we estimate reproducibility of ABA treated wood to be 3–4‰ from performance of internal reference materials. The earliest elevated radiocarbon due to atomic testing occurred in 1956 and peaked in 1964 (F¹⁴C = 1.8650 ± 0.0064). After 1964 it decreased to F¹⁴C = 1.0611 ± 0.0032 in the most recent ring (2014). We observe an increase in radiocarbon concentration for the 1971 ring that is likely due to the French and Chinese nuclear tests which occurred from 1966 to 1970 [27]. The data set from the *F. sylvatica* tree was compared to atmospheric ¹⁴C data from CALIBomb Northern Hemisphere Zone 1 (Fig. 3), though it is noted that Cape Cod is on the Northern Hemisphere Zone 1 and Zone 2 boundary [12]. The radiocarbon concentration peak

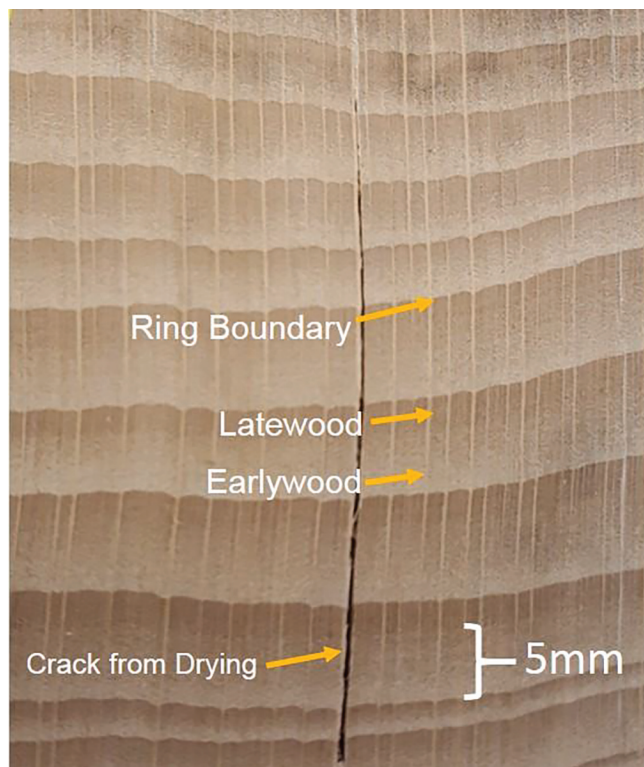


Fig. 2. A close up view of tree rings of the *Fagus sylvatica* tree from Woods Hole, MA, USA. This shows approximately 9 rings with the lighter earlywood and darker latewood transitions. Ray cells are also visible perpendicular to the ring boundaries.

for this *F. sylvatica* tree is in 1964, following a consistent pattern of other temperate northern hemisphere deciduous trees [4,14,11,22,31]. Intra-annual sample pairs of EW and LW from the 1960, 1961 and 1982 rings were analyzed to assess annual variability in assimilated carbon (Table 2, Fig. 3). The difference in radiocarbon content of early and late season annual growth is more than 2 sigma in all pairs, but the differences do not follow a consistent pattern.

4. Discussion

The radiocarbon bomb curve recorded in the radiocarbon concentrations in the tree rings of this coastal *F. sylvatica* tree is very similar in timing and amplitude to the CALIBomb Northern Hemisphere zone 1 atmospheric data [12], Fig. 3). Data for the *F. sylvatica* tree are also compared to two red oak (*Quercus rubra*) trees from New York [4] and a mollusc shell (*Arctica islandica*) from local waters- Georges Bank in the Gulf of Maine [45], Fig. 4). The *F. sylvatica* tree shows similar changes in assimilated radiocarbon to the *Q. rubra* tree from Bear Mountain State park in New York [4]. Our data from Woods Hole are consistent with assimilation of carbon representing the atmosphere with no strong input from carbon with a reduced ^{14}C signal, either from CO_2 from fossil fuel combustion as seen in the *Q. rubra* from New York City or CO_2 from surface ocean radiocarbon as recorded by the *A. islandica* shell from the Gulf of Maine.

Using repeated counts on two cleaned radial transects on the trunk cross-section, we estimate our ring count to be accurate to within 1

Table 1

Summary of Fraction Modern (F^{14}C) results from the *Fagus sylvatica* tree in Woods Hole, MA, USA. Errors are reported in one standard deviation.

Ring	Year	F^{14}C
119	1895	0.9882 ± 0.0021
110	1904	0.9865 ± 0.0030
100	1914	0.9755 ± 0.0020
90	1924	0.9763 ± 0.0022
80	1934	0.9941 ± 0.0026
70	1944	0.9810 ± 0.0031
62	1952	0.9691 ± 0.0025
61	1953	0.9740 ± 0.0021
60	1954	0.9751 ± 0.0021
59	1955	0.9730 ± 0.0022
58	1956	0.9971 ± 0.0023
57	1957	1.0340 ± 0.0032
56	1958	1.0731 ± 0.0025
55	1959	1.1324 ± 0.0024
54	1960	1.2238 ± 0.0027
53	1961	1.2584 ± 0.0028
52	1962	1.4114 ± 0.0036
51	1963	1.7774 ± 0.0044
50	1964	1.8639 ± 0.0064
49	1965	1.7923 ± 0.0052
48	1966	1.6845 ± 0.0055
47	1967	1.6328 ± 0.0035
46	1968	1.5692 ± 0.0050
45	1969	1.5610 ± 0.0033
44	1970	1.5184 ± 0.0032
43	1971	1.5305 ± 0.0034
42	1972	1.4640 ± 0.0053
40	1974	1.4037 ± 0.0039
39	1975	1.3918 ± 0.0029
38	1976	1.3600 ± 0.0030
36	1978	1.3266 ± 0.0030
34	1980	1.2798 ± 0.0030
30	1984	1.2113 ± 0.0030
29	1985	1.1998 ± 0.0029
18	1996	1.1207 ± 0.0032
1	2014	1.0611 ± 0.0032

ring. Another potential source of error is the difficulty of sampling the youngest growth ring due to its small size ($< 0.4 \text{ mm}$). In this study, “Ring 1” (calendar year 2014) was collected by boring axially into the outer surface of the cross-section in the initial sampling. In this section of the tree the average ring width for the first 10 rings is 0.45 mm . This first sample may comprise more than one year of growth.

There may also be uncertainty due to the choice of a relatively simple Acid-Base-Acid pretreatment method. Prior work indicates that lignins may not be separated from the cellulose in ABA treatments, and that samples treated thus may be more susceptible to contamination than pure cellulose samples [4,42,13,1]. However, data from Olsson and Possner [29], and Quarta et al. [31] found the difference between the ^{14}C concentration in the insoluble fraction and the cellulose fraction to be minor. Capano et al. [5] looked at different pretreatment of wood and found that most methods, including ABA and cellulose extraction give consistent and reproducible results for most samples, but that aged and radiocarbon-free wood may give more accurate results with cellulose extraction. ABA was chosen as a compromise for its simplicity and close agreement with cellulose extraction in most modern dendrochronological applications.

The data for the EW and LW pairs do not show a consistent trend. Large rings ($> 4 \text{ mm}$ wide) were chosen for EW-LW pair sampling to

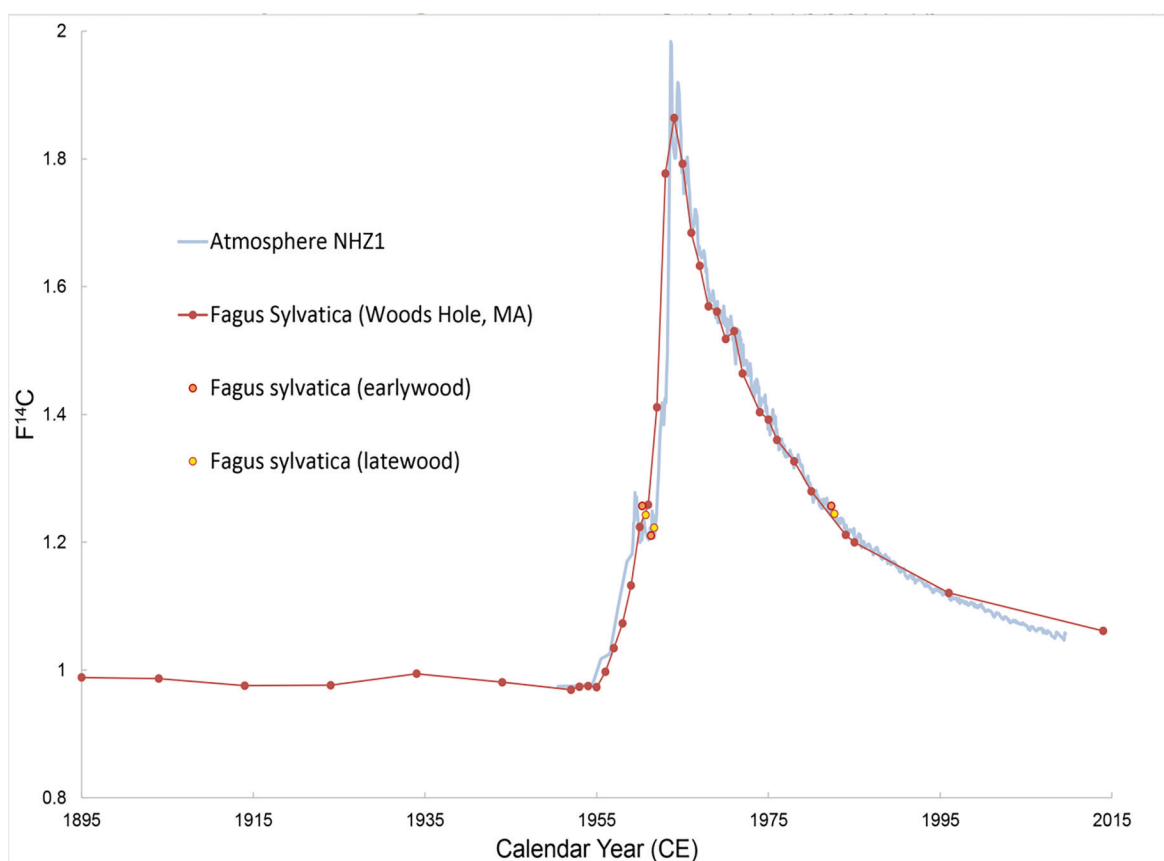


Fig. 3. $F^{14}C$ vs. calendar year for the *Fagus sylvatica* tree in Woods Hole, MA, USA. CaliBOMB Northern Hemisphere Zone 1 atmospheric data [12] are shown for comparison. Line Width = average $F^{14}C$ analytical error of ± 0.003 . For the earlywood and latewood 0.3 and 0.7 years (respectively) were added to the calendar year to represent the approximate time of development (late April to early May for earlywood and late August to early September for latewood).

help improve sample separation, however the boundary is somewhat arbitrary and is based on light vs. dark coloration. There is a suggestion that the ^{14}C detected in EW and LW may follow the bomb curve signal, i.e. in the rising limb of the bomb signal, LW has higher ^{14}C than EW, while in the post-bomb period LW has lower ^{14}C than EW. These pairs show no clear reduction in early season radiocarbon due to dilution by dead CO_2 from fossil-fuel combustion during the heating season (October through April). There is some uncertainty associated with the formation of EW in trees. Carbone et al. [6] found that nonstructural carbon used to support metabolism was one to two years old in spring before leaves emerged, but reflected current-year photosynthetic

products in late summer. Sample preparation may also affect data from EW-LW pairs. As discussed, ABA pretreatment may not properly leach younger lignin from latewood and therefore may produce inconclusive results, but Kudsk et al. [16] produced good results for EW-LW pairs using a bleach-acid-base-acid pretreatment. Only three *F. Sylvatica* EW-LW pairs were analyzed for this work. Analysis of more pairs (especially in pairs after bomb radiocarbon reached equilibrium) would improve interpretation of any differences with the pairs.

5. Conclusion

A new bomb-curve record from northeastern North America was produced by measuring the radiocarbon concentrations of annual tree rings from a *Fagus sylvatica* tree in Woods Hole, MA, USA. The results are consistent with published tree-ring data sets and Northern Hemisphere atmospheric ^{14}C data. There is little evidence for local effects of fossil fuels on the radiocarbon isotopic concentration of EW when compared to LW in the same growing year. This data set supports the NH1 model published in CALIBomb, and may add to the accuracy and robustness of that calibration dataset.

Table 2

Fraction Modern ($F^{14}C$) results from earlywood and latewood of three rings from *Fagus sylvatica* in Woods Hole, MA, USA.

Ring	Year	Growth	$F^{14}C$
54	1960	Early	1.2569 ± 0.0029
		Late	1.2429 ± 0.0028
53	1961	Early	1.2105 ± 0.0028
		Late	1.2230 ± 0.0029
32	1982	Early	1.2569 ± 0.0030
		Late	1.2444 ± 0.0027

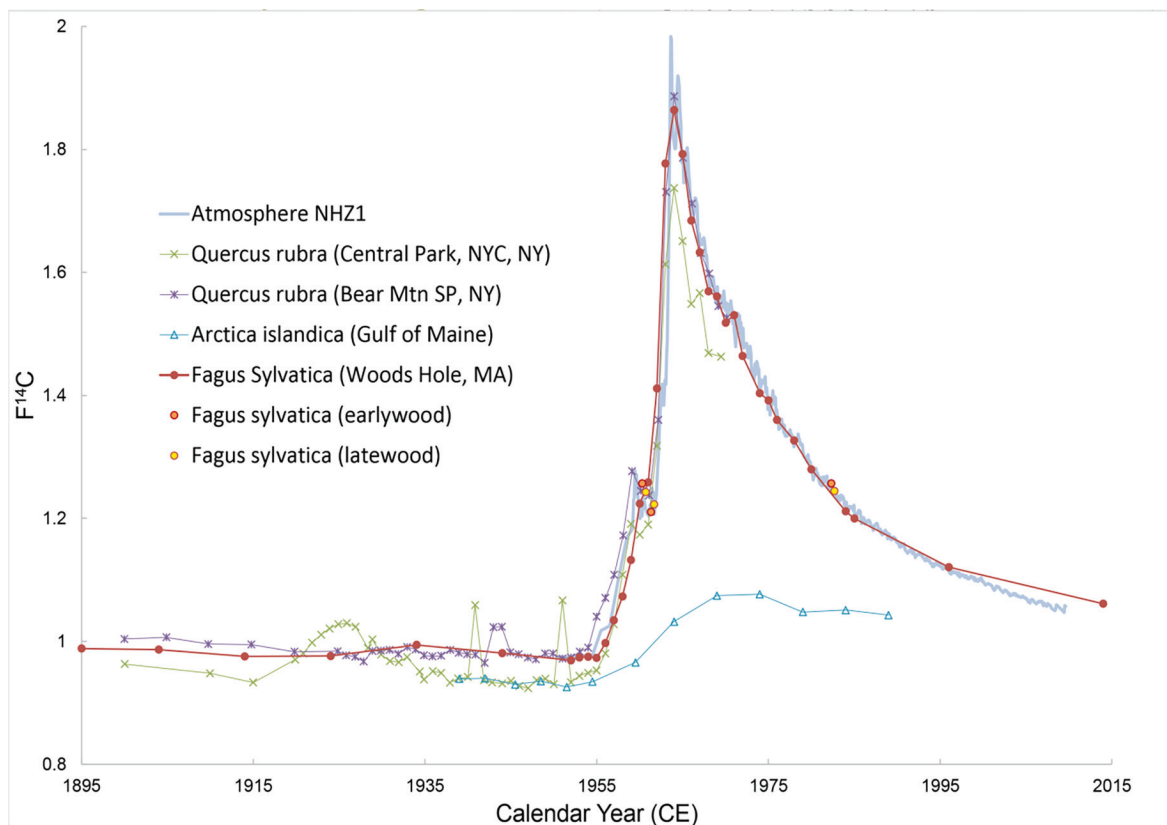


Fig. 4. $F^{14}C$ vs. calendar year from the *Fagus sylvatica* tree are compared to atmospheric ^{14}C data from BombCal NH zone 1 [12], two red oak (*Quercus rubra*) tree ring sets from New York [4] and a mollusc shell (*Arctica Islandica*) chronology from Georges Bank [45].

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