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Research paper

Annual growth rings in a sample of Paraná pine (*Araucaria angustifolia*): Toward improving the ^{14}C calibration curve for the Southern Hemisphere

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ABSTRACT

This work reports the first high-precision ^{14}C-AMS dating of holocellulose from a Brazilian subtropical species for the period 1927–1997, with the goal to identify suitable Southern American tree species that will serve as benchmarks for improving the calibration of the ^{14}C time-scale for the Southern Hemisphere (SH). The tree rings analyzed here came from a single tree of Paraná pine (*Araucaria angustifolia*) growing at 22°50′S, 46°04′W (Camanducaia, Minas Gerais, Brazil). A slight depletion of atmospheric ^{14}C after 1927 AD was observed, due to the Suess effect. Our ^{14}C results also showed the rise and rapid decrease of atmospheric ^{14}C concentrations associated with the detonation of nuclear weapons during the late 50s, and its subsequent uptake by other large C sinks. Current ^{14}C data can be used for the study of the global carbon cycle, forensic sciences applications, and the determination of the age and growth rate of tropical trees without annual ring patterns. The remarkable overall agreement of our tree-ring/^{14}C data with the SH Zone 1–2 compilation dataset shows this subtropical tree species' potential to refine the ^{14}C calibration curve.

1. Introduction

Conversion of radiocarbon (^{14}C) values to calendar dates is normally accomplished by calibration using independent curves built up from materials of pre-determined ages and techniques that can identify these ages. Conventionally, ^{14}C calibration curves have been based on pairing datasets of ^{14}C measurements of tree rings predated by dendrochronology. But a series of other types of samples (corals, planktonic foraminifera, speleothems, varved sediments, tropospheric CO_2 samples) and techniques (U–Th dating and varve counting) also have been employed (Reimer et al., 2009). Existing ^{14}C calibration curves, which can help to infer absolute calendar ages, are divided into hemispheres as well as pre- and post-nuclear bomb values. For pre-bomb values, the ^{14}C calibration curve of the Northern Hemisphere runs from 0 to 50 ka cal BP, with much less resolution beyond 26 ka cal BP (Reimer et al., 2009). The ^{14}C time scale for the Southern Hemisphere (SH) spanning from 0 to 11 ka cal BP (SHCal02) comprises measurements of ^{14}C and dendrochronology methodologies from decadal wood samples growing between AD 950 and 1850 (McCormac et al., 2002; Hogg et al., 2002). Beyond this dataset, the SH curve was initially expanded based on the Northern Hemisphere dataset and a random effects model (McCormac et al., 2004), which took into account a variable ^{14}C offset between the Northern and Southern Hemisphere datasets. For post-bomb ^{14}C values (1950–2010), in addition to the differences between the hemispheres, the ^{14}C calibration curves are divided into different zones (1–3) based upon modeled atmospheric circulation (Hua et al., 2013 and references therein).

Recently, the SH ^{14}C time scale curve has been extended to 50 ka cal yr BP, with the addition of new tree-ring/^{14}C values (SHCal13) curve (Hogg et al., 2013), and assuming an inter-hemispheric offset similar to those measured for the past 0–2000 cal BP. Nevertheless, a South American ^{14}C curve from dendrochronologically-dated wood is still lacking, especially

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within the tropical or subtropical zones, which should experience seasonal shifts in atmospheric CO_2. The first step toward improving SHCal curve calibration is to assess the annual makeup of the growth rings of long-lived tree species. This goal can be achieved by ^{14}C bomb-pulse dating of selected individual dendrochronologically-dated rings. Although certain efforts have been proposed to expand the SH calibration of the ^{14}C time scale, the lack of rings and/or false rings in many tropical tree species make it challenging to accurately model ^{14}C dates (Worbes, 2002). Previous attempts to ^{14}C measure selected tree ring sequences of a subtropical Brazilian Paraná pine tree species (*Araucaria angustifolia*) by scintillation counting confirmed its overall atmospheric nuclear bomb-pulse profile (Lisi et al., 2001), suggesting that it may be promising to use annual resolution data from this long-lived species to reconstruct a ^{14}C calibration curve.

The above-ground testing of atomic bombs during the 1950s and 1960s produced a large amount of atmospheric ^{14}C, creating a pulse that today can be used as a time-specific signal, especially from new plant growth tissue. Since this anthropogenic variation in atmospheric ^{14}C concentration has been well-documented from the early 1950s onward in both hemispheres, bomb-produced ^{14}C can be a valuable tool in age validation of dendrochronologically-dated, long-lived tree species. Consequently, tree species can be assessed by tree-ring/^{14}C cross-dating, and thereby can be screened as possible candidates to fulfill temporal gaps from existing datasets in order to improve the accuracy of existing ^{14}C atmospheric records of ^{14}C time-scales.

In the present study, we revisited the selected tree rings of the *A. angustifolia* tree growing at 22°50′S, 46°04′W (13 tree rings previously measured by scintillation counting in Lisi et al., 2001) and expanded this dataset to 71 wood rings (i.e., the complete cross-section from the pith to inner bark layer, comprising the years from 1927 to 1997). Although *A. angustifolia* seasonal cambium activity has been confirmed (Oliveira et al., 2009) and its master chronology has been obtained by dendrochronology techniques (Oliveira et al., 2010), Lisi et al. (2001) produced only the overall ^{14}C layout of the bomb pulse by the scintillation counting method. Therefore, longitudinal variations in atmospheric ^{14}C concentration cannot be fully disregarded. In addition, previous ^{14}C analyses using *A. angustifolia* at 27′S, 50′W (1960–1969) have been judged to be erroneous, due either to the presence of carbohydrate reserves from the previous year in the tree-ring analyzed and/or lack of a proper chemical pretreatment before making ^{14}C measurements (Hua et al., 1999). Therefore, *A. angustifolia* seasonal cambial activity as well as its future use toward improving the ^{14}C calibration curve for the SH still need to be properly confirmed. Tree-ring/^{14}C cross-dating analysis has been demonstrated to be a solid and concrete approach for validating or refuting the annual pattern of tree species (Biondi and Fessenden, 1999; Hua et al., 1999; Fichtler et al., 2003; Menezes et al., 2003; Biondi et al., 2007; Hua, 2009; Wils et al., 2009; Soliz-Gamboa et al., 2011). Here, we chose to use high-precision ^{14}C-AMS (Accelerator Mass Spectrometry) dating of holocellulose extracts of consecutive single tree rings of a single specimen within the bomb pulse period to:

a) Validate the annual makeup of the growth rings of this long-lived tree species by dating material from the pre-to post-bomb periods;
b) Build a chronology for a terrestrial archive in South America that is both reliable and relatively recent, particularly at this latitude; and
c) Demonstrate the robustness of our holocellulose procedure by testing it on wood samples that belong to the bomb pulse period. For this purpose, we made an extra effort to measure the entire ARA-07 consecutive single tree rings (71 tree rings) rather than a few selected rings, as per Lisi et al. (2001).

Once the annual growth pattern of Araucariaceae is confirmed, we can use dendrochronological sequences of these South American evergreen cone-bearing trees to refine the existing ^{14}C time scale for the Southern Hemisphere.

2. The Paraná pine (*A. angustifolia*) tree species and its potential for use in ^{14}C calibration

The Paraná pine, also called the candelabra tree (*A. angustifolia*), is included in the Araucariaceae family. This ancient family of evergreen coniferous trees, which during the Jurassic and Cretaceous periods was distributed worldwide, became confined to New Caledonia, Norfolk Island, eastern Australia, New Guinea, Argentina, Chile, and southern Brazil (Bittencourt, 2007; Ledru and Stevenson, 2012). Among the 19 Southern Hemisphere species of Araucaria, *A. angustifolia* is the only one in the Brazilian territory. It can be also found in the northeast of Argentina (Misiones and Corrientes) and in Paraguay (Alto Paraná). The majestic *A. angustifolia* tree, like its cousin *A. araucana*, found in Argentina and Chile, can reach a maximum height of 50 m and a diameter of 2 m at breast height. Growth ring counting shows that this tree species can reach several hundred years of age (ca. 400 years; Oliveira et al., 2010), making these trees ideal for long-term studies. In Brazil, this subtropical tree species (Fig. 1) occurs primarily at elevations between 500 and 1400 m in southern Brazil (between 24° and 30° S) (Bittencourt, 2007), and in a few isolated patches at elevations between 1400 and 1800 m in the southeastern region (between 18° and 24° S) (Behling et al., 2004; Tomazello Filho, 1986).

Few studies have addressed the Brazilian Araucariaceae tree species in terms of cambial activity, seasonality signals, and their potential to produce chronologies. At high elevations within the Paraná state (25° 34′S, 50° 05′W; 780 m a.s.l), Seitz and Kanninen (1989) developed a ring-width chronology using 10 trees of *A. angustifolia*. In Minas Gerais (the studied area in this work; see Fig. 1). Lisi et al. (2001) reproduced the overall outline of the ^{14}C bomb-pulse, whose ^{14}C patterns were reflected in the tree's annual ring formation. Oliveira et al. (2009) confirmed the seasonal growth patterns of *A. angustifolia* when studying 18 trees growing in an old-growth forest stand located between 27 and 31°S at 900 m. a.s.l. Recently, Stepka (2012) uncovered *A. angustifolia* trees as old as 264 years in Santa Catarina State. Therefore, this tree species has already proven to be of value for dendrochronological analyses, and possibly for tree-ring/^{14}C cross-dating. Thus, the tree's use as a starting point toward improving the accuracy of existing ^{14}C atmospheric records of the SH ^{14}C time-scale is encouraging. Obviously, chronologies derived from living trees are not enough to produce ^{14}C datasets for calibration curves. The extension of tree-ring/^{14}C records also relies on using wood specimens found elsewhere (e.g., in historic buildings, or in wood that was somehow preserved on the forest floor or in bogs). Once a fragmented-floating tree-ring width chronology is established and matched with an existing tree-ring chronology, the ^{14}C calibration curve can be determined.

Before European settlements established themselves in the southern and southeastern Brazilian regions during the late 17th and 18th centuries, the araucaria forest covered an area of approximately 200,000 km^2 (Fig. 1). Due to a long history of the tree's exploitation for timber, fruit, and seeds for animal and human consumption, and in forest clearance for agricultural activities, it is believed that just 2%–4% of the original araucaria forest remains standing today (Mantovani et al., 2004). Despite its present

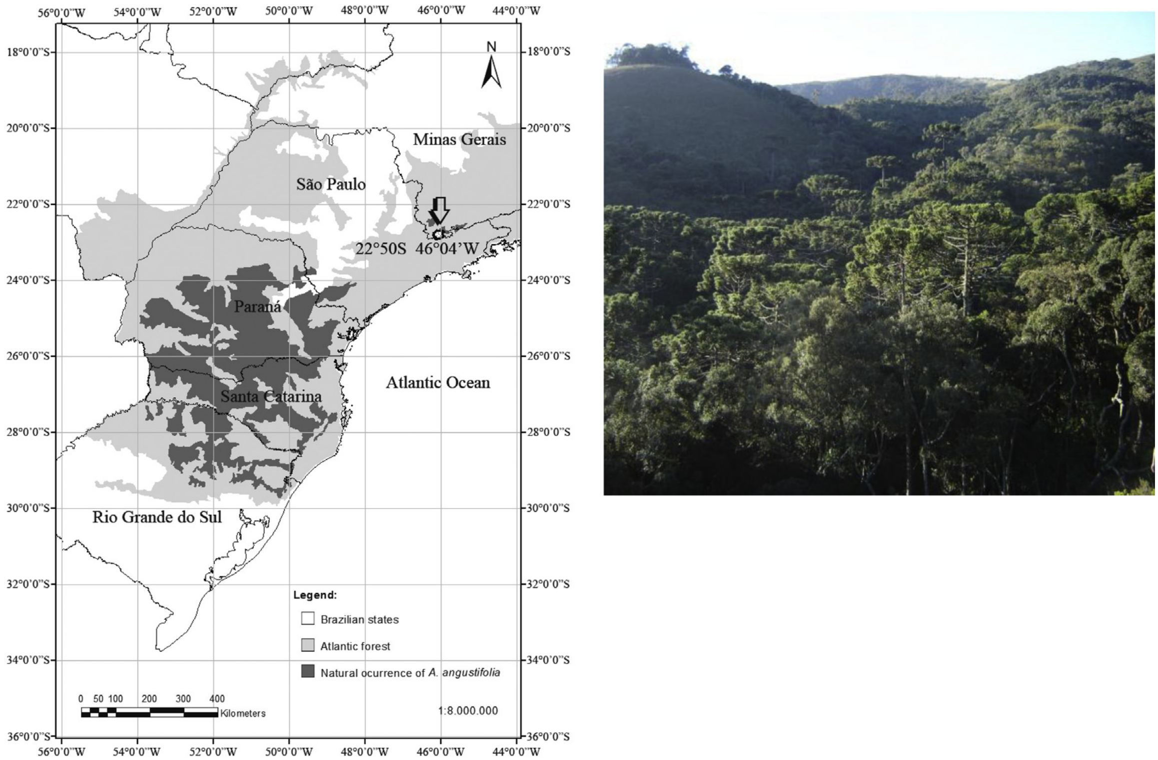

Fig. 1. Map showing primitive araucaria forest distribution in the Brazilian territory (http://www.rbma.org.br/anuario/mata_03_anosdedesttuicao_dest_araucaria.asp Source: CN-RBMA: Projeto Inventário dos Recursos Florestais da Mata Atlântica), with the location of the present study site (marked by an arrow). Aerial photo of the mixed Ombrophilous Forest of Camanducaia, Minas Gerais, showing patches of native trees of *A. angustifolia*.

fragmentation, some of its original timber (which was heavily used for public buildings, such as chapels, schools, and community centers, as well as households and farms) has been preserved within those edifices in the states of Minas Gerais (Cruz, 2008), Santa Catarina (Brandt, 2008, 2010), Paraná and Rio Grande do Sul (Obrzut, 2006; Batista, 2007; Kohlraush, 2007; Zani, 2013), and São Paulo (Morales et al., 2011), with or without other groups of wood from native tree species. The significance of the araucaria timber for Brazil's economy, and especially for the development of the southern regions, motivated the documentation of many colonial architectural structures found today in this region (Zani, 2013) (Fig. 2). Although the future of some of these structures remains unclear unless effective plans for preserving them can be implemented, their old timbers may also help to establish dendrochronological libraries, which can later be synchronized with ^{14}C atmospheric signatures for this latitude.

3. Geographical setting and sample details

The study area is the Levantina Farm unit of the Melhoramentos Florestal S.A., a paper and wood products company located in the Atlantic forest biome in the Municipality of Camanducaia (22°50′S, 46°04′W), Minas Gerais, in Brazil's southeastern highlands (Fig. 1). Its nearly 12 thousand hectares, which consist of mostly pines for local paper cellulose production, include patches of mixed Ombrophilous Forest with native trees of *A. angustifolia*, which still dominate the upper canopy in the valleys and slopes (Fig. 1). The region is characterized by an average annual temperature of 18 °C with precipitation of 1660 mm/year, which is measured locally at the meteorological station. Precipitation events are higher during the summer (between December and April) and are coupled with longer periods of sunlight. The drier seasons range from April to August, with occasional frost observed during the coolest winter months.

The tree rings analyzed here by ^{14}C-AMS analysis were from a single native *A. angustifolia* tree (ARA-07) with a growth unit age estimated as >45 years, growing at an altitude of 1300 m. The ARA-07 tree was selected from among 21 other native tree specimens (Lisi, 2000), and cut down in 1998. From a cross section of wood disk of approximately 10 cm in thickness cut at 1.3 m DBH (Diameter Breast Height), a radial wood sample of (10 mm × 2 mm) was removed with the help of double-blade circular saw equipment. Once the radial strip was properly secured and allowed to air-dry, wood X-ray density profiles were obtained using a tree-ring analyzer (QMS densitometer, Model QTRS-01X) at the University of São Paulo, Piracicaba (Fig. 3). The wood X-ray densitometric profiles helped to determine the tree-ring boundaries for further dendrochronological dating. Wood anatomical features and dendrochronological counting revealed the presence of 71 tree rings (Fig. 3) from the pith to inner bark. Four wood anatomical structures were also observed. The dendrochronological analyses were obtained previously, and fully described in Lisi (2000) and Lisi et al. (2001), and can be summarized as follows:

Once the dendro-timescale of ARA-07 has been established, high-precision ^{14}C can be performed and then compared with the SH datasets on atmospheric $^{14}CO_2$ concentration, as an independent check to confirm the annual ring patterns of the *A. angustifolia* tree species.

4. Radiocarbon sample processing and analysis

From each annual tree ring, earlywood was separated from latewood. Wood samples of 10–100 mg were subdivided into 3 periods (before, during, and after nuclear tests), and then randomly

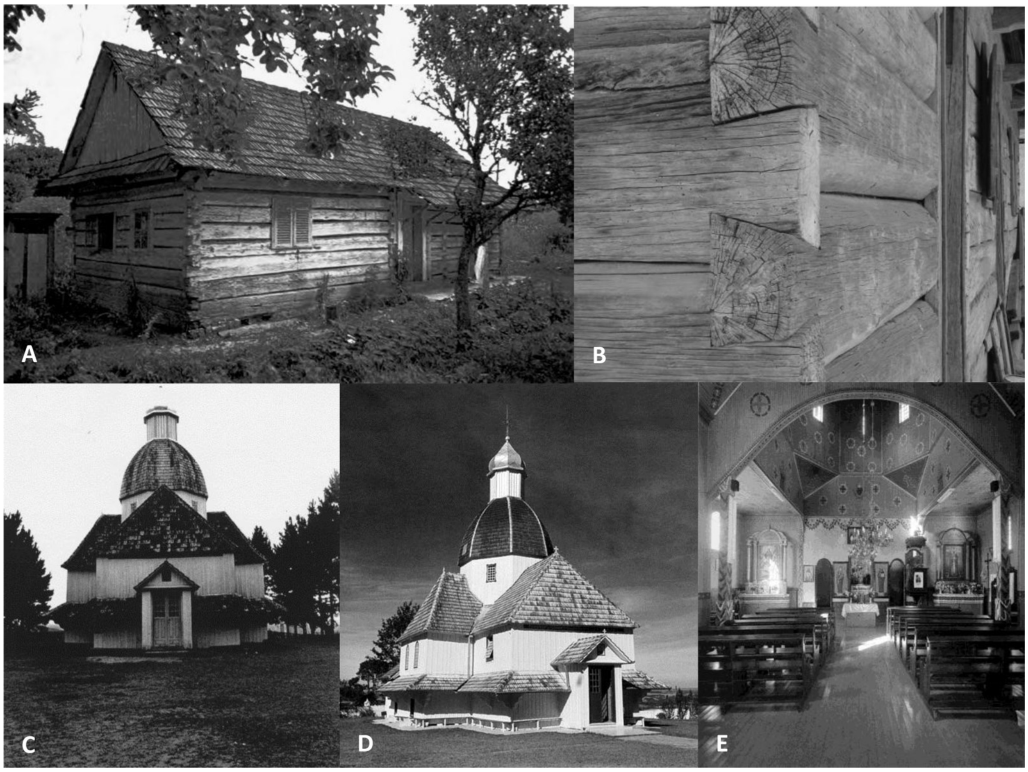

Fig. 2. Examples of wood buildings from the early 19th century made up exclusively from *A. angustifolia*: A) shows a household structure in wood, under the traditional Polish system of connecting larger timber logs without the use of iron nails (details in panel B); panel C) shows the São Miguel Arcanjo church (Ukrainian Orthodox) located at Dorizon, Municipality of Mallett, Paraná, whose building was begun in 1897 (photo from the archive of CPHA – SEEC, Pr.; Zani, 2013); D & E) show the same church after some of its sections underwent restoration, which was completed in 2011 (exterior and interior, respectively).

^{14}C-processed and measured in 3 batches. To evaluate precision and accuracy, wood samples from *A. angustifolia* were performed side by side with reference materials from the Fourth International Radiocarbon Inter-comparison exercise (FIRI) and the International Atomic Energy Agency (IAEA). Besides the 71 individual tree rings of ARA-07, another 12 random duplicates were also measured to aid accuracy. These were produced from random large tree rings, and processed separately from the original leftover materials. Radiocarbon dead (or ^{14}C-free) Queets-A wood was also added to batches to serve as blank material (Southon and Magana, 2010; Santos and Ormsby, 2013), as their ^{14}C values can be used for background corrections.

Extractives and the lignin from the original wood samples were removed by a holocellulose extraction procedure carried out on subsamples of 10–20 mg placed into 13 mm culture tubes. Wood aliquots were initially subjected to an acid-base-acid (ABA) pretreatment (Santos and Ormsby, 2013) at 70 °C with 1 N HCl for 30 min, 1 N NaOH for 60 min (the bath solution was repeated until supernatant was clear), followed by 1 N HCl for another 30 min. Holocellulose was isolated using equal volumes of 1 N HCl and 1 M $NaClO_2$ at 70 °C in a fume hood for approximately 4 h, or until the yellow/golden color subsided. After bleaching, the holocellulose was rinsed with warm Milli-Q water (70 °C/30 min) until pH >6 (Southon and Magana, 2010). Samples were then dried at 50 °C for several hours on a heat block, and the tubes were capped with gas-tight closures prior to undergoing combustion and graphitization for ^{14}C analysis. Samples were loaded into pre-baked quartz tubes with Ag wire and 60 mg CuO in preparation for combustion offline, then evacuated, sealed, and baked at 900 °C for 3 h. The CO_2 was cryogenically cleaned and then reduced to graphite in the presence of H_2 at 550 °C on pre-reduced Fe powder following established protocols (Santos et al., 2004, 2007). Graphite samples were pressed into Al targets and placed in the sputter ion source for measurement.

High-precision ^{14}C-AMS measurements were performed using a modified compact AMS system (NEC 0.5 MV 1.5SDH-1), which uses a fast beam switcher for sequential injection of ^{12}C, ^{13}C, and ^{14}C (Beverly et al., 2010). All results were corrected using the online δ^{13}C AMS values of the respective graphite samples investigated, following instrumental analysis described in Santos et al. (2007). The blank correction was obtained from ^{14}C analysis of Queets-A wood samples, subjected to the standard holocellulose extraction as discussed above. Samples of a subfossil FIRI-H wood, used here as a secondary standard, were also subjected to holocellulose extraction. Accuracy and precision, which were evaluated by a set of OX-I (as primary standard), and several secondary standards (FIRI-H, FIRI-G; Scott et al., 2004; and IAEA-C3 cellulose; leClercq et al., 1998), yielded ~2.0‰. Likewise, a recent evaluation of quality assurance between the KCCAMS/UCI and four other international labs (Hogg et al., 2013), when measuring decadal kauri samples ~10,000 ^{14}C years in age, demonstrated that the UCI laboratory can provide highly consistent ^{14}C results for holocellulose extract samples, and consequently no other crosschecks were considered necessary.

5. Radiocarbon results and discussion

The resulting Fraction Modern Carbon (FmC) ^{14}C signatures are reported in two formats. Table A1 (appendix) shows all ^{14}C results. A comparison of plots (Figs. 4 and 5) shows the same results against the SH datasets for atmospheric $^{14}CO_2$ concentrations. Even though the averaged calendar year values were previously assigned, we

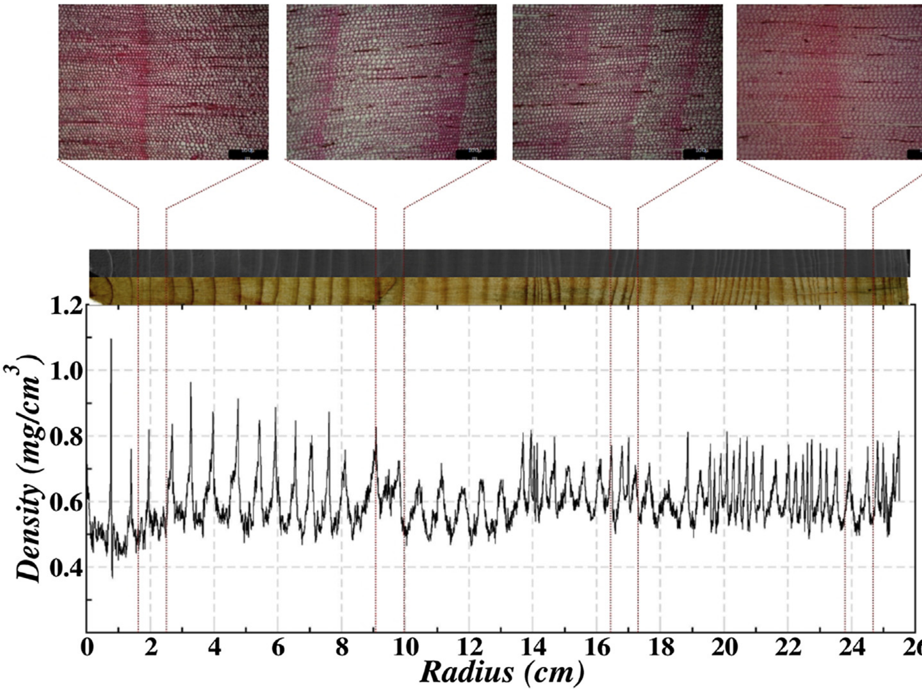

Fig. 3. The density (specific mass) of ARA-07 is shown, including wood anatomical images of four regions with different growth ring structures (bars of 100 μm) and radius cross-sections.

- The tree ring pattern shows distinct differences in color between earlywood (lighter) and latewood (darker). In addition, we observe significant differences in tree ring thickness along of the radial direction of the wood sample, possibly due to variations in past climate and/or tree-to-tree competition during growth.
- The variations in the tree ring wood density range from 0.4 to 1.1 g/cm^3, with a mean density value of 0.6 g/cm^3 across the wood sample;
- The differences observed in the tree ring wood density are associated with the structural changes of the tracheids' cell wall thickness of the early and latewood, as exemplified by the wood anatomical features of the four radial positions shown in Fig. 3.
- Direct tree ring counts from the edge of the incomplete bark inward, which are clearly visible to the naked eye, allow the calendar age of the Paraná pine ARA-07 (1927–1997) tree to be determined.

reevaluated whether those reflected local climate conditions that would promote C fixation for vegetation growth. Fig. 4 shows ^{14}C data for ARA-07 against the bomb-pulse SHCal data for zones 1–2 (Hua et al., 2013) for the period of 1955–1970, where the slopes of the curve are very sharp. For each panel (Fig. 4a–c) the dendrochronological dates have been shifted by increments of a few months until both datasets coincided. Although Fig. 4b and c shows no distinguishable differences, an adjustment of 0.95 was chosen

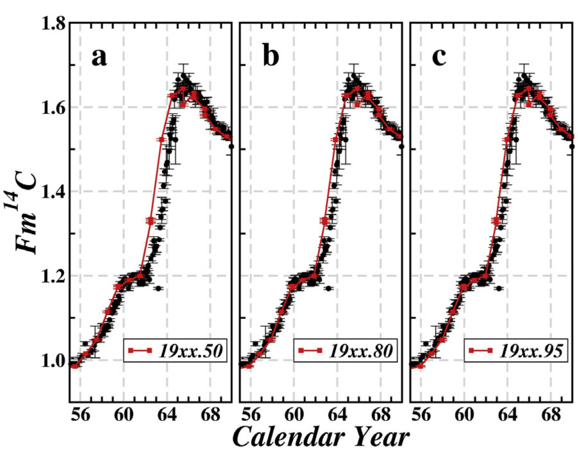

Fig. 4. The FmC ^{14}C values of the *A. angustifolia* of Camanducaia (22°50′S, 46°04′W) against the atmospheric record compilations for SHCal zones 1–2 (November–February) from 1950 to 1970 (Hua et al., 2013). Our data and the atmospheric record are represented in red and black, respectively. Each individual panel shows the month increments applied to nominal dendrochronological dates to evaluate the best fit for the calendar year. In a) an increment of 0.50 places the calendar year in the winter season, while the increments of b) 0.80 and c) 0.95 place it at the beginning or middle-wet summer season, respectively.

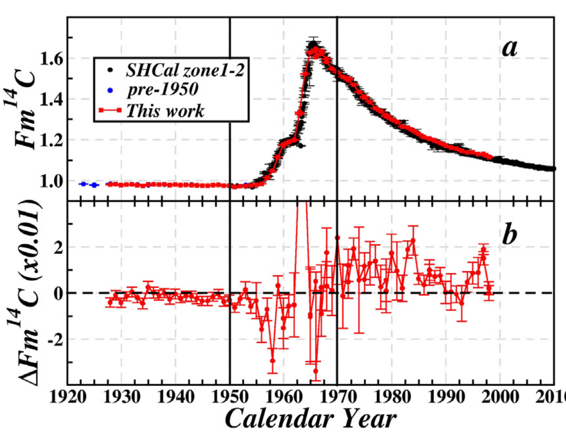

Fig. 5. To confirm the annual growth pattern of the *A. angustifolia* tree of Camanducaia (22°50′S, 46°04′W) by tree-ring/^{14}C cross-dating, we plot: a) the complete set of FmC ^{14}C values of ARA-07 and compare them with the atmospheric record for SH (November–February) from 1950 to 2010 and NH (May–August) (Hua et al., 2013). For the pre-bomb period of 1920–1948, represented in blue, we plotted FmC ^{14}C values from tree-rings of New Zealand and South Africa (McCormac et al., 1998, 2002; Hogg et al., 2002). The statistical uncertainties, shown as error bars, are smaller than the symbols; and b) to point out important patterns in our dataset, we use a cubic spline curves using a mathematical routine (Press et al., 2007). Uncertainties in datasets were also considered in the splines, and are represented as error bars.

for all calendar ages shown in Fig. 5. Most importantly, the calendar age chosen represents the averaged point in time when atmospheric ^{14}C exchange with the biosphere most likely occurred, which synchronizes with the middle-wet summer season running from the beginning of November to the end of February in Brazil's southeastern region.

High-precision ^{14}C-AMS results (Fig. 5a) showed the rise and rapid decrease of atmospheric ^{14}C concentrations associated with the detonation of nuclear weapons during the late 1950s, and its subsequent uptake by other large C sinks (such as oceans and the biosphere). A slight depletion of the atmospheric ^{14}C signal after 1920 is observed, possibly due to the Suess effect (a $^{14}C/C$ dilution response to burning of fossil fuel since the industrial revolution; McCormac et al., 1998). We also observed a slight but statistically significant divergence of our tree-ring/^{14}C data between 1975 and 1997 in regard to the atmospheric record compilations for SH and NH (which overlapped from the early 1970s onward).

To better evaluate local effects on the ^{14}C atmospheric content imprinted in *A. angustifolia* trees, we compare our present ^{14}C ARA-07 dataset to the reference pre-1950 period for South Africa and New Zealand tree rings (McCormac et al., 1998, 2002), and the post-1950 period SHCal zones1–2 (Hua et al., 2013). A cubic spline (Press et al., 2007) (Fig. 5b) was performed on the reference data points to estimate the values at exactly same calendar year as in the ARA-07 dataset, allowing for a direct subtraction. Thus we define $\Delta Fm^{14}C$ and its uncertainty as follows:

$$\Delta Fm^{14}C = \left[\text{This work}\right] - \left[\text{spline}\right] \tag{1}$$

$$\sigma\Delta^2 = \sigma^2_{\text{This work}} + \sigma^2_{\text{spline}} \tag{2}$$

Uncertainty of the interpolation at each calendar year was estimated as the averaged experimental uncertainty of neighbor points.

For the pre-1950 period we obtained an excellent agreement, as $\Delta Fm^{14}C$ is close to zero. Between 1950 and 1970 the $\Delta Fm^{14}C$ is quite variable due to the rapid increase of ^{14}C in the atmosphere. We suspected that some monthly variability played a role. This variability is more evident from the duplicates at the height of the bomb curve (Table A1) produced from separate pieces of wood. Despite our efforts to identify the orientation of the growth of the individual tree rings selected to undergo duplicates, we suspect that the small scale changes of the fiber direction under the microscope mean that we separated wood material from monthly intervals rather than from the intended full tree ring. The FmC ^{14}C result associated with the dendrochronological date AD 1965.95 (UCIAMS#116680 – Table A1), however, appears to be a true outlier. But because we did not know why this result appeared to be faulty, the result was not discarded. After 1970 the ARA-07 tree-ring/^{14}C dataset shows systematically higher ^{14}C activities (an increase of approximately 10‰) than the SHCal Zone 1–2 curve, despite the relative uncertainties. Since the C within tree rings remains fixed after formation, this increased ^{14}C production may reflect the following effects:

1) The phenomenon could be correlated with ^{14}C latitudinal differences (Braziunas and Stuiver, 1995; Krakauer et al., 2006). For instance, in the NH mid-latitudes fossil fuel emissions (which are ^{14}C-free) deplete atmospheric $^{14}CO_2$. However, carbon from the terrestrial biosphere can respire higher levels of bomb $^{14}CO_2$, which will enrich the ^{14}C signals above tropical forests. In addition, the exchange of ^{14}C-depleted C in the Southern Ocean can further reduce atmospheric ^{14}C in the southern mid-latitudes, enhancing the ^{14}C latitudinal difference toward the tropics.
2) Another aspect to be considered regarding this local ^{14}C increase is the huge Brazilian production of sugarcane (for commercial biomass energy through ethanol to replace gasoline). Brazil is among the largest producers of sugarcane for bioenergy consumption, which historically started in the early 1970s and has been concentrated mostly in the country's south and northeast regions (Goldemberg, 2008). Aside from a regional enriched $^{14}CO_2$ biogenic boost due to the Atlantic forest biome's proximity to the studied site, an increase in sugarcane production may also compete with the global atmospheric $^{14}CO_2$ dilution caused by fossil fuel burning. More evidence is needed to support this hypothesis.

To summarize, the remarkable agreement between our annual tree-ring/^{14}C data and the high-frequency SH Zone 1–2 compilation data (Hua et al., 2013) shows this subtropical tree species' potential for refining ^{14}C calibration curves. Furthermore, the present dataset (AD 1927–1997) can be used to determine the age and growth rate of tropical trees without annual tree ring patterns, to study the global carbon cycle, and in forensic sciences applications.

6. Conclusions and future directions

Here we report the first set of high-precision (0.2–0.3%) ^{14}C-AMS of a single tree growing at 22°50′S, 46°04′W (Camanducaia, Brazil) from AD 1927 to 1997. The annual formation of the *A. angustifolia* tree rings was confirmed by 83 ^{14}C dates, as significant agreement was found between this tree-ring/^{14}C data and the high-frequency SH Zone 1–2 compilation datasets (AD 1950–2010) and the pre-bomb tree ring sets from New Zealand and South Africa.

On a chemical procedural matter, the agreement between the dendrochronological dates and the ^{14}C atmospheric record confirms that our holocellulose extract procedure is suitable when treating wood samples for the purpose of expanding the ^{14}C calibration curve. In addition, the present ^{14}C data can be used to determine the age and growth rate of tropical trees without annual ring patterns, to study the global carbon cycle, and in forensic sciences applications.

Ultimately, we plan to continue the ^{14}C-AMS dating of other dendrochronologically-dated wood samples of species of *Araucariaceae* from different latitudes in South America, well preserved wood buildings from the late 18th century as well as other assemblies of subtropical Latin American tree species. By combining ^{14}C-AMS and dendrochronology, we aim to obtain ^{14}C cross-dated ancient wood archives, and use them to refine the existing SH ^{14}C calibration curve. In addition, natural and anthropogenic biogenic $^{14}CO_2$ from the Brazilian native and urban areas has not been thoroughly studied. By extending our present tree-ring/^{14}C data to current calendar years, and by comparing these data with the status of carbon fluxes on land, we may be able to better understand the CO_2 impacts of bioenergy and its contribution to the global carbon cycle.

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Appendix

Table A1
Extended ^{14}C data for *A. Angustifolia* tree, Camanducaia (22°50′S, 46°04′W), Brazil. All ^{14}C results are shown as Fraction Modern Carbon (FmC) signatures. The individual statistical error bars of the ^{14}C data were calculated based on counting statistics and scatter in multiple measurements on each sample, along with propagated uncertainties from normalization to a primary modern standard (OX-I), background subtraction based on processed Queets-A (^{14}C free wood), and isotopic fractionation corrections provided by the on-line $\delta^{13}C$-AMS values, following instrumental analysis described in Santos et al. (2007). Results of 12 duplicates are also shown (marked by asterisks). Large deviations among some of the duplicates are due to wood month variability, since tree-ring samples were not homogenized before being split to undergo duplicate sample processing and analyses.

Year (AD)	UCIAMS#	Fm^{14}C	
		Mean	1σ
1927.95	119253	0.9807	0.0012
1928.95	119254	0.9831	0.0014
1929.95	119255	0.9790	0.0013
1930.95	114749	0.9809	0.0012
1931.95	114750	0.9818	0.0015
1932.95	116666	0.9787	0.0016
1933.95	116667	0.9752	0.0015
1934.95	119256	0.9812	0.0017
1935.95	119257	0.9822	0.0014
1936.95	114771	0.9811	0.0013
1937.95	114752	0.9820	0.0012
1938.95	116668	0.9789	0.0015
1939.95	116669	0.9808	0.0015
1940.95	114753	0.9786	0.0012
1941.95	114754	0.9791	0.0014
1942.95	116670	0.9786	0.0016
1943.95	116671	0.9776	0.0017
1944.95	119258	0.9765	0.0012
1945.95	114755	0.9765	0.0012
1946.95	114756	0.9783	0.0012
1947.95	119259	0.9793	0.0013
1948.95	119260	0.9758	0.0014
1949.95	116672	0.9762	0.0015
1950.95	116673	0.9695	0.0016
1951.95	114757	0.9730	0.0012
1952.95	114758	0.9746	0.0012
1953.95	119261	0.9770	0.0018
1954.95	116674	0.9797	0.0015
1955.95	116675	0.9851	0.0016
1956.95	114759	1.0149	0.0013
1957.95	114760	1.0481	0.0017
1958.95	116676	1.1145	0.0017
1959.95	116677	1.1718	0.0023
1959.95*	119273	1.1761	0.0018
1960.95	116678	1.1897	0.0020
1961.95	114761	1.1997	0.0019
1962.95	114762	1.3342	0.0020
1962.95*	119270	1.3258	0.0018
1963.95	119262	1.5227	0.0023
1964.95	116679	1.6270	0.0026
1964.95*	119274	1.6280	0.0021
1965.95	116680	1.6050	0.0027
1965.95*	119275	1.6439	0.0023
1966.95	114763	1.6215	0.0022
1966.95*	119271	1.6328	0.0021
1967.95	114764	1.5811	0.0020
1967.95*	119272	1.5956	0.0020
1968.95	119263	1.5474	0.0022
1969.95	119264	1.5303	0.0020
1970.95	119265	1.5082	0.0020
1971.95	116681	1.4928	0.0024
1971.95*	119276	1.4999	0.0019
1972.95	114765	1.4699	0.0019
1973.95	114766	1.4336	0.0018
1974.95	116682	1.4057	0.0025
1974.95*	119277	1.4115	0.0018
1975.95	119266	1.3850	0.0023
1976.95	114767	1.3547	0.0017
1977.95	114768	1.3402	0.0017
1978.95	116683	1.3159	0.0025
1978.95*	119278	1.3206	0.0017
1979.95	116684	1.3010	0.0020
1980.95	114769	1.2873	0.0016
1981.95	114770	1.2638	0.0017
1982.95	116685	1.2596	0.0020
1983.95	116686	1.2438	0.0019
1984.95	114751	1.2287	0.0015
1985.95	114772	1.2131	0.0015
1986.95	116687	1.2045	0.0022
1986.95*	119279	1.2066	0.0018
1987.95	116688	1.1953	0.0019
1988.95	119267	1.1838	0.0017
1989.95	114773	1.1713	0.0015
1990.95	114774	1.1621	0.0015
1991.95	119268	1.1562	0.0017
1992.95	116689	1.1448	0.0019
1993.95	116690	1.1372	0.0018
1994.95	119269	1.1384	0.0014
1995.95	114775	1.1305	0.0014
1996.95	116691	1.1271	0.0024
1996.95*	119280	1.1310	0.0017
1997.95	116692	1.1148	0.0021
1997.95*	119281	1.1169	0.0015

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