# <sup>14</sup>C BOMB EFFECT IN TREE RINGS OF TROPICAL AND SUBTROPICAL SPECIES OF BRAZIL

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#### ABSTRACT

Atmospheric nuclear tests in the early 1960s introduced large amounts of radiocarbon into the atmosphere, which resulted in an increase of tropospheric  $^{14}\text{CO}_2$  concentration by nearly 100% during the years 1964–1965. The bomb-produced  $^{14}\text{C}$  was then gradually incorporated within the global carbon cycle. The history of  $^{14}\text{C}$  concentration in the troposphere is preserved within annual growth layers of trees and can be reconstructed for those areas where direct measurements of  $^{14}\text{C}$  in the atmosphere were not performed. The paper presents results of  $^{14}\text{C}$  activity measurements in tree rings of tropical and subtropical species from Brazil, for the period 1945–1997. We investigated two species (*Araucaria angustifolia* and *Parkia* sp.) growing at three sites covering the latitudinal band between 7°S and 24°S. The results indicate that the maximum  $^{14}\text{C}$  activity in the Southern Hemisphere occurred in 1965, with the  $^{14}\text{C}$  values reaching around 700%. Significant differences in  $^{14}\text{C}$  were recorded among the studied sites for the period of maximal  $^{14}\text{C}$  levels in the atmosphere, with the highest level observed at the tropical site and lowest in the subtropical zone. This reflects the dynamics of interhemispheric transport of  $^{14}\text{C}$  during the years of high spatial and temporal gradients of this isotope in the atmosphere.

Keywords: 14C bomb effect, tree-ring, Parkia sp., Araucaria angustifolia.

## INTRODUCTION

The nuclear bomb atmospheric tests began in 1952 (see *e.g.* Enting 1982) and resulted in a massive input of several radioactive isotopes into the atmosphere. Among them was <sup>14</sup>C, which is naturally produced by interaction of cosmic rays with the atmosphere. As a result, the <sup>14</sup>C levels in atmospheric CO<sub>2</sub> nearly doubled during the years

1960–1965. Measurements of  $^{14}\text{CO}_2$  content in the Northern Hemisphere indicate  $\Delta^{14}\text{C}$  values up to 1036‰ (71°06′N) in 1963 (Nydal and Lovseth 1996) (all ‰ figures are relative to pre-1950 levels). In the Southern Hemisphere Vogel and Marais (1971), analysed atmospheric  $^{14}\text{CO}_2$  in a site close of Pretoria, South Africa, and determined the  $^{14}\text{C}$  contents for the years of 1968–1970. The  $\Delta^{14}\text{C}$  values varied from 577‰ to 517‰. The maximum

 $^{14}$ CO<sub>2</sub> content for this hemisphere was recorded by Manning *et al.* (1990) in 1965 at Wellington (41°S, 174°E), New Zealand, where they found a  $\Delta^{14}$ C value of 695‰. After this period, the  $^{14}$ C levels have gradually declined, mostly due to transfer of  $^{14}$ C into the ocean, assimilation of radiocarbon by plants, and fossil fuel dilution (*e.g.* Nydal and Gislefoss 1996; Hua *et al.* 1999).

In this study, we measured the <sup>14</sup>C content in tree rings of three trees from tropical and subtropical sites in Brazil. The species studied were Parkia sp. from Amazonia tropical rainforest and pantropical forests, and Araucaria angustifolia from subtropical sites in Brazil. Parkia sp. attain heights of 20-40 m, and trunk diameters of 100 cm. The wood is used for civil construction (Lorenzi 1992; Camargos et al. 1993; Paula and Alves 1997; Ribeiro et al. 1999). A. angustifolia is a majestic tree that attain heights of 25-50 m, with a straight trunk 1-2 m in diameter. This species occurs in natural stands in southeastern Brazil and northeastern Argentina and Paraguay (Seitz and Kanninen 1988; Tomazello Fo et al. 2000). The annual growth rings of these species are distinct in trunk cross sections, allowing dendrochronological dating.

The objectives of this study were to determine the <sup>14</sup>C bomb effect in the tree-rings of tropical (Humaitá, southern Amazon state, Amazon region, 7°S) and subtropical (Camanducaia, Minas Gerais state, 22°S, southeastern Brazil and Arapotí, Paraná state, 24°S, southern Brazil) species (*Parkia* sp. and *Araucaria angustifolia*, respectively), during the pre- and post-nuclear tests periods (1943–1997), and to compare the results with previous work in the Northern and Southern hemispheres.

## **METHODS**

In the tropical region, the study sites are located in the Humaitá-AM region (7°56'S, 63°20'W), at an elevation of ca. 90 m.a.s.l. The mean temperature is around 26°C and the annual precipitation varies around a mean of 2,600 mm. In the subtropical regions the sites are (1) Camanducaia-MG (22°50'S, 46°04'W), at an elevation of 1,300 m.a.s.l., with a mean annual temperature of 18°C and mean annual rainfall of 1,660 mm; and (2)

Arapoti-PR (24°11′S, 49°58′W), at an elevation of 722 m.a.s.l., with a mean annual temperature of 20°C and mean annual rainfall of 1,670 mm. In both regions the analyzed trees were located in native forest, far from any sources of anthropogenic pollution.

For the wood anatomical analysis and identification of tree rings boundaries by X-ray densitometry, we collected disk samples at 1.3 m BHD (Breast Height Diameter) from the trees, dried them in the laboratory and polished the disk cross sections with a series of increasingly fine sand papers. For the X-ray densitometry method, 1 mm thick pith to bark wood samples were conditioned in a climate controlled room and exposed to an X-ray source. The wood X-ray films were analysed microdensitometrically and revealed radial wood density profiles (Schweingruber 1988; Amaral and Tomazello Fo 1998).

The tree rings were dated by wood anatomy and dendrochronological methods. For <sup>14</sup>C analysis we selected the annual growth rings formed before, during and after the nuclear testing period. In each annual tree ring, we separated the earlywood and part of the latewood of the following years: 1943, 1952, 1956, 1957, 1959, 1960, 1962 to 1966, 1968, 1971 to 1973, 1977, 1981, 1985, 1997. These are fragmented, crushed to ca. 0.2 mm and homogenised. The extraction of cellulose was based on the modified method of Green (1963). To remove the resins the wood samples received alcohol-toluene extraction, followed by alcohol extraction in the Soxhlet, and washed with distilled water. Then to remove lignin the samples were treated with sodium chlorite solution and acetic acid solution in a hot water bath (80°C). Finally, the extracted celluloses were washed several times with distilled water to remove any residual lignin and sodium chlorite. The cellulose samples were burned to carbon dioxide and converted to benzene (Pessenda and Camargo 1991). The <sup>14</sup>C activities were measured by liquid scintillation spectrometry. The results are expressed in  $\Delta^{14}$ C (%) and corrected for isotopic fractionation using δ<sup>13</sup>C values (Stuiver and Polach 1977).

The dendrochronological and  $\Delta^{14}$ C and  $\delta^{13}$ C determinations were carried out at the Tree-Ring Laboratory, Department of Forest Sciences, and

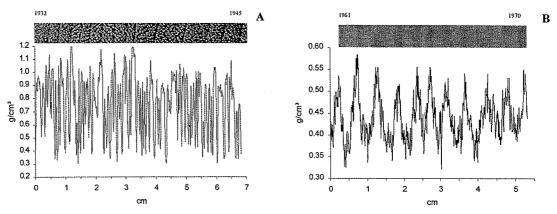


Figure 1. Wood anatomical structure and density profile by X-ray densitometry of Parkia sp. (A) and A. angustifolia (B) trees.

Radiocarbon and Stable Isotopes Laboratories, Center for Nuclear Energy in Agriculture, University of São Paulo, respectively.

# RESULTS

The wood density profiles by X-ray densitometry and the respective wood anatomical structure of microsections are shown in Figure 1, representing the years 1932–1945 and 1960–1970 for *Parkia* sp. and *A. angustifolia* trees, respectively. The wood anatomical struture of *Parkia* sp. is characterized by growth rings delimited by thin confluent parenchyma bands and fibrous zones. The annual growth rings of *A. angustifolia* are delimited by latewood with thick tracheid walls (Figures 1A, B) (Camargos *et al.* 1993; Seitz and Kanninen 1988; Tomazello Fo *et al.* 2000).

The *Parkia* sp. trees growing in Humaitá had 121 rings (1869–1989), and the *A. angustifolia* trees had 72 (1926–1997) and 51 annual growth rings (1945–1995) from Camanducaia and Arapoti, respectively. Table 1 and Figure 2 summarize the  $\Delta^{14}$ C and  $\delta^{13}$ C values of 27 tree rings of the selected years from the three trees. Six samples represent the pre-bomb period (1943 to 1959) and reveal the natural level of  $^{14}$ C, with  $\Delta^{14}$ C values fluctuating around 0% (-32.2% for Humaitá to 15.2% for Arapoti).

From 1960–1965 a significant increase was observed in the  $\Delta^{14}$ C values in the three locations, rising from 212.9‰ to 702.3‰, for Camanducaia site. Between 1966–1997 the values gradually de-

creased in all three locations, declining from 673.6% to 159.5%, in the Camanducaia record.

## DISCUSSION

The fibrous zones of Parkia sp. and the latewood of A. angustifolia trees were correlated with the higher values of X-ray wood density, permitting the establishment of an absolute chronology of the surveyed tree species. The 14C level for Humaitá in 1952 ( $\Delta^{14}$ C = -32.2%) is lower than observed for the same year at Camanducaia ( $\Delta^{14}$ C = 13.7%) and Arapotí ( $\Delta^{14}$ C = 15.2%). This may reflect a regional effect associated with recycling of CO<sub>2</sub> through the Amazon rainforest system. The significantly lower δ<sup>13</sup>C values for the Humaitá tree, compared with two other analyzed localities, seem to support this hypothesis. The <sup>14</sup>C analyses of trees growing in the Manaus region, Central Amazon, published by Worbes and Junk (1989) revealed very similar  $\Delta^{14}$ C values during the same period (1950–1954). The respiration flux of CO<sub>2</sub> from the soils would have slightly lower 14C concentrations due to an ageing effect in the soil reservoir. Differences of up to several per cent between the current 14C level in the atmosphere and respiration CO<sub>2</sub> were observed in a European forest (Dörr and Münnich 1986). Further measurements are needed to test this hypothesis in the context of tropical climate and the extent of the Amazon rainforest.

The highest <sup>14</sup>C levels in the analyzed tree-ring sequences occurred in 1965–1966 (Figure 2), ap-

<b>Table 1.</b> $\Delta^{14}$ C and $\delta^{13}$ C values in	1943–1997 annual	l growth rings of	f <i>Parkia</i> sp.	(Humaitá)	and $A$ .	angustifolia	(Camanducaia
and Arapotí).							

Years	Humaitá—AM 7°56'S, 63°20'W			Camanducaia—MG 22°50'S, 46°04'W			Arapotí—PR 24°11′S, 49°58′W			
	Lab n°	δ <sup>13</sup> C (‰)	Δ14C (%e)	Lab n°	δ <sup>13</sup> C (‰)	Δ14C (‰)	Lab n°	δ <sup>13</sup> C (‰)	Δ14C (%o)	
1943				679	-22.5	8.2 ± 7.4		_		
1952	_		_	973	-23.1	$13.7 \pm 7.8$	_	_	_	
1956	707	-26.9	$-32.2 \pm 7.8$	_	_	_	658	-21.3	$15.2 \pm 7.5$	
1957	708	-26.2	$-31.1 \pm 7.2$			_	_	_		
1959	709	-26.1	$-9.2 \pm 8.5$		_	_		-	_	
1960	_	_	_	747	-23.5	$212.9 \pm 9.1$	_	_	_	
1962	_		_	680	-22.9	$212.4 \pm 8.5$	_	_		
1963	718	-27.2	$520.9 \pm 10.1$	681	-22.1	$498.5 \pm 10.2$	662	-21.8	$317.5 \pm 9.2$	
1964	_		_	683	-23.1	$652.9 \pm 11.1$	_	_	_	
1965		_	_	677	-23.4	$702.3 \pm 11.4$	661	-22.0	$651.6 \pm 11.1$	
1966	_			676	-22.7	$673.6 \pm 11.3$	740	-22.5	$608.5 \pm 10.6$	
1968	717	-27.7	$598.4 \pm 10.7$	_	_	_	_		_	
1971	_	<del></del>				_	739	-20.8	$479.0 \pm 9.8$	
1972	716	-27.4	$490.6 \pm 9.9$	_			738	-22.6	$445.9 \pm 9.6$	
1973	_		<del></del>	750	-23.5	$418.9 \pm 10.5$	_	_		
1977	_		_	682	-22.1	$378.5 \pm 9.5$	660	-23.8	$321.9 \pm 9.2$	
1981		_	_	675	-22.2	$245.8 \pm 8.8$	_		_	
1985	_			749	-23.7	$233.3 \pm 9.4$	659	-22.8	$230.8 \pm 8.7$	
1997	_		_	674	-23.2	$159.5 \pm 8.3$	_	_	_	

proximately two years later than the maximum recorded in the Northern Hemisphere in 1963–1964 (Vogel and Marais 1971; Nydal *et al.* 1979; Levin *et al.* 1985; Dai and Fan 1986; Manning *et al.* 

1990; Dai *et al.* 1992). This agrees with other treering studies carried out on the South American continent (Worbes and Junk 1989; Yonenobu *et al.* 1995) and at other sites in the Southern Hemi-

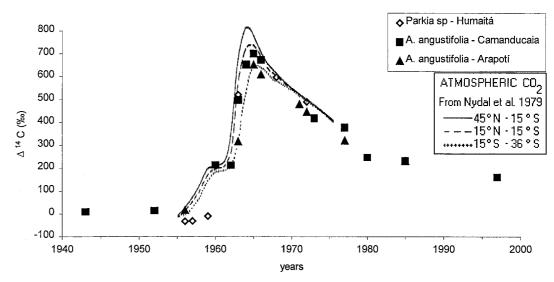


Figure 2.  $\Delta^{14}$ C in annual growth rings of *Parkia* sp. and *A. angustifolia* trees from three sites and determined by Nydal *et al.* (1979).

sphere (Vogel and Marais 1971; Manning *et al.* 1990; Yonenobu *et al.* 1995). The observed northsouth gradient in  $\Delta^{14}$ C reflects the dynamics of the inter-hemispheric transport of bomb-produced <sup>14</sup>C in the atmosphere. Figure 2 also illustrates the differences between the results obtained by Nydal with atmospheric  $CO_2$  in the Northern and Southern Hemispheres and the results obtained in this study.

As observed in the Figure 2, the years of maximum  $^{14}\text{CO}_2$  content in the Northern Hemisphere (1963–1964) reveal differences between Nydal's results and this study. Table 1 also show the same tendency among the analysed trees of Humaitá (7°S) and Arapoti (24°S). A distinct north-south gradient in the  $^{14}\text{C}$  content is observed: in 1963 in both sites, with  $\Delta^{14}\text{C}$  value of 521‰ at Humaitá and 317‰ at Arapotí. The apparent gradient in  $\Delta^{14}\text{C}$  is reduced from ca. 200‰ in 1963–1964 to approximately 45‰ in 1972–1973.

As reported in other publications (Vogel and Marais 1971; Nydal *et al.* 1979; Manning *et al.* 1990) and as can be seen in the Figure 2, after the years of maximum <sup>14</sup>C content the values gradually decreased in the Northern and Southern Hemispheres, mainly due to the CO<sub>2</sub> dilution from fossil fuels. The lowest value of 159.5‰ was obtained for Camanducaia site in 1997, and at present the value is around 90 to 100‰ in atmospheric CO<sub>2</sub> (Levin and Kramer 1997).

# **CONCLUSIONS**

The anatomy and X-ray densitometric analysis of wood samples of *Parkia* sp. and *Araucaria angustifolia* allowed us to establish an absolute chronology and to correlate to calendar years with  $^{14}{\rm C}$  activity and  $\delta^{13}{\rm C}.$   $^{14}{\rm C}$  analysis of tree-ring sequences, including the atmospheric nuclear bomb test period, allowed reconstruction of atmospheric  $^{14}{\rm C}$  levels during the period of relatively rapid changes of this isotope in the global atmosphere.

The results of <sup>14</sup>C content measurements in treering sequences belonging to tropical and subtropical Brazilian species (*Parkia sp* and *Araucaria angustifolia*) are consistent with available data for the Southern Hemisphere. Our results confirm that the maximum atmospheric <sup>14</sup>C concentration oc-

curred in the Southern Hemisphere during 1965-1966 and was considerably lower than that observed in the Northern Hemisphere. Our results also suggest the existence of a north-south gradient in <sup>14</sup>CO<sub>2</sub> concentration within the Southern Hemisphere and characteristic changes of this gradient with time. This reflects the dynamics of interhemispheric transport of 14C during the years of high spatial and temporal gradients of this isotope in the atmosphere, and the possible influence of the respiration CO2 flux from the Amazon rainforest. New sample collections should be carried out in the southern Amazon region, in order to improve the data base of this tropical site. Finally, the bomb 14C record in tropical trees may be a usefel tool for evaluating the growth rates of trees in cases where classical dendrochronological analysis provides doubtful results.

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