

## MODERN RADIOCARBON LEVELS FOR NORTHWESTERN MEXICO DERIVED FROM TREE RINGS: A COMPARISON WITH NORTHERN HEMISPHERE ZONES 2 AND 3 CURVES

Laura E Beramendi-Orosco<sup>1,2</sup> • Galia Gonzalez-Hernandez<sup>3</sup> • Jose Villanueva-Diaz<sup>4</sup> • Francisco J Santos-Arevalo<sup>5</sup> • Isabel Gómez-Martínez<sup>4</sup> • Edith Cienfuegos-Alvarado<sup>1</sup> • Pedro Morales-Puente<sup>1</sup> • Jamie Urrutia-Fucugauchi<sup>3</sup>

**ABSTRACT.** The radiocarbon variation for northwestern Mexico during the period 1950–2004 was studied by accelerator mass spectrometry (AMS) and liquid scintillation counting (LSC) analyses of tree rings. Two tree-ring sequences of *Pseudotsuga menziesii*, sampled in a site isolated from urban centers and active volcanoes (26.18°N, 106.3°W, 3000 m asl), were dendrochronologically dated and separated in annual rings prior to <sup>14</sup>C analysis. Results obtained show a similar profile to the values reported for the Northern Hemisphere (NH), having significant correlation coefficients with the compilation curves for NH zone 2 ( $r = 0.987, p < 0.001$ ) and NH zone 3 ( $r = 0.993, p < 0.001$ ). The maximum peak is centered at 1964.5 with a  $\Delta^{14}\text{C}$  value of  $713.15 \pm 9.3\%$ . The values obtained for the period 1958–1965 are lower than zone 2 values and higher than zone 3 values. For the period 1975–2004, the values obtained are higher than the NH compilation curve and other NH records. We attribute the first divergence to the North American monsoon that may have carried <sup>14</sup>C-depleted air from the south during the summer months; the second divergence may be attributable to <sup>14</sup>C-enriched biospheric CO<sub>2</sub>.

### INTRODUCTION

Radiocarbon levels increased significantly during the late 1950s and early 1960s as a result of atmospheric nuclear tests. After the Nuclear Test Ban Treaty in 1963, the atmospheric <sup>14</sup>C concentration started to decrease due to carbon exchange with other reservoirs. The excess atmospheric <sup>14</sup>C, produced mainly in high latitudes of the Northern Hemisphere, was heterogeneously distributed by atmospheric circulation during the bomb peak period (i.e. 1955–1969), creating 3 zones in the Northern Hemisphere (NH) (Hua and Barbetti 2004). The atmospheric <sup>14</sup>C reached equilibrium in the 1970s; therefore, its concentration became nearly homogeneous with differences of less than 16‰ between latitudes and longitudes for the NH (Hua and Barbetti 2004).

The NH division in 3 zones depends on atmospheric circulation and the seasonal positions of Hadley cell boundaries and the Intertropical Convergence Zone (ITCZ). These divisions were obtained by compiling monthly atmospheric and annual tree-ring <sup>14</sup>C records from different latitudes and longitudes (Hua and Barbetti 2004). However, the <sup>14</sup>C data compilations for NH zones 2 and 3 did not include any record from the American continent as all available records for the NH were from Europe, Asia, and Africa.

Mexico, situated in NH zone 2 with the summer ITCZ just off its Pacific southern coast, is a vast territory with active volcanoes and important urban and industrial areas. It lies between tropical and temperate latitudes (between 14°N and 32°N) and has a very diverse topography. As consequence, the climate varies from a tropical south to an arid north and temperate areas along the mountains, with dry winters and wet summers (Barajas et al. 1986). The trans-Mexican volcanic belt, located between 19°N and 21°N crossing the country from west to east, has been active during the last century at the Colima, Parícutin, and Popocatepetl volcanoes. The population in Mexico increased from

<sup>1</sup>Instituto de Geología, Universidad Nacional Autónoma de México, Ciudad Universitaria, México DF 04510, México.

<sup>2</sup>Corresponding author. Email: laura@geofisica.unam.mx.

<sup>3</sup>Instituto de Geofísica, Universidad Nacional Autónoma de México, Ciudad Universitaria, México DF 04510, México.

<sup>4</sup>Instituto Nacional de Investigaciones Forestales y Agropecuarias, CENID-RASPA, Gómez Palacio, Durango 35140, México.

<sup>5</sup>Centro Nacional de Aceleradores (CNA), Avda. Thomas Alva Edison 7, Isla de la Cartuja, Sevilla 41092, Spain.

25.8 million in 1950 to 103.3 million in 2005, with 76.5% concentrated in urban areas (INEGI 2008). All these characteristics make it important to study the  $^{14}\text{C}$  levels for Mexico during the second half of the 20th century because the  $\text{CO}_2$  emissions, either anthropogenic or volcanic, must have been considerable.

Previous work on modern  $^{14}\text{C}$  levels for Mexico has been scarce. The earliest  $^{14}\text{C}$  measurement for Mexico was reported by Tauber (1967) obtained from a grass sample collected outside Mexico City (19.42°N, 99.12°W) during the summer of 1960. The value reported by Tauber ( $\Delta^{14}\text{C} = 210 \pm 9\text{‰}$ ) is closer to the values for NH zone 3 than for NH zone 2. Other work reporting modern  $^{14}\text{C}$  values for Mexico was related to the dendrochronological potential of *Pinus lagunae* from Baja California (23.5°N, 109.9°W) (Biondi and Fessenden 1999). They attempted to check if growth rings were indeed annual by measuring tree-ring  $^{14}\text{C}$  contents and contrasting them with the NH modern  $^{14}\text{C}$  profile, but the  $^{14}\text{C}$  results suggested either missing or false rings. The values reported for one of the studied trees have a lag of about 5 yr with respect to the NH  $^{14}\text{C}$  curves; the highest value is centered at 1964–1965 with a  $\Delta^{14}\text{C}$  of 675‰, very similar to the NH zone 3 curve.

Our aim is to begin a characterization of the long-term variation of  $^{14}\text{C}$  levels, including the bomb-peak period, for the area of the American continent covered by NH zone 2. Here, we report  $^{14}\text{C}$  levels for northwestern Mexico obtained by accelerator mass spectrometry (AMS) and liquid scintillation spectrometry (LSC) analyses of tree rings.

## METHODS

### Sampling and Sample Preparation

The sampling site was selected with 2 criteria: first, it had to be isolated from  $\text{CO}_2$  sources, and second, it needed to have the presence of tree species with reliable annual rings suitable for dendrochronological dating.

The selected sampling area, located in the Sierra Madre Occidental (26.18°N, 106.3°W) at 3000 m asl, is a mixed coniferous forest characterized by subtropical climate with dominant summer precipitation. The nearest human settlement is located 0.9 km away with a population of just 100 inhabitants. The nearest urban and industrial area, Culiacan (790,000 inhabitants), is located 180 km southwest (Figure 1).

Cross-sections of 2 Douglas-firs (*Pseudotsuga menziesii*), one felled in 2003 and the other in 2005, were dated by standard dendrochronological techniques through a master tree-ring chronology previously developed for the site (Villanueva-Diaz et al. 2008, 2009). In order to construct a master plot, individual skeleton plots from increment cores and cross-sections of Douglas-fir were cross-dated; then, the master series was used to predate the wood (Stokes and Smiley 1996). The predated rings were divided in bands of total ring width, earlywood and latewood; and measured with a Velmex device to the nearest 0.001 mm (Cleaveland et al. 2003). The COFECHA program was used to control the quality of the dating (Holmes 1983) and the ARSTAN program was used to produce tree-ring indices (Cook 1987). The common interval time span of the chronology was 1829 to 2005 and had a mean sensitivity of 0.25, a correlation of 0.537 among all radii, 0.534 between trees, 0.719 within trees, signal-to-noise ratio of 30.9, and variance in the first eigenvector of 55.5%. In the last decade, several dendrochronologies have been constructed for northern Mexico using Douglas-fir (Therrell et al. 2002; Cleaveland et al. 2003; González-Elizondo et al. 2005; Villanueva-Diaz et al. 2007), including the master tree-ring chronology for the sampling site (Villanueva-Diaz et al. 2008).

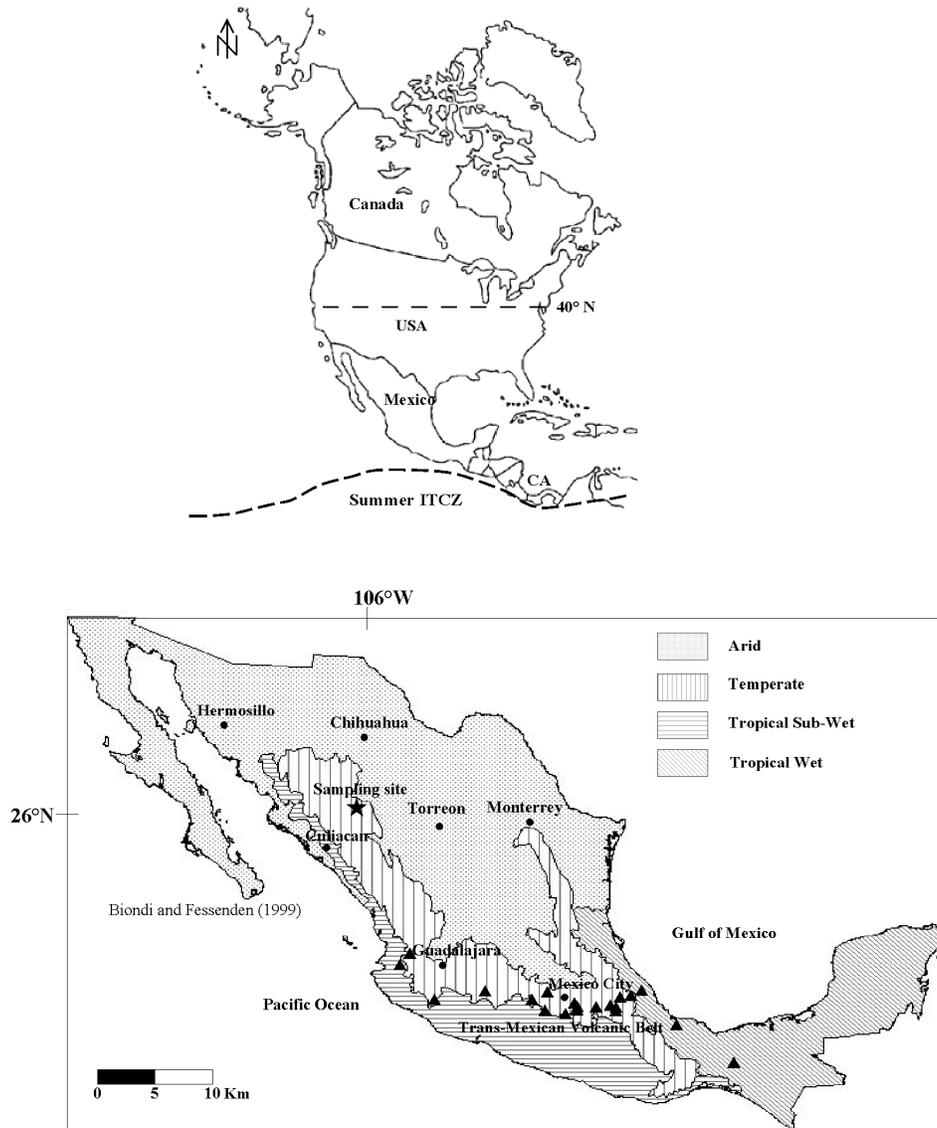


Figure 1 Map showing location of the sampling site. Shaded areas correspond to different to climatic zones (Barajas et al. 1986). Principal urban centers, the trans-Mexican volcanic belt ( $\Delta$ ), and the summer position of the ITCZ are also indicated. Sampling sites for previous studies on  $^{14}\text{C}$  for Mexico are included as a reference.

Pieces of wood were cleaned in a Soxhlet extractor with a mixture of ethanol:toluene (2:1) for more than 24 hr. Annual rings were separated using a stainless steel blade and milled in a ball mill. Samples for AMS analysis (Table 1) were further separated into early- and latewood. The AMS results presented here correspond to latewood of single tree rings from only 1 tree. Samples measured by LSC were a mixture of whole wood of consecutive rings, from both trees, in order to obtain a sample size suitable for analysis. The sampling resolution for the bomb-peak period (1955–1965) was annual because the highest variations in  $^{14}\text{C}$  atmospheric concentration took place during these years.

Table 1  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  data for Douglas-fir (*Pseudotsuga menziesii*) tree-rings from northwestern Mexico.

Year	Type of sample	Lab code	$\delta^{13}\text{C}$ (‰) $\pm 1 \sigma$	$\Delta^{14}\text{C}$ (‰) $\pm 1 \sigma$
1950.5	Latewood single ring	CNA-215	$-23.22 \pm 0.4$	$-22.98 \pm 2.99$
1955.5	Latewood single ring	CNA-216	$-23.72 \pm 0.2$	$-5.48 \pm 3.18$
1956.5	Latewood single ring	CNA-217	$-26.26 \pm 0.3$	$29.64 \pm 3.16$
1957.5	Latewood single ring	CNA-218	$-24.35 \pm 0.3$	$62.52 \pm 4.30$
1958.5	Latewood single ring	CNA-219	$-24.53 \pm 0.8$	$97.81 \pm 6.37$
1959.5	Latewood single ring	CNA-220	$-23.77 \pm 0.8$	$168.72 \pm 6.26$
1960.5	Latewood single ring	CNA-221	$-24.51 \pm 0.8$	$206.92 \pm 6.53$
1961.5	Latewood single ring	CNA-222	$-25.61 \pm 0.8$	$209.27 \pm 6.79$
1962.5	Latewood single ring	CNA-223	$-23.03 \pm 0.8$	$280.77 \pm 6.83$
1963.5	Latewood single ring	CNA-224	$-21.09 \pm 1.1$	$602.78 \pm 10.33$
1964.5	Latewood single ring	CNA-225	$-23.96 \pm 0.8$	$713.15 \pm 9.23$
1965.5	Latewood single ring	CNA-226	$-25.92 \pm 0.9$	$701.40 \pm 8.71$
1970.5	Latewood single ring	CNA-227	$-23.87 \pm 0.9$	$515.82 \pm 7.63$
1971–1973	Whole wood combined rings	UNAM-0832	$-24.45 \pm 0.2$	$444.64 \pm 2.28$
1975.5	Latewood single ring	CNA-228	$-24.04 \pm 0.8$	$385.75 \pm 7.41$
1980.5	Latewood single ring	CNA-229	$-26.18 \pm 0.8$	$293.29 \pm 6.91$
1981–1984	Whole wood combined rings	UNAM-0827	$-24.50 \pm 0.2$	$265.20 \pm 2.28$
1985–1989	Whole wood combined rings	UNAM-0828	$-24.57 \pm 0.2$	$222.28 \pm 2.18$
1990.5	Latewood single ring	CNA-230	$-27.46 \pm 0.8$	$162.45 \pm 6.24$
1991–1994	Whole wood combined rings	UNAM-0829	$-24.80 \pm 0.2$	$121.96 \pm 2.28$
1995–1999	Whole wood combined rings	UNAM-0830	$-24.98 \pm 0.2$	$109.28 \pm 2.28$
2000.5	Latewood single ring	CNA-231	$-25.97 \pm 0.8$	$84.81 \pm 6.34$
2001–2002	Whole wood combined rings	UNAM-0831	$-25.00 \pm 0.2$	$94.37 \pm 2.28$
2004.5	Latewood single ring	CNA-232	$-26.58 \pm 0.8$	$71.54 \pm 5.55$

### LSC Analysis

Cleaned and milled samples were transformed to benzene in a commercial benzene synthesis line (TASK Inc.). Benzene samples (1.5 mL) were mixed with 0.5 mL of scintillation cocktail (2,5-diphenyloxazole (PPO) + 1,4-Bis(5-phenyl-2-oxazolyl)benzene (POPOP) dissolved in dead spectrophotometric-grade benzene) in 3-mL Teflon<sup>®</sup> vials.

Analysis was performed in a Quantulus 1220<sup>™</sup> ultra-low-level liquid scintillation spectrometer as detailed in Beramendi-Orosco et al. (2006). Each sample was analyzed for 2500 min, alternating sample vials with oxalic acid SRM 4990C standard and background vials. The counting window was set to optimize the figure of merit with a <sup>14</sup>C counting efficiency higher than 60%.

Stable  $\delta^{13}\text{C}$  isotope analyses were performed at the Stable Isotopes Laboratory at the Institute of Geology, UNAM. Measurements were done using a Finnigan Delta Plus XL mass spectrometer with dual inlet and Elemental Analyzer Flash 1112EA and ConFloIII as interface. Samples were combusted in sealed quartz tubes with CuO, Cu, and Ag at 900 °C for 2 hr and at 650 °C for 1 hr for reduction of nitrogen oxides. The CO<sub>2</sub> obtained was purified by cryogenic separation in a vacuum line. Results are reported relative to the Vienna Pee-Dee belemnite (VPDB) standard with a precision of 0.2‰ and normalized with NBS-19 (calcite standard reference material) and LSVEC (lithium carbonate standard reference material) scale according to Coplen et al. (2006).

### AMS Analysis

Approximately 6 mg of cleaned sample was combusted in a vacuum-sealed quartz tube at 950 °C for 4 hr with CuO and Ag powder. The produced  $\text{CO}_2$  was then reduced to graphite by adding excess  $\text{H}_2$  using cobalt as a catalyst. The resultant mixture of graphite and cobalt was pressed into aluminium cathodes and kept under vacuum until analysis.

The AMS procedure we employed has been described elsewhere (Chamizo et al. 2008; Santos-Arevalo et al. 2009). Briefly, the terminal voltage is set at 1MV, and a charge state of 2+ is selected after the stripping process. The typical extracted  $^{12}\text{C}$  current in the low-energy side is about 25  $\mu\text{A}$ , and transmission is ~43%.  $^{14}\text{C}$  is identified in an ionization chamber working as a  $\Delta\text{E}-\text{E}_{\text{res}}$  detector. Selecting a charge state of 2+ after the stripper allows  $^7\text{Li}^+$  to reach the detector, which could produce a severe interference. The detector resolution is good enough to separate both signals created by  $^7\text{Li}$  and  $^{14}\text{C}$ , and problems only arise in the case of very intense  $^7\text{Li}^+$  count rates. Yet, by fine tuning the high-energy analyzers and using slits, the  $^7\text{Li}^+$  count rate can be reduced to a minimum, and spectra are clear enough to easily identify the  $^{14}\text{C}$  counts.

Samples were measured in 6 runs, consisting of 10 cycles of 30 s each. During data analysis, any of these cycles were eliminated if affected by any instantaneous instability of the system, or eventual bursts of Li that could saturate the detector.

Measurements were performed on batches of graphite cathodes from a variable number of samples, together with blank samples (1 per 10–15 unknown samples), oxalic acid SRM 4990C standard samples for normalization (1 per 7–8 unknown samples), and 1 or 2 intercomparison samples.

### RESULTS AND DISCUSSION

The  $^{14}\text{C}$  results, expressed as  $\Delta^{14}\text{C}$  corrected for isotopic fractionation and decay-corrected (Stuiver and Polach 1977), are plotted in Figure 2 and tabulated in Table 1. The bomb peak obtained for northwestern Mexico shows a similar profile to that reported for the Northern Hemisphere, having significant correlation coefficients with the compilation curves for NH zone 2 ( $r = 0.987$ ,  $p < 0.001$ ) and NH zone 3 ( $r = 0.993$ ,  $p < 0.001$ ). The peak is centered at 1964.5 with a  $\Delta^{14}\text{C}$  value of  $713.15 \pm 9.3\text{‰}$  ( $F^{14}\text{C} = 1.7252 \pm 0.009\text{‰}$ ), which is lower than the peak value of the NH zone 2 by 90.85‰ and higher than the peak of the NH zone 3 (centered at 1965.5) by 8.15‰ (Table 2). The  $\Delta^{14}\text{C}$  value for the pre-bomb period (1950) is  $-22.98 \pm 3.0\text{‰}$  ( $F^{14}\text{C} = 0.9839 \pm 0.003\text{‰}$ ); a value of  $71.54 \pm 5.6\text{‰}$  ( $F^{14}\text{C} = 1.0791 \pm 0.005\text{‰}$ ) was found for 2004.

An important feature of the profile for northwestern Mexico is that the  $\Delta^{14}\text{C}$  values for the period 1958–1965 are lower than those reported for the sites in the NH zone 2 compilation by a mean of 62.02‰. In contrast, the  $\Delta^{14}\text{C}$  values obtained for the period 1975–1999 are higher than the NH compilation by a mean of 11.76‰. Furthermore, for the period 2000–2004, the values obtained are also higher than those reported by other records for NH sites, such as for Jungfrauoch, Switzerland (Levin and Kromer 2004) and Niwot Ridge, Colorado, USA (Turnbull et al. 2007); both records from stations at similar altitude as the sampling site. For example, during the summer months of 2004 the Niwot Ridge record has a mean value of  $\Delta^{14}\text{C} = 64.14\text{‰}$ , lower than the value found in this study by 7.40‰.

The fact that the values obtained for the 1958–1965 period are lower than the values for NH zone 2 can be a consequence of the atmospheric circulation over northwestern Mexico, which during the summer months is characterized by winds from the south dominated by the North American monsoon (Higgins et al. 1999). This may affect the atmospheric  $^{14}\text{C}$  of the region, with more  $^{14}\text{C}$ -

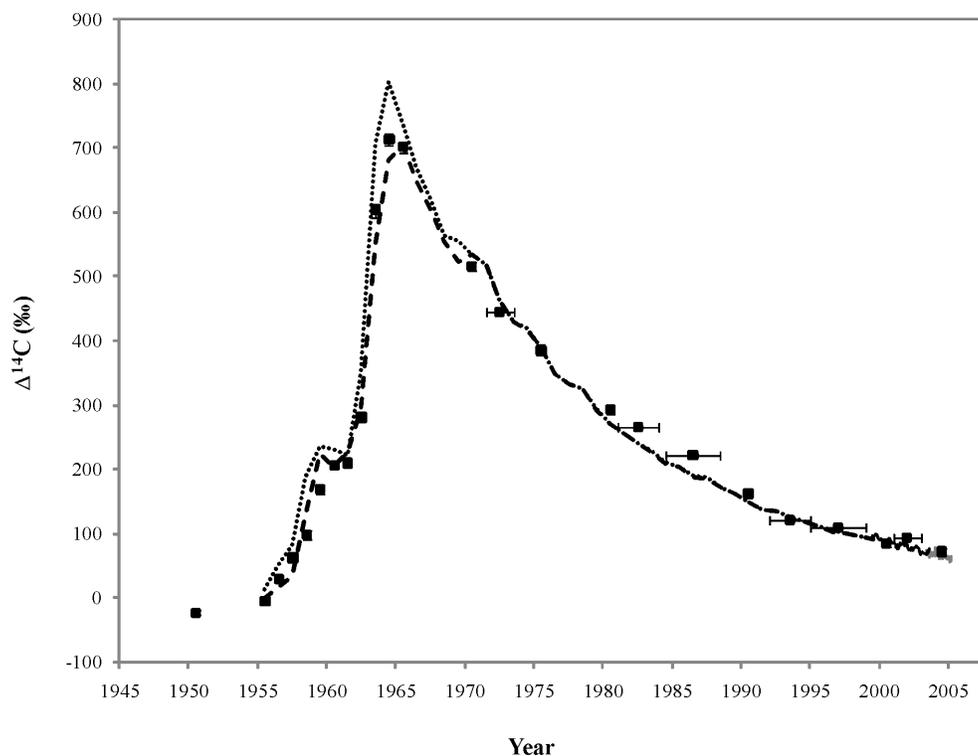


Figure 2 Plot of  $\Delta^{14}\text{C}$  values vs. years for Douglas-fir (*Pseudotsuga menziesii*) tree rings from northwestern Mexico (squares) and curves for the NH zone 2 (dotted line), NH zone 3 (dashed line) (Hua and Barbetti 2004), Jungfraujoeh, Switzerland (black line) (Levin and Kromer 2004), and Niwot Ridge, Colorado USA (gray line) (Turnbull et al. 2007). Vertical error bars correspond to  $\pm 1 \sigma$ ; horizontal error bars reflect the number of years comprising samples analyzed by LSC (see Table 1).

Table 2 Comparison between  $\Delta^{14}\text{C}$  data for northwestern Mexico and Northern Hemisphere zones 2 and 3 for the period 1955–2000.

	$\Delta^{14}\text{C}$ of peak (‰ $\pm 1 \sigma$ )	Peak year	Difference with Mexico (‰)	Correlation coefficient with Mexico ( $r, p < 0.001$ )
Northwestern Mexico	$713.15 \pm 9.3$	1964.5	—	—
NH zone 2	$804 \pm 8$	1964.5	-90.85	0.987
NH zone 3	$705 \pm 6$	1965.5	+8.15	0.993

depleted air from the south during the growing season months of the studied tree species (May to October). A similar effect has been reported for tropical Africa where air masses from the Southern Hemisphere, carried by the summer southwest Asian monsoon, may influence the atmospheric  $^{14}\text{C}$  level (Hua and Barbetti 2007). This could also explain the relatively low  $\Delta^{14}\text{C}$  value found by Tauber (1967) for the summer of 1960 outside Mexico City. Yet in this case, the fossil fuel effect cannot be ruled out as the reported sampling site is indeed very close to Mexico City, inside the Valley of Mexico.

For the period 1975–2004, although the differences between our data and the other NH records are within those reported by Hua and Barbetti (2004) in their compilation, the fact that the northwestern Mexico  $\Delta^{14}\text{C}$  values are higher than those reported for the NH could be attributed to the contribution of  $^{14}\text{C}$ -enriched biospheric  $\text{CO}_2$  derived from respiration, which may be considerable during the growing season months due to the high forest density of the sampling site, and also to the fact that the fossil fuel factor is negligible as a consequence of the isolation from possible fossil  $\text{CO}_2$  emissions. In recent decades, the respiration from northern ecosystems has become  $^{14}\text{C}$ -enriched relative to the contemporary background atmosphere because carbon residence time in vegetation and soils spans several years (Randerson et al. 2002). By measuring  $\Delta^{14}\text{C}$  from black spruce forest in Alaska, Randerson and coworkers inferred that most northern ecosystems have been a net source of  $^{14}\text{C}$  for the last decades. The  $\Delta^{14}\text{C}$  of the biosphere's respiration can be significantly higher than that of the atmosphere (Turnbull et al. 2006). Levin et al. (2008) estimated the biospheric  $\Delta^{14}\text{C}$  for Heidelberg in summer 1986 and 2006 to be 15 and 22‰ higher than the background  $\Delta^{14}\text{C}$ , respectively.

## CONCLUSIONS

$\Delta^{14}\text{C}$  levels for northwestern Mexico measured in Douglas-fir tree rings are consistent with the  $^{14}\text{C}$  variation profile reported for the Northern Hemisphere (NH). However, there are significant differences with the reported values for the compilation curve of the NH zone 2. Whereas the values obtained for period 1958–1965 are depleted in  $^{14}\text{C}$  relative to the NH zone 2, for the period 1975–2004 the values are enriched relative to the NH records. We attribute the first divergence to the fact that the summer climate for the studied area is influenced by the North American monsoon, which carries  $^{14}\text{C}$ -depleted air from the south. The second divergence may result from the contribution of  $^{14}\text{C}$ -enriched biogenic  $\text{CO}_2$  and the isolation from fossil  $\text{CO}_2$  sources.

The results presented here make evident the need for analyzing the remaining years of the tree-ring sequences, improving the resolution in order to better understand the factors that affect  $^{14}\text{C}$  levels for the area. It also becomes apparent that long-term  $^{14}\text{C}$  records must be generated for other parts of Mexico with different wind patterns and biogenic  $\text{CO}_2$  contribution, which will help to elucidate the reasons for the above divergences.

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