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ATMOSPHERIC ^{14}C VARIABILITY RECORDED IN TREE RINGS FROM PENINSULAR INDIA: IMPLICATIONS FOR FOSSIL FUEL CO_2 EMISSION AND ATMOSPHERIC TRANSPORT

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ABSTRACT. Radiocarbon analysis in annual rings of a teak tree (*Tectona grandis*) is reported in comparison with previously published results. Samples (disks) were collected from Hoshangabad (22°30′N, 78°E), Madhya Pradesh, in central India. The previously published sample was collected from Thane (19°12′N, 73°E), Maharashtra, near the west coast of India (Chakraborty et al. 1994). Two short $\Delta^{14}C$ time series were reconstructed with these tree samples to capture the bomb peak of atmospheric ^{14}C and the spatial variability in this record. These time series represent the periods 1954–1977 and 1959–1980 for Hoshangabad and Thane, respectively. The ^{14}C peaks in these places appear around 1964–1965. The Hoshangabad tree records a peak $\Delta^{14}C$ value of 708 ± 8‰, which conforms to the peak value of Northern Hemisphere Zone 3 as described in Hua and Barbetti (2004). But the peak $\Delta^{14}C$ at Thane is somewhat less (630 ± 8‰) probably due to the dilution by fossil fuel CO_2 free of ^{14}C emanating from the neighboring industrial areas. This depletion of peak values has been used to estimate the local emission of fossil fuel CO_2, which is approximately 2.3% of the background atmospheric CO_2 concentration.

INTRODUCTION

The variation of atmospheric radiocarbon activity has been used to study various natural processes. The atmospheric ^{14}C activity during the early 1960s rose to almost double relative to the pre-bomb (pre-1950s) level as a result of aboveground nuclear bomb testing. This transient peak of ^{14}C gave an opportunity to study various earth-system processes such as atmospheric transport (Manning et al. 1990; Land et al. 2002), air-sea gas exchange rates (Broecker et al. 1985; Chakraborty et al. 1994; Hua et al. 2003; Sweeney et al. 2007), soil carbon turnover (Harrison et al. 1993), and patterns of regional fossil fuel-derived CO_2 concentration (Tans 1978; de Jong and Mook 1982; Levin and Kromer 1997; Levin et al. 2003; Turnbull et al. 2006; Hsueh et al. 2007). Accurate estimates of bomb ^{14}C inventory in the atmosphere and the change over time are crucial for validating theoretical models of carbon pool turnover rates (Guilderson et al. 2000; Naegler et al. 2006; Sweeney et al. 2007).

The maximum production of bomb ^{14}C was in the Northern Hemisphere as a result of being the region of the majority of the atmospheric nuclear tests. ^{14}C was then transferred southwards by atmospheric circulation, depending on regional wind patterns and the resistance of atmospheric cell boundaries (Hua and Barbetti 2004). It is also believed that the interhemispheric exchange of ^{14}C is also controlled by the monsoon air circulation modulated by seasonal movement of the intertropical convergence zone (ITCZ) (Hua and Barbetti 2004). Once the ^{14}C excess is transported across the ITCZ, atmospheric ^{14}C diffuses rapidly with fast mixing in the Southern Hemisphere, resulting in reduced amplitude and a lower latitudinal gradient compared to the Northern Hemisphere (Hua et al. 1999). Hua and Barbetti (2004) classified the global tropospheric ^{14}C records for the later half of the 20th century into 4 zones, 3 for the Northern Hemisphere and 1 for the entire Southern Hemisphere. These zones are based on the amplitude and occurrence of the bomb ^{14}C peak, from monthly atmospheric $^{14}CO_2$ measurements or from ^{14}C measured in tree rings from various geographical locations. These data show a progressive decrease of $\Delta^{14}C$ peak values across the latitudes from a high

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of about 1040‰ during summer of 1963 at Fruholmen (71°N), Norway (Nydal and Lövseth 1996), to a low of 628‰ recorded in tree rings formed in 1965 at Armidale (30°S), Australia (Hua et al. 2003). However, from detailed analysis of atmospheric ^{14}C data from various latitudes, Hua and Barbetti (2007) concluded that there is no simple latitude dependence of atmospheric ^{14}C gradients, but that seasonal atmospheric circulation also played a role during the peak bomb period between 1963 and 1967.

Out of the 4 global zones, India, SE Asia, and part of Africa were classified in the Northern Hemisphere (NH) Zone 3 with a peak $\Delta^{14}C$ value at 705 ± 6‰ during 1965 (Hua and Barbetti 2004). The $\Delta^{14}C$ profile for this zone was a composite curve based on the analysis of atmospheric $^{14}CO_2$ from Debre Zeit in Ethiopia (9°N, 39°E; Nydal and Lövseth 1996), in combination with tree-ring ^{14}C data from Doi Inthanon, Thailand (19°N, 99°E; Hua et al. 2000); Saigon, Vietnam (11°N, 107°E; Kikata et al. 1992); and Mandla, central India (22°35′N, 80°22′E; Murphy et al. 1997) (Table 1). Though the atmospheric $\Delta^{14}C$ data used in the Hua and Barbetti (2004) compilation have a narrow range of peak values (692‰ to 705‰) within NH Zone 3, other places in this same geographical region do show large variability in peak $\Delta^{14}C$ values. Hua et al. (2000) reported $\Delta^{14}C$ values of 694 ± 7‰ in 1965 and 653 ± 9‰ in 1966 measured in a 3-leaf pine tree from Doi Inthanon, NW Thailand. Chakraborty et al. (1994) reported a peak bomb ^{14}C of 630 ± 8‰ for 1964–1965, measured in a teak tree collected from Thane (19°30′N, 74°12′E), near Mumbai (formerly Bombay), India. Meanwhile, Murphy et al. (1997) reported a bomb ^{14}C peak of 705 ± 9‰ for 1965 measured in teak tree rings from Mandla, ~1300 km ENE of Thane. Table 1 summarizes the peak $\Delta^{14}C$ values for NH Zone 3.

Table 1 Peak $\Delta^{14}C$ in atmospheric CO_2 recorded from various locations of NH Zone 3.

Location	Sample type	Peak $\Delta^{14}C$ (‰)	Peak year	Reference
Saigon, Vietnam (11°N, 107°E)	Tree ring	692 ± 16	1965	Kikata et al. (1992)
Thane, western India (19°30′N, 74°12′E)	Tree ring	630 ± 8	1964–1965	Chakraborty et al. (1994)
Debre Zeit, Ethiopia (9°N, 39°E)	Atm. CO_2	717 ± 6	1965	Nydal and Lövseth (1996)
Mandla, central India (22°35′N, 80°22′E)	Tree ring	705 ± 9	1965	Murphy et al. (1997)
Doi Inthanon, northern Thailand (19°N, 99°E)	Tree ring	694 ± 7	1965	Hua et al. (2000)
Hoshangabad, central India (22°30′N, 78°E)	Tree ring	708 ± 8	1965	This study

The variability in the peak $\Delta^{14}C$ values ranging from 630 to 717‰ recorded in trees from NH Zone 3 indicates regional influences on tropospheric ^{14}C distribution. Therefore, it is important to understand the factors that control the regional and temporal pattern of the $\Delta^{14}C$ variation: e.g. oceanic CO_2 evasion through ocean-atmosphere exchange, biospheric CO_2 release, and CO_2 emission from fossil fuel combustion (Randerson et al. 2002). A spatially large database is thus required to accomplish this task (Hsueh et al. 2007). It may be of significant interest to study spatio-temporal variability of ^{14}C in atmospheric CO_2 near the Indian subcontinent, as this region is affected by seasonally reversing monsoon winds and is a highly populated and industrialized area. Continuous records of ^{14}C measurements from India are scarce. To date, there exist only 2 tree-ring ^{14}C profiles from India (Chakraborty et al. 1994; Murphy et al. 1997) for the post-bomb period (1950 onwards) and no record exists prior to that. The earliest atmospheric ^{14}C measurements from India were reported by Agrawal and Kusumgar (1968), wherein data from Gulmarg (34°04′N, 74°25′E, 2745 m asl), Bombay (18°56′N, 72°51′E, at sea level, presently Mumbai) and Kodaikanal (10°15′N, 77°31′E, 2300 m asl) were reported for the period from November 1963 to April 1965. It is important to note that these ^{14}C data were not corrected for fractionation of ^{13}C. Apart from the tree-ring records, maritime atmospheric $^{14}CO_2$ measurements are available from the adjoining oceanic regions, measured dur-

ing the spring seasons between 1993 and 2001 (Bhushan et al. 1997; Dutta et al. 2006). We have undertaken a larger project to reconstruct the ^{14}C activity for the bomb period in central and southern India using tree rings. In this paper, we report some of the results of this reconstruction based on the ^{14}C analysis of a teak tree collected from Hoshangabad (22°30′N, 78°E) in central India, and compare them with the ^{14}C results from Thane (19°30′N, 74°12′E) on the west coast of India. The work on the southern India tree is underway and will be published elsewhere.

MATERIALS AND METHODS

Sample Locations

Teak trees (*Tectona grandis*) in natural forests mostly grow in central and southern parts of India (Troup 1921). They form well-defined annual growth rings (Chowdhury 1940) that have been studied extensively to relate teak growth and climate variability (Borgaonkar et al. 1994, 1996; Bhattacharyya et al. 1992; Bhattacharyya and Yadav 1999; Pumijumnog and Park 1999; Shah et. al. 2007). However, the isotopic analysis (especially carbon) of Indian teak is less studied. Chakraborty et al. (1994) carried out ^{14}C analysis of a teak sample (*T. grandis*) collected from the west coast of India to determine the air-sea gas exchange rates. Murphy et al. (1997) also analyzed teak collected from a central Indian location to determine the atmospheric bomb ^{14}C level.

Madhya Pradesh is one of the states in central India that harbors teak forest. Climate in this location is dry with annual rainfall ~1000 mm and soils that are shallow, porous, or clayey. Teak trees are present in high proportion and are often associated with *Anogeissus latifolia* and *Terminalia* spp. in mature forest (Champion and Seth 1968). Tree disks were collected from logged trees by AB and SKS from Hoshangabad (22°30′N, 78°E; Figure 1), Madhya Pradesh, in March 1997. The ring-width chronology of this tree record (labeled as HBD) was established for the period 1836–1997 (Shah et al. 2007). Here, we present ^{14}C measurements with short temporal resolution for the period of 1954–1977 to span the peak $\Delta^{14}C$ value for the bomb period.

^{14}C Analysis

The HBD sample had well-developed rings with widths in the range of 2–5 mm. ^{14}C was measured both by liquid scintillation counting (LSC) at the Birbal Sahni Institute of Palaeobotany, and by accelerator mass spectrometry (AMS) at the Center for Applied Isotope Studies (CAIS), University of Georgia, USA. For LSC analysis, 2–3 rings from the same disk were combined together to make an adequate amount of benzene. The rings were separated by hammer and chisel and crushed in a Wiley Mill. Due to their narrow width, it was not possible to separate the early- and late-wood portions of the rings; instead, whole rings were used for ^{14}C analysis. The rings were chemically pretreated according to the method of Cain and Suess (1976). The samples were pyrolyzed under vacuum to form charcoal (Leavitt et al. 1982). The charcoal was then combusted in a vacuum system in the presence of oxygen to form carbon dioxide. Subsequently, the CO_2 was converted into benzene following the standard method (Noakes et al. 1965) and counted in a Quantulus 1220 liquid scintillation counter. The 1-σ error bars reported in Figure 2 are based on counting statistics only. The same procedure was followed for the previously published Thane samples (Chakraborty et al. 1994) and maintained here as well in order to compare results. For AMS analysis, the wood samples were treated to make α-cellulose following the techniques outlined in Hua et al. (2000). The graphitization was done at the Department of Botany, University of Florida, Gainesville, USA, and at the Institute of Physics AMS Facility, Bhubaneswar, India. The results are reported in $\Delta^{14}C$ notation corrected for isotopic fractionation based on the measured or estimated $\delta^{13}C$ values, and radioactive decay since the year of growth (Stuiver and Polach 1977).

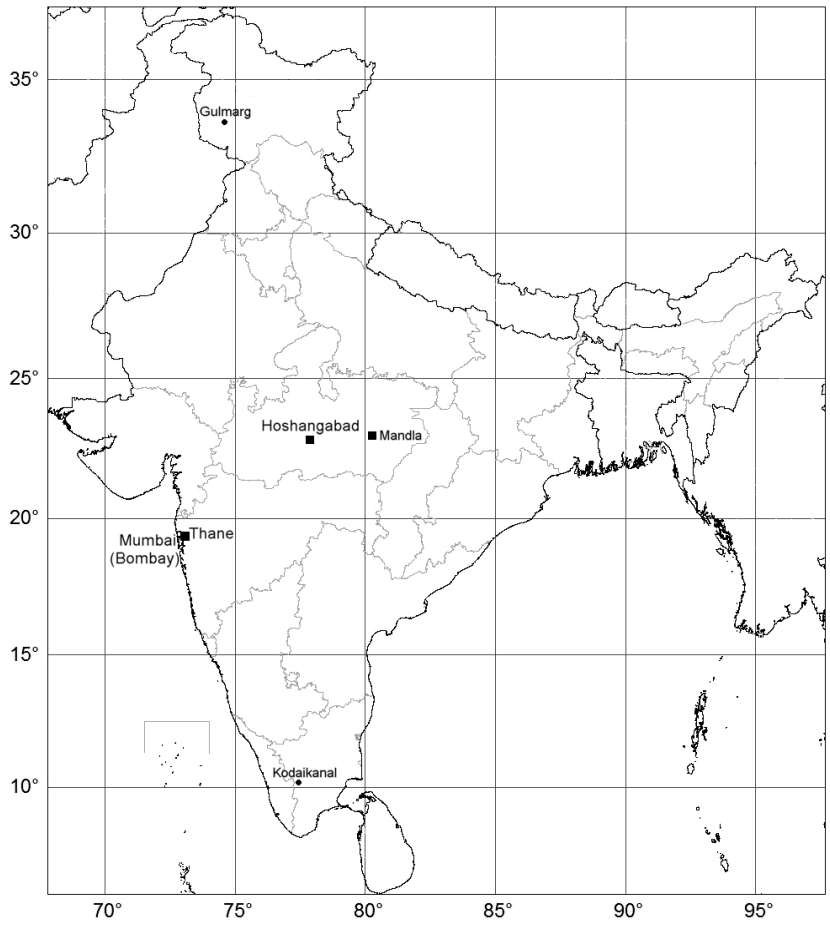

Figure 1 Locations in India of samples of tree-ring (filled squares) and atmospheric CO_2 (filled circles).

Back-Trajectory Analysis

We used back-trajectory analysis to determine the approximate source regions of air parcels that moved over the locations of the trees sampled, and to observe the general trend of air mass movements during different seasons. To determine the back-trajectories, we used the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Rolph 2003) for 10 m above ground level at 3 locations near Hoshangabad, Thane, and Mandla, using archived global NCEP Reanalysis data sets (1948 to present).

Estimation of Fossil Fuel Component

The fossil fuel component has been calculated by assuming a 2-component mixing of CO_2, i.e. fossil fuel CO_2 is mixing with the background CO_2, while assuming that photosynthetic uptake of CO_2 did not significantly alter the CO_2 partitioning in this area. The following equation was used as outlined in Levin et al. (1989):

$$C_{fossil} = C_{bkg} \times \left(\frac{{}^{14}C_{bkg} - {}^{14}C_{ms}}{{}^{14}C_{bkg}} \right) \quad (1)$$

where C_{bkg} is the CO_2 concentration in the clear air (in ppm) and $^{14}C_{bkg}$ and $^{14}C_{ms}$ are the ^{14}C concentration in clean air and the measured ^{14}C concentration for the examined region, respectively, expressed as percent modern carbon (pMC) (Stuiver and Polach 1977).

RESULTS AND DISCUSSION

The Hoshangabad (HBD) sample $\Delta^{14}C$ peak value is 708 ± 8‰ and is in very good agreement within the limits of analytical uncertainties with that of the Mandla record (Murphy et al. 1997) as well as with that of the composite $\Delta^{14}C$ profile constructed for NH Zone 3 as categorized in Hua and Barbetti (2004) (Table 2; Figure 2). These compiled results clearly show that there is a considerable variation in tropospheric ^{14}C activities within a narrow belt of 19°N–23°N and 74°E–100°E during the later half of the 20th century. Though the HBD $\Delta^{14}C$ values are not uniformly enriched relative to the Thane values, the average $\Delta^{14}C$ of HBD between 1964 and 1965 (657 ± 13‰) is higher by 27 ± 15‰ than that of Thane (630 ± 8‰) for the same period. Apparently, this enrichment is manifested more during the peak when the bomb-produced ^{14}C enhanced the effect than at other times. Here, we discuss the possible reasons for these variations and their implications, mainly in the context of comparing the Hoshangabad and Thane records, which are from a similar region but differ by ~27‰. The possible reasons for this difference could be partly due to the averaging effect because

Table 2 ^{14}C data of teak tree-ring samples from peninsular India.

Lab #	Sample	Years	Method	$\Delta^{14}C$ (‰) ±1 σ
Hoshangabad (HDB) tree rings				
	HBD-TEGR-005455	1954–1955	AMS	47 ± 5
	HBD-TEGR-565758	1956–1958	LSC	35 ± 5
	HBD-TEGR-005960	1959–1960	AMS	102 ± 6
	HBD-TEGR-006162	1961–1962	LSC	215 ± 5
	HBD-TEGR-006300	1963	AMS	534 ± 8
	HBD-TEGR-006400	1964	AMS	605 ± 10
	HBD-TEGR-000065	1965	AMS	708 ± 8
	HBD-TEGR-006800	1968	LSC	518 ± 18
	HBD-TEGR-000077	1977	LSC	313 ± 9
Thane (Th) tree rings[a]				
CH-191	Th-17	1960	LSC	238 ± 6
CH-190	Th-16	1961	LSC	260 ± 6
CH-187	Th-15	1962	LSC	338 ± 6
CH-186	Th-14	1963	LSC	565 ± 8
CH-185	Th-13	1964–1965	LSC	630 ± 8
CH-184	Th-12	1966	LSC	587 ± 8
CH-183	Th-11	1967	LSC	560 ± 7
CH-182	Th-10	1968	LSC	534 ± 7
CH-181	Th-9	1969–1970	LSC	476 ± 7
CH-180	Th-8	1971–1972	LSC	434 ± 7
CH-179	Th-7	1973	LSC	400 ± 7
CH-178	Th-6	1974	LSC	420 ± 7
CH-177	Th-5	1975	LSC	354 ± 6
CH-176	Th-4	1976	LSC	340 ± 7
CH-173	Th-3	1977	LSC	311 ± 6
CH-172	Th-2	1978	LSC	299 ± 6
CH-171	Th-1	1979–1980	LSC	260 ± 7

[a]Data from Chakraborty et al. (1994) and Bhushan et al. (1994).

2 rings were combined for LSC analyses in the case of 1964–65, 1969–70, and 1971–72 samples. This averaging will cause a reduction of ~12‰ of the bomb ^{14}C peak (of 1964–65 averaged sample) from its maximum value of 705 ± 6‰ during 1965 (Hua and Barbetti 2004). Another likely possibility is regional fossil fuel emissions that could lower the ^{14}C content of the regional air. Thane belongs to the industrial belt of Mumbai and so could be influenced by enormous amounts of industrial emissions. Fossil fuel emission plays a significant role in controlling the $\Delta^{14}C$ of local/regional atmospheric CO_2 (Levin and Kromer 1997; Randerson et al. 2002; Levin et al. 2003; Turnbull et al. 2006; Hsueh et al. 2007). During the late 1960s and early 1970s, with atmospheric CO_2 concentration of ~320 ppm, the addition of 1 ppm of fossil fuel-derived CO_2 (with $\Delta^{14}C$ –1000‰) would have caused a depletion in atmospheric $\Delta^{14}C$ of ~5‰ from its background value of 705‰. In this region of India, the contribution of fossil fuel CO_2 has not been quantified yet from $\Delta^{14}C$ or the $\delta^{13}C$ analysis of tree rings. Dutta et al. (2006) demonstrated that the air from the South Asian continent introduces a significant amount of fossil CO_2 into the clean air over the Indian Ocean region through meridional transport. The effect of this transported CO_2 observed over the Arabian Sea could reduce the atmospheric $\Delta^{14}C$ by up to 8.1‰ relative to the background value (Dutta et al. 2006).

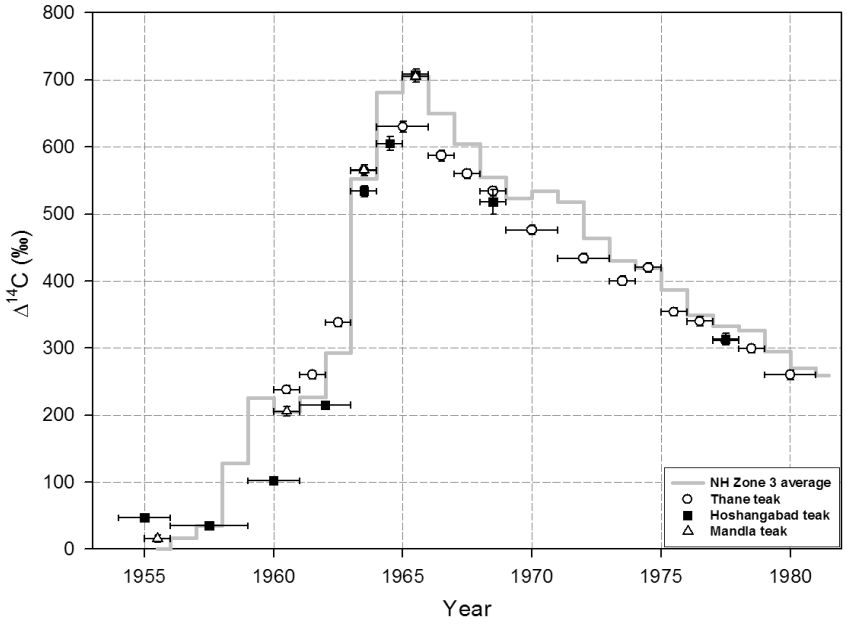

Figure 2 Atmospheric $\Delta^{14}C$ time series from ^{14}C in teak tree rings from Hoshangabad (filled squares, this study), Thane (open circles; Chakraborty et al. 1994), and Mandla (open triangles; Murphy et al. 1997), compared with the annual average atmospheric $\Delta^{14}C$ of Northern Hemisphere Zone 3 (thick gray line; Hua and Barbetti 2004).

Another potential mechanism that may deplete the atmospheric $\Delta^{14}C$ near the west coast of India is oceanic upwelling. It is known that the Arabian Sea is a perennial source of oceanic CO_2 to the atmosphere, since the partial pressure of CO_2 of surficial Arabian Sea water is higher than that of the atmosphere (Takahashi 1989). Sarma et al. (1998) estimated an average flux of CO_2 from the Arabian Sea of ~45 Tg C yr^{-1}. During the mid-1960s, oceanic CO_2 released from the surficial Arabian Sea was significantly depleted in ^{14}C relative to that of the atmosphere. During 1964–65, the difference in $\Delta^{14}C$ of atmospheric CO_2 (708 ± 8‰) and that of surface Arabian Sea (120 ± 7‰, as measured in a coral from the northern Arabian Sea, reported by Chakraborty et al. 1994), was

588 ± 10‰. Thus, during the mid-1960s, oceanic evasion of CO_2 could have potentially decreased terrestrial atmospheric $\Delta^{14}C$ records near the Arabian Sea coasts, providing that sufficient contact time existed for ocean-atmosphere CO_2 exchange. Atmospheric $\Delta^{14}C$ records from coastal regions are reported to be influenced by coastal upwelling, as observed in pre-bomb tree rings (Damon et al. 1999) and atmospheric $^{14}CO_2$ measurements in the post-bomb period (Rozanski et al. 1995). The back-trajectory analysis shows that during the summer (May–Sep), which encompasses the main growing season of the tree rings, the predominant flow of wind was from the middle of the Arabian Sea towards peninsular India. Intense upwelling takes place in the central Arabian Sea during the summer (or southwest) monsoon, which may potentially influence the atmospheric carbon isotopic signatures through evasion of oceanic CO_2. High wind speeds over the Arabian Sea during the summer monsoon season (Jun–Sep) result in a given mass of air moving over the Arabian Sea within just ~3–4 days (Figure 3a). The short residence time of air over the Arabian Sea during the southwest monsoon season excludes the possibility of any significant isotopic change in the atmospheric CO_2. During the winter and early spring seasons (Dec–Feb), the winds blow mainly over the continents (Figure 3b). However, winter atmospheric ^{14}C signatures may not be well recorded by these tropical trees, which mainly grow during summer seasons. From monthly atmospheric $\delta^{13}C$ measurements in flask samples from Cape Rama, Goa (15°05′N, 73°50′E, 60 m asl) on the west coast of India, Bhattacharya et al. (2002) demonstrated changes in isotopic composition related to seasonally reversing winds and biogenic CO_2 emission, but excluded strong marine influence.

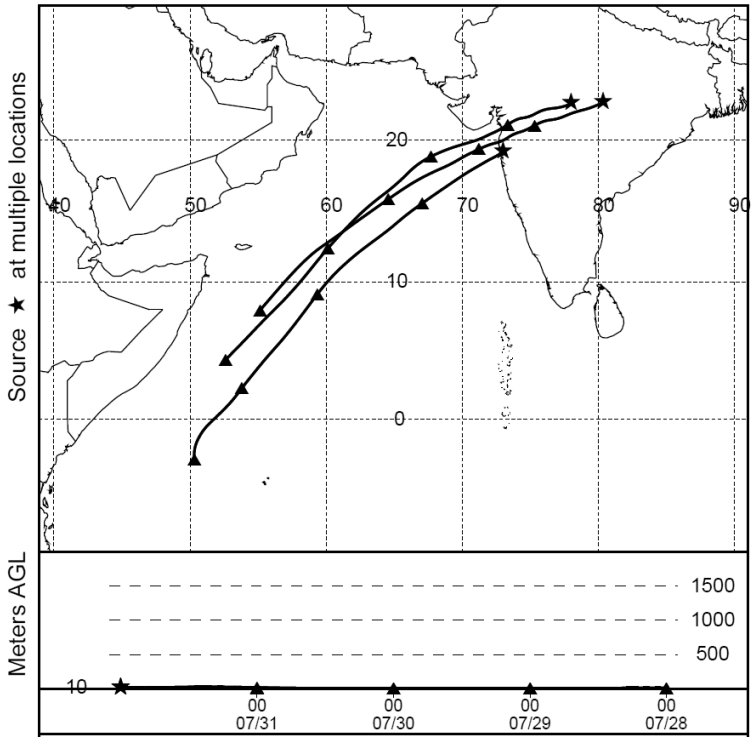

Figure 3a Four-day wind back-trajectory plots between 28 July and 1 August 1963 (summer), produced by HYSPLIT (Draxler and Rolph 2003). Air movement is from the Arabian Sea towards India. The stars denote the locations of Thane, Hoshangabad, and Mandla. Latitudes and longitudes are marked on the grids. The bottom panel shows the temporal variation of the heights of the air masses in meters above ground level (AGL), initial heights being zero for all.

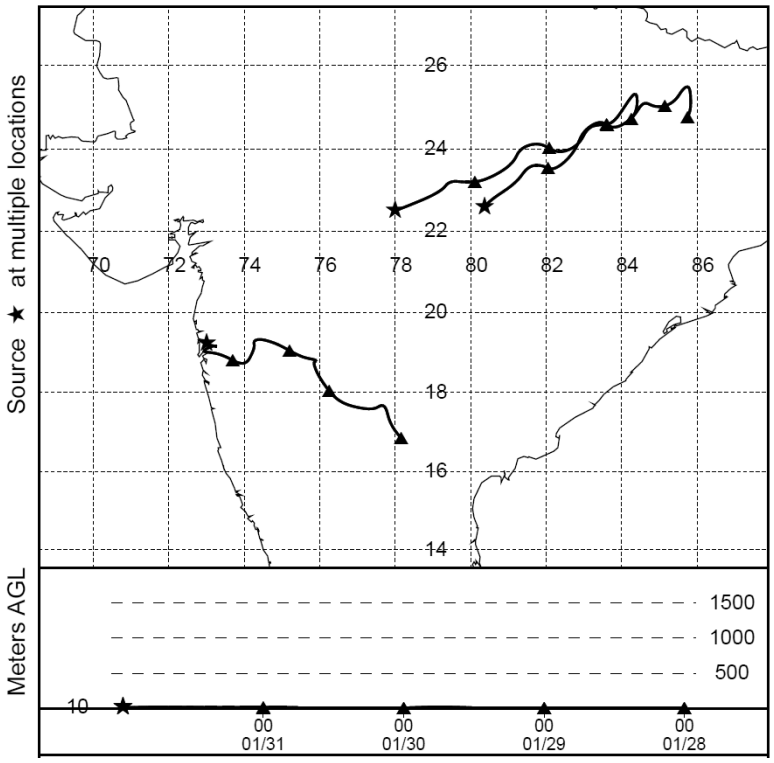

Figure 3 Same as in Figure 3a, but between 30 January and 1 February 1963 (winter time). Air circulation is over the continent.

The anomalously low $\Delta^{14}C$ observed in the Thane record is therefore most likely due to the regional effects of fossil fuel CO_2 emission, as Thane is close to the highly industrialized suburbs of Mumbai. It is thus possible to use this anomaly to estimate the local flux of CO_2 depleted in ^{14}C that is responsible for lowering the $\Delta^{14}C$ peak using Equation 1. The difference in peak value was calculated by taking the difference of the weighted average values of 1964 and 1965 for the HBD tree and the Thane peak value (already an average of these 2 yr). This difference is equal to 27 ± 15‰. We then estimated the fossil fuel CO_2 (C_{fossil}) using the Thane tree-ring ^{14}C record (as $^{14}C_{ms}$), the global average CO_2 concentration (Keeling et al. 2001) (as C_{bkg}), and the HBD ^{14}C data (as $^{14}C_{bkg}$). The fossil fuel CO_2 component estimated in the tropospheric air of Thane between 1963 and 1968 varied from 0.8% to a maximum of 3.8% of the background CO_2 concentration, with a mean value of 2.3 ± 1.2% (1 σ).

From ^{14}C measurements in annual crops of corn across the North America for 2004, Hsueh et al. (2007) concluded that the contribution of fossil fuel-derived CO_2 near urban areas can be up to an order of magnitude larger relative to unpolluted rural regions. Hsueh et al. (2007) estimated fossil fuel CO_2 concentration was higher by 2.7 ± 1.5 ppm and 4.3 ± 1.0 ppm relative to the background value of 378 ppm during 2004, for the eastern United States and the Ohio-Maryland region, respectively. This translates to fossil fuel CO_2 contributions of about 0.7 ± 0.3% and 1.1 ± 0.2%, respectively, of the background atmospheric CO_2. Turnbull et al. (2006) estimated fossil fuel CO_2 of 0 to ~5.5 ppm from $^{14}CO_2$ measurements during 2004 in the boundary layer of New England in the northeastern United States. Our estimated fossil fuel CO_2 contribution at Thane, near the highly urban area of Mumbai, is of similar magnitude relative to the less urban Hoshangabad.

CONCLUSION

^{14}C analysis of the annual rings of a teak tree collected from central India recorded a peak atmospheric $\Delta^{14}C$ value of 708 ± 8‰ during 1965. This value was in excellent agreement with measurements reported by other investigators for this region. However, peak $\Delta^{14}C$ recorded in 1964–65 tree rings that grew at Thane near Mumbai on the west coast of India was lower by ~27‰. This depletion in $\Delta^{14}C$ was attributed to the local fossil fuel emission from the neighboring industrial area of Mumbai. The anomaly in $\Delta^{14}C$ was used to calculate the local emission of fossil fuel CO_2, which was estimated to be 2.3 ± 1.2% of the contemporary ambient atmospheric CO_2 concentration.

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