Influence of atmospheric circulation on regional $^{14}CO_2$ differences

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Received 10 August 2006; revised 15 December 2006; accepted 16 December 2006; published 4 October 2007.

[1] Detailed analyses of published ^{14}C data from tree rings and atmospheric CO_2 samples for the northern tropics in Asia (India, Thailand, and Vietnam) and Africa (Ethiopia) have been performed for the heavily bomb-influenced period 1963–1967 A.D. The results show that the Asian summer monsoon and Intertropical Convergence Zone (ITCZ) position influenced atmospheric $^{14}CO_2$ over the study area. Similar analyses of atmospheric records for northern and western Europe, northwestern Africa, and the northeastern United States and tree ring data for east Asia show that the Northern Hemisphere distribution of bomb ^{14}C for 1963–1967 depended on atmospheric circulation controlled by the seasonal positions of Hadley cell boundaries and the ITCZ. The distribution of ^{14}C did not have a simple latitudinal dependence. This work shows that the seasonal atmospheric circulation patterns are crucial for the description of atmospheric ^{14}C gradients during the bomb peak period. These principles can be applied to the interpretation of the small intrahemispheric ^{14}C offsets of the remote past.

Citation: Hua, Q., and M. Barbetti (2007), Influence of atmospheric circulation on regional $^{14}CO_2$ differences, *J. Geophys. Res.*, *112*, D19102, doi:10.1029/2006JD007898.

1. Introduction

[2] Natural radiocarbon or ^{14}C is formed continuously in the atmosphere by interactions of the secondary neutron flux with atmospheric ^{14}N. About 55% of ^{14}C is produced in the stratosphere and 45% in the upper troposphere [*Gäggeler*, 1995]. Following its production, ^{14}C is oxidized to form atmospheric ^{14}CO and then $^{14}CO_2$. Note that throughout this paper we use atmospheric ^{14}C as a shortened form of atmospheric $^{14}CO_2$.

[3] It has been long assumed that, for any given time period, atmospheric ^{14}C is homogeneously distributed throughout the lower troposphere [*Libby*, 1955]. This allows us to use a single universal data set for the calibration of radiocarbon dates. However, *Lerman et al.* [1970] were the first to note a difference in natural atmospheric ^{14}C between the two hemispheres when they measured ^{14}C in tree rings at different locations. Recent studies have confirmed the finding of Lerman and colleagues, and also reported interhemispheric and intrahemispheric ^{14}C offsets and temporal variations thereof [*Vogel et al.*, 1993; *Stuiver and Braziunas*, 1998; *McCormac et al.*, 1998; *Kromer et al.*, 2001; *Barbetti et al.*, 2004].

[4] In previous papers, we have investigated regional atmospheric ^{14}C in the tropics during the latter part of the Little Ice Age (LIA) and 1938–1954 A.D. using cross-dated tree rings from north-central and northwestern Thailand, respectively [*Hua et al.*, 2004a, 2004b]. For both periods, the ^{14}C results from Thai tree rings clearly followed the trend of Southern Hemisphere ^{14}C levels and we concluded that atmospheric ^{14}C over Thailand during the growing season of the tree rings was strongly influenced by the entrainment of Southern Hemisphere air parcels in the southwest Asian summer monsoon. As discussed by *Hua et al.* [2004b], during the summer of the Northern Hemisphere the Intertropical Convergence Zone (ITCZ) moves northward (up to 30°N or more) over the Asian continent and inhibits Northern Hemisphere air masses from entering Thailand and southern Asia (because winds at the ITCZ are commonly light or nonexistent [*Linacre and Geerts*, 1997; *McGregor and Nieuwolt*, 1998]). The air mass over these regions during the summer months is mainly a mixture of Southern Hemisphere winter air carried north by the southwest Asian monsoon, and Northern Hemisphere winter air which flowed into southern Asia a few months earlier.

[5] A large amount of ^{14}C was artificially produced when hundreds of nuclear weapons were detonated in the atmosphere between 1945 and 1980. Nuclear bomb blasts produced intense fluxes of thermal neutrons, which in turn reacted with atmospheric ^{14}N to form ^{14}C. Most of the detonations occurred in the northern atmosphere in the late 1950s and early 1960s. They caused a dramatic increase in the concentration of ^{14}C and created large ^{14}C disequilibria within the troposphere (e.g., north versus south, high versus low latitudes) during the bomb peak period (1963–1967 A.D.) [*Nydal and Lövseth*, 1983; *Manning et al.*, 1990; *Levin and Hesshaimer*, 2000], as depicted in Figure 1. Bomb ^{14}C significantly increased the contrast between different air masses during the bomb peak period. With the above scenario of air mixing over Thailand, any

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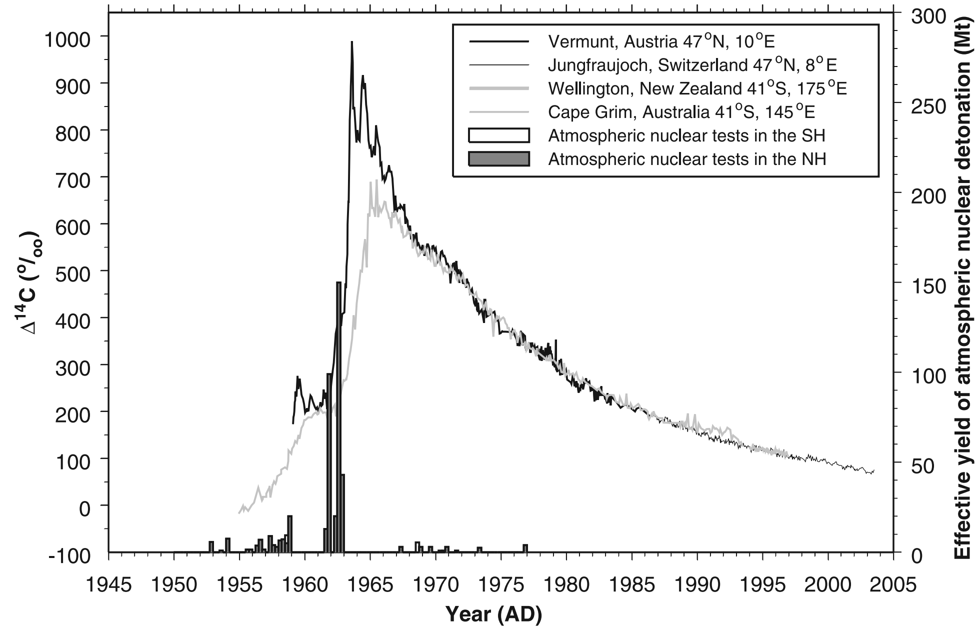


Figure 1. Atmospheric ^{14}C for the past 50 years and the magnitude of atmospheric nuclear detonation. Lines represent atmospheric ^{14}C data. Data sources are *Levin and Kromer* [2004] for Vermunt and Jungfraujoch, *Manning and Melhuish* [1994] for Wellington, and *Levin et al.* [1996, 1999] for Cape Grim. Bars represent effective yields of atmospheric nuclear detonations for 3-month periods [*Enting*, 1982].

contribution of Southern Hemisphere air masses (containing much less ^{14}C than those from the Northern Hemisphere during the bomb peak) carried by the southwest Asian monsoon to southern Asia and adjacent areas is expected to cause a significant decrease in atmospheric ^{14}C over these regions during the summer monsoon period.

[6] Therefore, in this paper, we use published bomb ^{14}C data for the northern tropics to test the above hypothesis for tropical Asia and Africa. We then expand our investigation to examine possible influences of atmospheric circulation on regional atmospheric ^{14}C during the bomb peak period for northern midlatitudes in the North Atlantic region and in east Asia, based on the existing records of tree ring and atmospheric ^{14}C.

2. Monsoon Circulation and Atmospheric ^{14}C in the Northern Tropics

2.1. African/Asian Monsoons and Atmospheric ^{14}C Over Tropical Africa

[7] Short atmospheric ^{14}C records from Dakar (Senegal, 15°N, 17°W) and Debre Zeit (Ethiopia, 9°N, 39°E) [*Nydal and Lövseth*, 1996] for the bomb peak are plotted in Figures 2a and 2b for comparison. Note that in this paper, atmospheric ^{14}C levels are expressed as $\Delta^{14}C$ values, after corrections for isotopic fractionation using $\delta^{13}C$ and radioactive decay [*Stuiver and Polach*, 1977]. The issue arising here is why was there a significant ^{14}C gradient during the bomb peak between Dakar (15°N, 17°W) and Debre Zeit (9°N, 39°E), which are not much different in latitude? For example, during the 1963–1964 summers, some individual ^{14}C differences between Dakar and Debre Zeit reach a level of 150‰ (Dakar is higher; see Figure 2b).

[8] During the bomb peak period, there was a large ^{14}C gradient between the troposphere and other carbon reservoirs, causing a large flux of excess ^{14}C from the atmosphere to the oceans and terrestrial biosphere (~32% of excess ^{14}C produced by atmospheric nuclear detonations was taken up by the two carbon reservoirs in the period 1963–1967 [*Naegler and Levin*, 2006]). These fluxes might contribute to regional atmospheric ^{14}C offsets, and cause $\Delta^{14}C$ differences between Dakar and Debre Zeit for the bomb peak period in particular. The air-sea exchange flux caused ^{14}C differences between land and sea for the period immediately following nuclear detonations. By using a regional carbon cycle model, *Randerson et al.* [2002] calculated atmospheric $\Delta^{14}C$ anomalies arising from air-sea exchange for the year 1965. They reported an atmospheric $\Delta^{14}C$ anomaly of up to −7‰ for coastal stations in northwestern Africa (e.g., Dakar) relative to land stations in northeastern Africa (e.g., Debre Zeit). This effect, which would generate a negative ^{14}C gradient between Dakar and Debre Zeit, cannot be responsible for large positive ^{14}C differences between the two stations during the bomb peak period. The atmosphere–terrestrial biosphere exchange, during the bomb peak caused atmospheric ^{14}C offsets between land and ocean, and between lands having different residence times of carbon in terrestrial systems. However, *Randerson et al.* [2002] reported no significant atmospheric $\Delta^{14}C$ anomalies for coastal regions in northwestern Africa such as Dakar and land stations in northeastern Africa such as Debre Zeit for the year 1965. This terrestrial biospheric flux was not responsible for the large ^{14}C gradient between Dakar and Debre Zeit during the bomb peak as shown in Figures 2a and 2b. Later in this section, atmospheric ^{14}C records from other stations in northwestern Africa and

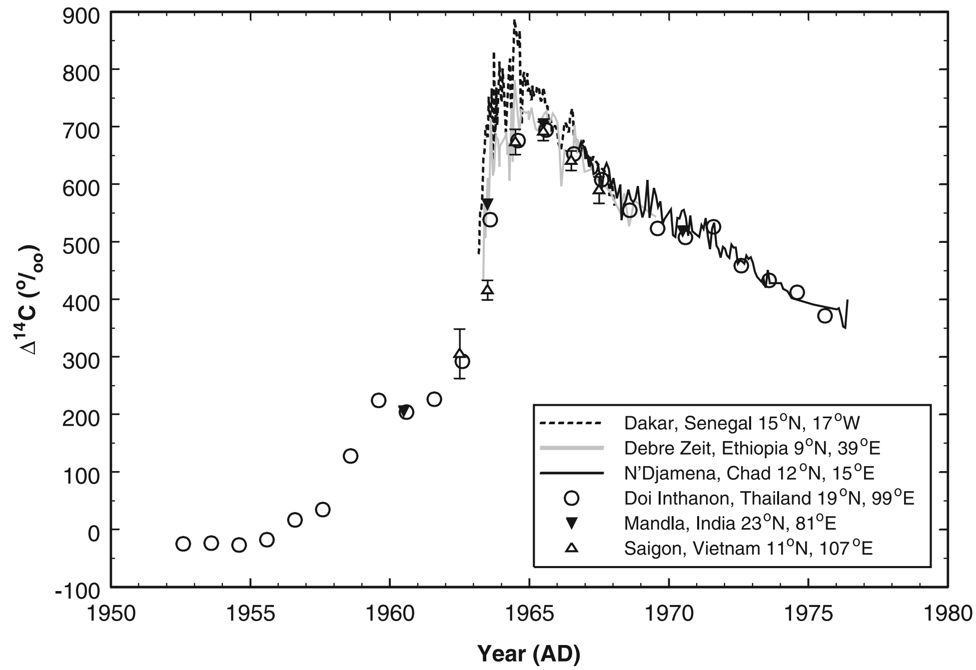


Figure 2a. Radiocarbon in tree rings versus atmospheric ^{14}C at similar latitudes for the northern tropics. Lines represent atmospheric ^{14}C data. Symbols depict measured $\Delta^{14}C$ values for tree rings. Error bars for Mandla and Doi Inthanon are too small to be shown. Error bars for Saigon are 1σ. Data sources are *Nydal and Lövseth* [1996] for Dakar, N'Djamena, and Debre Zeit; *Murphy et al.* [1997] for Mandla; *Hua et al.* [2000, 2004a] for Doi Inthanon; and *Kikata et al.* [1992, 1993] for Saigon.

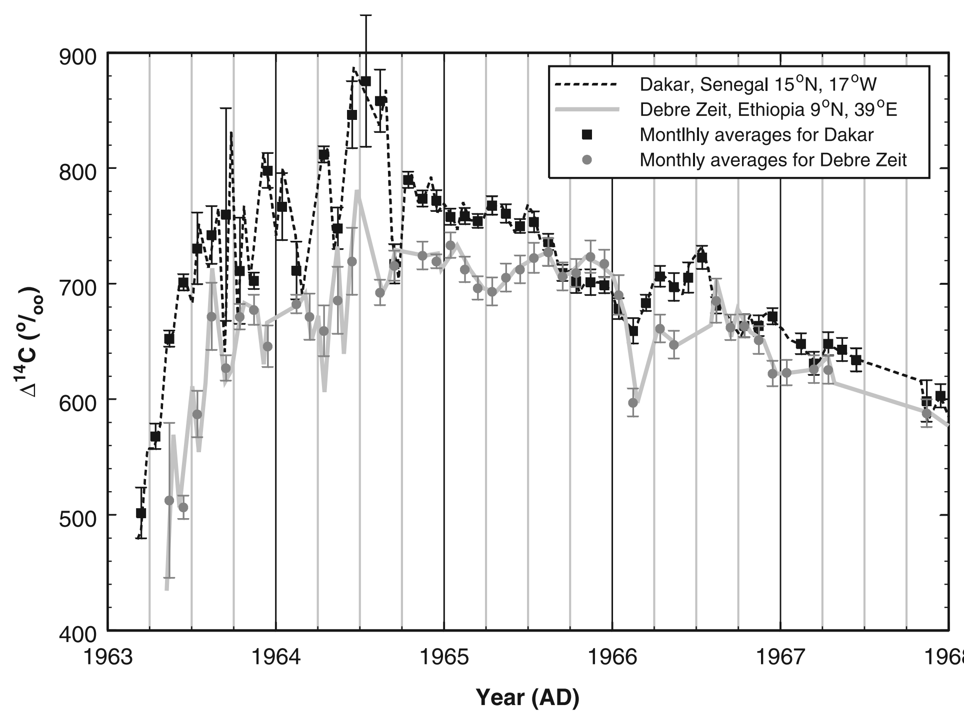


Figure 2b. Close-up of atmospheric ^{14}C records for Dakar and Debre Zeit during the bomb peak period. Lines represent measured atmospheric $\Delta^{14}C$ data. Symbols depict their monthly average values. Error bars are 1σ.

northwestern Europe will be discussed together with Dakar and Debre Zeit. During the bomb peak period, the air-sea exchange flux caused either no significant atmospheric Δ^{14}C differences between these stations or a negative north-south ^{14}C gradient [*Randerson et al.*, 2002], which is opposite to the positive north-south ^{14}C gradient of air mass origins discussed in this section. Similarly, during the bomb peak period the atmosphere–terrestrial biosphere exchange caused no significant anomalies for northern low latitudes to midlatitudes covering most of our study regions because of similar carbon cycling rates within terrestrial ecosystems of these regions. An atmospheric Δ^{14}C anomaly of up to −20‰ was reported for high latitudes relative to low latitudes and midlatitudes because of cooler temperatures and slower turnover time for terrestrial systems in the former regions [*Randerson et al.*, 2002]. Again, this negative north-south ^{14}C gradient is opposite to the positive north-south ^{14}C gradient of air mass origins discussed in this section. Therefore regional ^{14}C offsets due to oceanic and terrestrial biospheric fluxes during the bomb peak period can be neglected in the following discussion.

[9] The significant differences between Dakar and Debre Zeit might be due to the summer African/Asian monsoon, and the position of the ITCZ during the summer time relative to the locations of the two stations. During the boreal winter, the ITCZ moves southward (up to ~15°S) as shown in Figure 3. Both Dakar and Debre Zeit are located well to the north of the winter ITCZ position and therefore receive Northern Hemisphere air coming from midlatitudes and high latitudes. However, during the summer of the Northern Hemisphere, the latitude of the ITCZ over Africa is ~15°N and the two stations might be on different sides of the ITCZ (Figure 3). Dakar might be located north of the summer ITCZ and might continue to receive air masses from northern midlatitudes and high latitudes. Meanwhile, Debre Zeit is located south of the summer ITCZ and enjoys Southern Hemisphere air carried by the summer African monsoon (for eastern Africa this summer monsoon is actually a part of the southwest Asian monsoon [*Linacre and Geerts*, 1997; *McGregor and Nieuwolt*, 1998]). Thus atmospheric ^{14}C over Debre Zeit might be influenced by Southern Hemisphere air, with a ^{14}C level significantly lower than that for the northern midlatitudes and high latitudes during the bomb peak. A small difference in latitude between the two stations might nevertheless permit a large ^{14}C gradient between them in summer. If the above scenario is valid for the bomb peak, one can expect a large difference in ^{14}C between the two stations for summer months, and a much smaller difference for winter months. During the bomb peak, however, there was a large ^{14}C gradient in the Northern Hemisphere in summer caused by a significant injection of excess ^{14}C from the stratosphere into the troposphere in high latitudes in spring and summer. One may therefore argue that possible larger differences between Dakar and Debre Zeit for summer months compared to those for winter months during the bomb peak, are due to the latter effect rather than due to influence of the southwest Asian monsoon.

[10] In order to test the above two hypotheses, mean differences between several stations in the Northern Hemisphere for two different seasons, summer (May–August) and winter (November–February) of the Northern Hemisphere, were calculated for the period 1963–1970 and are presented in Figure 4. Differences between Fruholmen (Norway, 71°N, 24°E), in the high-latitude zone where injection of excess ^{14}C from the stratosphere occurred, and stations in low latitudes to midlatitudes north of the summer ITCZ position (Santiago de Compostela, Mas Palomas, Izaña and Dakar) are shown in Figure 4a. Differences between stations in low latitudes to midlatitudes north of the summer ITCZ and Debre Zeit are illustrated in Figure 4b. To avoid a possible problem of interlaboratory offsets in ^{14}C measurement, which may create anomalies in the results of the following analyses, the atmospheric ^{14}C data sets employed in Figure 4 were chosen because they were measured in the same laboratory of *Nydal and Lövseth* [1996]. A large north-south ^{14}C gradient is obvious for 1964, and there is a larger difference between Fruholmen and stations from low latitudes to midlatitudes north of the summer ITCZ position, for summer compared to that for winter (Figure 4a). To simplify our analyses, summer and winter in Figure 4 were respectively defined as May–August and November–February, although the ^{14}C seasonal variations of the records presented in Figure 4 are not always in phase because of the seasonality of the transport of bomb ^{14}C from north to south [*Enting and Mansbridge*, 1987]. This simplification might be the reason for the scatter of the data in summers (e.g., summer 1965 in particular), and for not seeing the north-south ^{14}C gradient in 1963 as summertime differences shown in Figure 4a are equal to or less than wintertime differences. However, data for the differences between Fruholmen and low latitudes to midlatitudes (north of the summer ITCZ position) have similar trends. On average, the magnitude of the (summer-winter) difference for 1964 is 35 ± 23‰. This summer-winter difference becomes smaller for 1965 and 1966.

[11] There are large differences between stations in low latitudes to midlatitudes north of the summer ITCZ and Debre Zeit for summer months compared to that for winter months for 1963 and 1964 (Figure 4b), when there was a significant difference between northern and southern air masses in terms of ^{14}C. On average, the magnitude of this (summer-winter) difference is 85–90‰. When the north-south ^{14}C gradient was reduced, this summer-winter difference becomes smaller, e.g., for 1965. As only one 7-day integrated CO_2 sample from Debre Zeit was measured for summer 1966, no reliable comparison between low latitudes to midlatitudes and Debre Zeit is available for summer 1966 (Figure 4b). The pattern of summer-winter differences in Figure 4b is similar to that in Figure 4a. However, on average the summer-winter difference in Figure 4b (85 ± 23‰) is significantly larger than that in Figure 4a (35 ± 23‰) for 1964. Even for 1965, when the north-south ^{14}C gradient was reduced, the summer-winter difference in Figure 4b (35 ± 39‰) is slightly higher than that in Figure 4a (27 ± 39‰). These analyses indicate that larger summertime differences in ^{14}C between stations in low latitudes to midlatitudes (north of the summer ITCZ position) and Debre Zeit, and smaller differences in wintertime for 1963–1965, are unlikely to be due solely to the enhanced summertime gradient of atmospheric ^{14}C caused by the injection of excess ^{14}C from the stratosphere, especially given the greater distance from the injection zone. It is much more likely that Debre Zeit was strongly influenced by

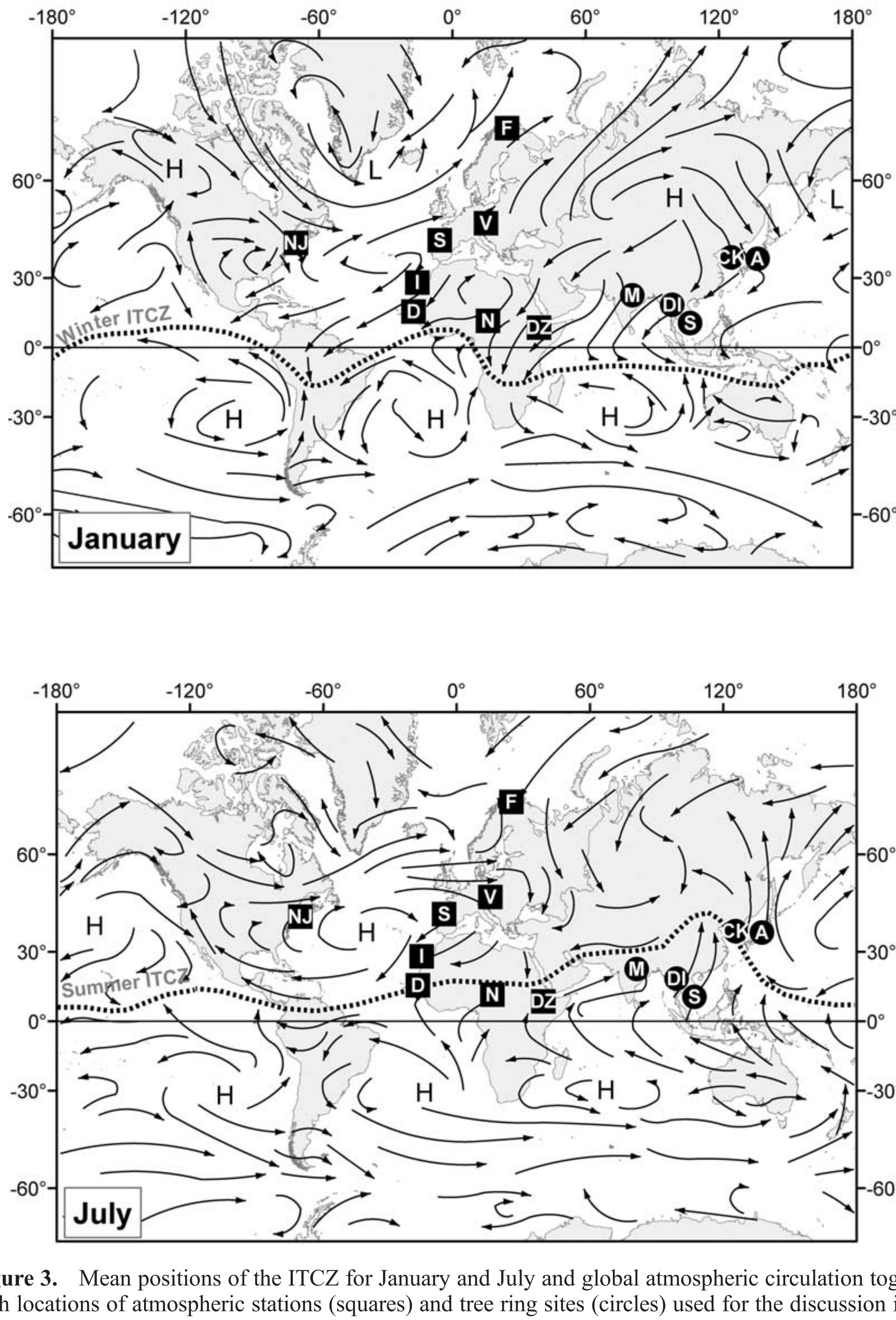


Figure 3. Mean positions of the ITCZ for January and July and global atmospheric circulation together with locations of atmospheric stations (squares) and tree ring sites (circles) used for the discussion in the text. H is a center of high atmospheric pressure, and L is a center of low atmospheric pressure. Atmospheric stations indicated are F, Fruholmen; V, Vermunt; S, Santiago de Compostela; I, Izaña; D, Dakar; NJ, New Jersey; N, N'Djamena; and DZ, Debre Zeit. Tree ring sites indicated are A, Agematsu; CK, Mounts Chiak and Kyeryong; DI, Doi Inthanon; M, Mandla; and S, Saigon. Adapted from *Linacre and Geerts* [1997]. Copyright 1997 Taylor and Francis. Reproduced by permission of Taylor and Francis Books UK.

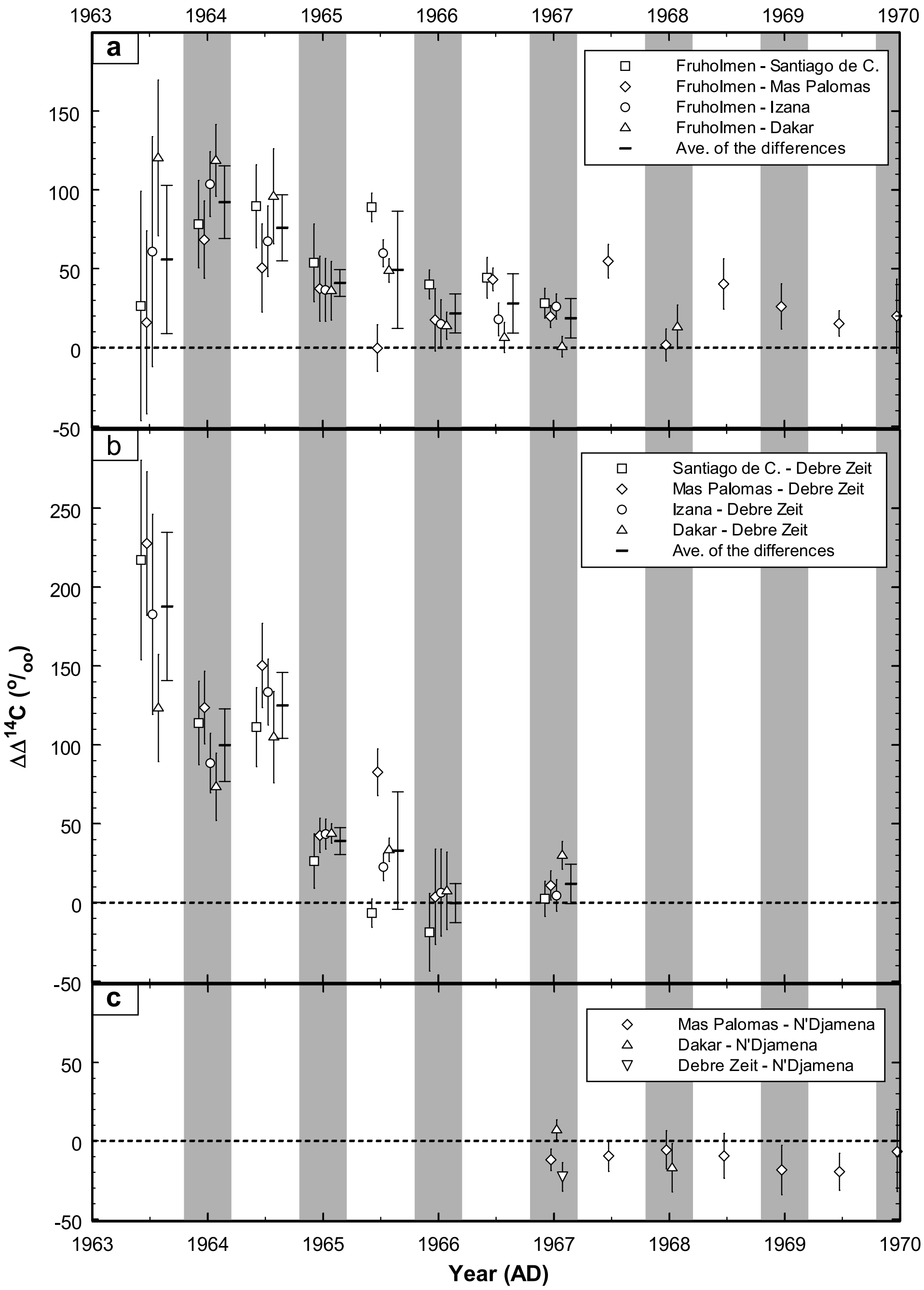


Figure 4. Differences in $\Delta^{14}C$ between sites in the Northern Hemisphere for two different seasons, summer (May–August) and winter (November–February): (a) Fruholmen versus northern low latitudes to midlatitudes north of the summer ITCZ, (b) northern low latitudes to midlatitudes north of the summer ITCZ versus Debre Zeit, and (c) northern low latitudes to midlatitudes versus N'Djamena. All symbols should be plotted in the middle of each year (for summers) and at the beginning of each year (for winters). However, they are displayed in the diagram with small temporal offsets for reason of clarity. Error bars are 1σ. Gray stripes depict winters of the Northern Hemisphere.

Southern Hemisphere air carried by the southwest Asian monsoon.

[12] There is an atmospheric ^{14}C record in north-central Africa from late 1966 to mid 1976: N'Djamena (Chad) at 12°N, 15°E reported by *Nydal and Lövseth* [1996] (see Figure 2a). In order to see whether N'Djamena was influenced by Southern Hemisphere air masses during the mid to late 1960s, the mean differences in ^{14}C between stations in low latitudes to midlatitudes and this station for the two different seasons were calculated for the period 1966–1970 and are presented in Figure 4c. However, there are short overlaps for Debre Zeit–N'Djamena and Dakar-N'Djamena.

Only one valid comparison between Mas Palomas and N'Djamena for the two seasons is available. The mean difference between the two stations for summer of -13 ± 6‰ is similar to that for winter of -11 ± 6‰ (N'Djamena values are higher in both cases). With only one valid comparison between Mas Palomas and N'Djamena, with the overlapping time of the two records being from October 1966 to November 1969 when the north-south ^{14}C gradient was reduced, no conclusion about the influence of Southern Hemisphere air masses on atmospheric ^{14}C at N'Djamena for the above period can be reliably inferred.

2.2. Asian Monsoons and Atmospheric ^{14}C Over Southern Asia

[13] Published ^{14}C records from tree rings at Mandla (India, 23°N, 81°E [*Murphy et al.*, 1997]), Doi Inthanon (Thailand, 19°N, 99°E [*Hua et al.*, 2000, 2004a]) and Saigon (Vietnam, 11°N, 107°E [*Kikata et al.*, 1992, 1993]) are also plotted in Figure 2a for comparison with atmospheric ^{14}C records from Dakar (Senegal, 15°N, 17°W) and Debre Zeit (Ethiopia, 9°N, 39°E). The tree ring data match each other very well (except the 1963 datum for Saigon). They also match very well with ^{14}C data for Debre Zeit, but all of them are much lower than the atmospheric data for Dakar. For example, the difference in ^{14}C between Dakar and Thailand is up to 150‰ for the 1964 summer (Figure 2a). It may well be asked why ^{14}C levels during the bomb peak at 19°–23°N in southern Asia are similar to those at 9°N in northeastern Africa and at 11°N in Southeast Asia (equatorial region), and much lower than those at 15°N in northwestern Africa. Our explanation is that Mandla, Doi Inthanon and Saigon (tree ring sites) are located well south of the mean summer position of the ITCZ (Figure 3), and the air mass over these sites during the growing season of tree rings is influenced by Southern Hemisphere air masses carried by the southwest Asian monsoon. During the bomb peak, these air masses containing much less ^{14}C than those from the Northern Hemisphere cause reduced atmospheric ^{14}C over India and Southeast Asia during the growing season of tree rings as shown in Figure 2a.

2.3. Estimate of the Contribution of Southern Hemisphere Air Masses to Northern Sites South of the Summer ITCZ During the Bomb Peak Period

[14] With the availability of atmospheric ^{14}C records for northern low latitudes to midlatitudes north of the summer ITCZ (Mas Palomas, Izaña and Dakar [*Nydal and Lövseth*, 1996]) and southern low latitudes (Suva, Fiji, 18°S, 178°E [*Manning et al.*, 1990]; and Fianarantsoa, Madagascar, 21°S, 47°E [*Nydal and Lövseth*, 1996]), and ^{14}C data from atmospheric and tree ring samples for the regions influenced by the southwest Asian monsoon (Debre Zeit [*Nydal and Lövseth*, 1996], Mandla [*Murphy et al.*, 1997], Doi Inthanon [*Hua et al.*, 2000] and Saigon [*Kikata et al.*, 1992, 1993]), one can estimate the contribution of Southern Hemisphere air masses to northern tropical sites south of the summer ITCZ for summer months during the bomb peak period.

[15] To simplify our calculation, we assume that:

[16] First, oceanic and terrestrial biospheric sources and sinks are small and can be neglected in our calculation. As mentioned at the beginning of this section, there was no significant difference in atmospheric $\Delta^{14}C$ for land sites, between Debre Zeit in northeastern Africa and southern Asia (Mandla, Doi Inthanon and Saigon), arising from air-sea exchange of CO_2 for the bomb peak [*Randerson et al.*, 2002]. There was a small annual mean atmospheric $\Delta^{14}C$ anomaly of up to -7‰ for coastal and oceanic stations in northwestern Africa (Dakar, Mas Palomas and Izaña), compared to the above sites for the year 1965 [*Randerson et al.*, 2002]. As ocean exchange is strongest during winter [*Enting and Mansbridge*, 1987], the level of atmospheric $\Delta^{14}C$ anomalies for coastal and oceanic stations in northwestern Africa (relative to the land stations) is much lower in magnitude than -7‰ in April (presummer) and May–August (summer) (see discussion later), and might be close to zero for the bomb peak period. Therefore this effect can be neglected in our calculation. There was also no significant difference in atmospheric ^{14}C between low latitudes and midlatitudes in Africa and southern Asia during the bomb peak, arising from atmosphere–terrestrial biosphere exchange [*Randerson et al.*, 2002].

[17] Secondly, there are three horizontally and vertically well-mixed atmospheric boxes: low latitudes to midlatitudes north of the summer ITCZ, between the summer ITCZ and the equator, and low latitudes in the Southern Hemisphere. Atmospheric $\Delta^{14}C$ values of the first box are average values of three stations with altitudes from near surface level to 2400 m above sea level (asl). They are Izaña (28°N, 17°W, 2400 m asl), Mas Palomas (28°N, 16°W, 10–100 m asl) and Dakar (15°N, 17°W, 10–25 m asl). Four ^{14}C records representing the second box are Debre Zeit (9°N, 39°E, 1900 m asl), Mandla (23°N, 81°E, <1000 m asl), Doi Inthanon (19°N, 99°E, <1500 m asl) and Saigon (11°N, 107°E, few m asl). Summer (May–August) $\Delta^{14}C$ values for Debre Zeit are similar to tree ring ^{14}C data from the other three sites during the bomb peak period (see Figure 2a). Two records for the last box are Suva (18°S, 178°E, <200 m asl) and Fianarantsoa (21°S, 47°E, 1100 m asl). As there is no large ^{14}C gradient for the Southern Hemisphere during the bomb peak, atmospheric $\Delta^{14}C$ values for the two records are very similar [*Hua and Barbetti*, 2004].

[18] Thirdly, atmospheric ^{14}C levels for Debre Zeit, Mandla, Doi Inthanon and Saigon would be similar to that for low latitudes to midlatitudes in northwestern Africa north of the summer ITCZ if there were no influence of Southern Hemisphere air masses carried by the southwest Asian monsoon. In other words, as oceanic and terrestrial biospheric sources and sinks are small only atmospheric mixing by monsoon circulation caused changes in the atmospheric ^{14}C content for the former sites. This is a reasonable assumption.

[19] In general, the atmospheric ^{14}C level, T, for the northern sites south of the ITCZ during the summer months is determined by

$$T = n_1N_1 + n_2N_2 + sS$$

where N_1 and N_2 are atmospheric ^{14}C levels for northern low latitudes to midlatitudes, which were not influenced by the southwest Asian monsoon such as Izaña, Mas Palomas and Dakar, for the presummer period (April), and for the summer season (May–August), respectively; S is the atmospheric ^{14}C level during the summer months (May–August) for southern low latitudes, such as Suva and

Table 1. Estimate of the Contribution of Southern Hemisphere Air Masses to Northeastern Africa, and Southern Asia During May–August, the Period That the Northern Tropics are Influenced by the Southwest Asian Monsoon

Year A.D.	Atmospheric $\Delta^{14}C \pm 1\sigma$, ‰ N_1[a]	S[b]	T[c]	$s \pm 1\sigma$, %[d]
1964	787 ± 21	566 ± 7	681 ± 9	48 ± 12
1965	749 ± 13	648 ± 6	705 ± 6	43 ± 16
1966	687 ± 10	633 ± 4	650 ± 8	68 ± 27
1967	650 ± 9	591 ± 3	605 ± 9	77 ± 25
Weighted average				52 ± 13

[a]Average atmospheric $\Delta^{14}C$ values for presummer (April) for northern low latitudes to midlatitudes north of the summer ITCZ (Mas Palomas, Izaña, and Dakar) derived from the data of *Nydal and Lövseth* [1996].

[b]Mean atmospheric $\Delta^{14}C$ for May–August for southern low latitudes (Suva and Fianarantsoa) derived from the data of *Manning et al.* [1990] and *Nydal and Lövseth* [1996].

[c]Average values of $\Delta^{14}C$ in tree rings from Mandla, Doi Inthanon, and Saigon and summer atmospheric ^{14}C for Debre Zeit (May–August). These values were reported as average values of summer atmospheric ^{14}C for Northern Hemisphere zone 3 by *Hua and Barbetti* [2004].

[d]Contribution of Southern Hemisphere air masses to northern tropics.

Fianarantsoa; and n_1, n_2 and s are the associated fractions of air masses of N_1, N_2 and S, respectively.
Note that

$$n_1 + n_2 + s = 1$$

[20] To further simplify the calculation, we assume that the contribution of air masses from northern low latitudes to midlatitudes to eastern Africa, and southern Asia during the summer months is much smaller than the contribution from the other two air masses, or $n_2 \ll n_1$, and $n_2 \ll s$. Note that the magnitude of n_2 depends on the penetration of the ITCZ into the Northern Hemisphere during summer and the "strength" of the southwest Asian monsoon, and, in general, is not large as winds at the ITCZ are commonly light or nonexistent [*Linacre and Geerts*, 1997; *McGregor and Nieuwolt*, 1998]. Thus s is determined by

$$s = \frac{(N_1 - T)}{(N_1 - S)}$$

Using this equation, the contribution of Southern Hemisphere air masses to the northern tropical sites in eastern Africa, and southern Asia during the southwest Asian monsoon for the period 1964–1967 A.D. is estimated to be 52 ± 13% (Table 1).

3. Regional Atmospheric Circulation and Atmospheric ^{14}C in the Northern Midlatitudes and High Latitudes

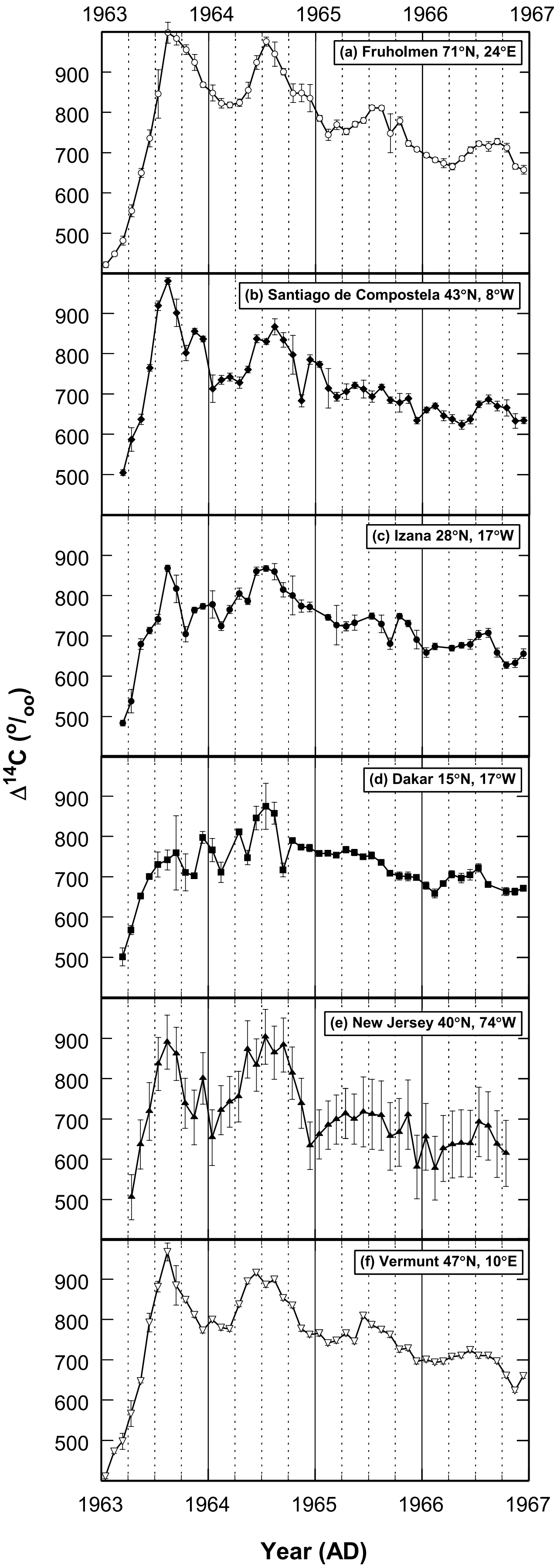
3.1. North Atlantic Region

[21] To examine influences of wind patterns on atmospheric ^{14}C for the midlatitudes and high latitudes in the North Atlantic region, atmospheric ^{14}C records from several stations in the region during the bomb peak have been analyzed. Figure 5 shows monthly mean $\Delta^{14}C$ values for Fruholmen (Norway, 71°N, 24°E), Santiago de Compostela (Spain, 43°N, 8°W), Izaña (Canary Islands, 28°N, 17°W), Dakar (Senegal, 15°N, 17°W) [*Nydal and Lövseth*, 1996], New Jersey (United States, 40°N, 74°W) [*Feely et al.*, 1963, 1966a, 1966b], and Vermunt (Austria, 47°N, 10°E) [*Levin et al.*, 1994] for the period 1963–1966 A.D. Fruholmen is at high latitudes where the injection of bomb ^{14}C from the stratosphere occurs; it has annual peaks in July–September and troughs in February–April (Figure 5a). This seasonal structure is mainly due to the injection of excess ^{14}C from the stratosphere, which occurs during spring and summer [*Meijer et al.*, 1995; *Nydal and Gislefoss*, 1996; *Levin and Hesshaimer*, 2000; *Randerson et al.*, 2002]. Similar to Fruholmen, the Santiago de Compostela record shows peaks in July–August. However, there is more than one trough per year, with a main trough in January–March (except 1966, where the trough is in May), and a secondary trough in October–December (October 1963, November 1964, December 1965, and November 1966; see Figure 5b). *Nydal* [1968], and *Nydal and Lövseth* [1983] showed a correlation between minima in the Santiago de Compostela $\Delta^{14}C$ record and troughs in local weather (wind velocity), at least for October 1963, January 1964 and November 1964. The authors argued that fossil fuel CO_2 was the cause for these 1963–1964 minima as accumulation of CO_2 free of ^{14}C from the combustion of fossil fuel became significant on calm days and thus affected atmospheric samples. If the authors were correct, the anthropogenic CO_2 contamination level for the Spanish ^{14}C record could be estimated using a two component mixing of the (observed) affected atmospheric CO_2 samples (whose $\Delta^{14}C$ values are secondary $\Delta^{14}C$ troughs), namely $CO_{2\ \mathrm{affected}} = CO_{2\ \mathrm{background}} + CO_{2\ \mathrm{fossil\ fuel}}$. According to *Levin et al.* [1980, 1989], the fossil fuel CO_2 contribution can be calculated from the following equation:

$$\frac{CO_{2\ \mathrm{fossil\ fuel}}}{CO_{2\ \mathrm{background}}} = \frac{\Delta^{14}C_{\mathrm{background}} - \Delta^{14}C_{\mathrm{affected}}}{\Delta^{14}C_{\mathrm{affected}} + 1000}$$

where $\Delta^{14}C_{\mathrm{affected}}$ refers to the value of a secondary $\Delta^{14}C$ trough, and $\Delta^{14}C_{\mathrm{background}}$ is an assumed value derived from two neighboring unaffected $\Delta^{14}C$ values of the record by linear interpolation. All $\Delta^{14}C$ values in the above equation are in per mil.

[22] Using the above equation, an estimated fossil fuel CO_2 contribution of 4–7% is required to cause the secondary troughs in the Santiago de Compostela record for 1963–1964 A.D. Winter fossil fuel CO_2 contribution levels for some European sites were estimated by *Levin et al.* [1989]. They are 2%, 1.5% and 10% for Westerland (Germany, 55°N, 8°E), Schauinsland (Germany, 48°N, 8°E) and Heidelberg (Germany, 49°N, 9°E) during the 1980s, respectively. The anthropogenic CO_2 contamination level for the Spanish station estimated above is much higher than those for Westerland (a coastal station in northwestern Europe), and Schauinsland (a rural mountain station in central Europe). It is worth noting that contamination by fossil fuel CO_2 is highest during winter [*Levin et al.*, 1985; *Meijer et al.*, 1995], and fossil fuel CO_2 emissions for West Germany



during the 1980s ($\sim$187 × 10^{12} gC) were much higher than those for Spain during the mid-1960s ($\sim$19 × 10^{12} gC) [*Marland et al.*, 2003]. If these two factors are taken into account, the contamination level estimated for the Spanish record of 4–7% for autumn to early winter during 1963–1964 is effectively much higher than the value of 10% estimated for Heidelberg (a highly populated and industrialized area in central Europe) during the 1980s. In brief, as Santiago de Compostela is only a few tens of kilometers inland from the westernmost coast of mainland Europe and away from major industrial CO_2 sources in northern Spain ($\sim$150 km at least), the above estimated fossil fuel CO_2 contribution of 4–7% for 1963–1964 is too high and unexpected for this site. This suggests that the secondary ^{14}C troughs observed in the Spanish record cannot be explained solely by contamination with fossil fuel CO_2 from sources within a few hundred kilometers. As mentioned by *Nydal* [1968] and *Nydal and Lövseth* [1983], the atmospheric samples at Santiago de Compostela were taken in a corner of a small airport, away from the runway, 11 km from the town, and the traffic at this airport was small (less than one flight per day in 1963). Local fossil fuel CO_2 contamination of 4–7% for 1963–1964 is also unlikely (see discussion later).

[23] Atmospheric ^{14}C records for Izaña and Dakar (Figures 5c and 5d) have $\Delta^{14}C$ seasonal structures similar to that of Santiago de Compostela. Both the Izaña and Dakar records have main troughs in January–April and secondary troughs in September–November (October 1963, September 1965 and October–November 1966 for Izaña, and October–November 1963, September 1964 and October–November 1966 for Dakar). One may argue that the secondary ^{14}C troughs in Santiago de Compostela, Izaña and Dakar records are simply due to experimental scatter. This is unlikely as atmospheric ^{14}C samples for these three records and Fruholmen were analyzed in the same laboratory [*Nydal and Lövseth*, 1996], and the latter record shows no secondary troughs (Figure 5a). Another interesting feature of these three atmospheric records from western Europe and northwestern Africa (Figures 5b–5d) is that the magnitudes of their secondary ^{14}C troughs are similar. For examples, decreases of $\sim$77‰ in $\Delta^{14}C$ in October 1963 for Santiago de Compostela, $\sim$86‰ in October 1963 for Izaña, and $\sim$61–83‰ in October–November 1963 for Dakar, are observed. If the latter two secondary troughs were due to local fossil fuel CO_2 contamination, their magnitudes could not be similar to that of the Spanish record. If the latter two secondary troughs were due to fossil fuel CO_2 contamination from sources within a few hundred kilometers, their

Figure 5. Monthly atmospheric ^{14}C during the bomb peak for stations in Europe, northwestern Africa, and the northeastern United States: (a) Fruholmen (Norway), (b) Santiago de Compostela (Spain), (c) Izaña (Canary Islands), (d) Dakar (Senegal), (e) New Jersey (United States), and (f) Vermunt (Austria). Monthly mean values are derived from the data of *Nydal and Lövseth* [1996] for Fruholmen, Santiago de Compostela, Izaña, and Dakar; *Levin et al.* [1994] for Vermunt; and *Feely et al.* [1963, 1966a, 1966b] for New Jersey. Error bars are 1σ.

magnitudes could not be similar because fossil fuel CO_2 emissions for Spain during the mid-1960s ($\sim 19 \times 10^{12}$ gC) are much higher than those for Senegal ($\sim 0.36 \times 10^{12}$ gC) [*Marland et al.*, 2003], and anthropogenic CO_2 emissions for the Canary Islands would also be small. In addition, ^{14}C data for New Jersey shown in Figure 5e have a seasonal structure similar to that of the Santiago de Compostela record (Note that this similarity was mentioned by *Nydal* [1968]). The New Jersey record has main peaks in July August and primary troughs in January–February. As the record was not measured with high precision (~5% precision [*Feely et al.*, 1966a]), its secondary troughs may be masked by large measurement uncertainties and not apparent (Figure 5e). However, there are decreases in monthly mean ^{14}C values in September–December (October–November 1963, September–October 1965 and December 1965) and these ^{14}C decreases may be evidence of secondary troughs in the record. If Nydal and Lövseth were correct, how could local weather around the Spanish station be related to the ^{14}C seasonal structure of Izaña, Dakar and possibly New Jersey, which are far away from Santiago de Compostela? Note that distances between Izaña, Dakar and New Jersey, and Santiago de Compostela are ~2000 km, ~3500 km and ~6500 km, respectively. Also, the atmospheric record for Vermunt in central Europe (Figure 5f) has secondary minima (although they are much smaller in magnitude compared to those of Santiago de Compostela, Izaña, Dakar and New Jersey) in November–December (December 1963, November 1966).

[24] As mentioned in section 2, fluxes from the oceans and terrestrial biosphere also contributed to atmospheric ^{14}C variations during the bomb peak period. These fluxes may therefore contribute to the secondary troughs of atmospheric $\Delta^{14}C$ records in the North Atlantic region, which occurred during the autumn season. During the bomb peak period, the air-sea exchange flux caused decreases in atmospheric $\Delta^{14}C$ for oceanic and coastal stations in the North Atlantic region [*Randerson et al.*, 2002]. However, as ocean uptake of atmospheric CO_2 is largest in winter [*Enting and Mansbridge*, 1987], the contribution of the air-sea exchange flux to the above secondary troughs of atmospheric $\Delta^{14}C$ during autumn may not be large. The terrestrial biospheric flux also caused decreases in atmospheric $\Delta^{14}C$ for the North Atlantic region during the bomb peak period. The effect was strongest during autumn when respiration rates were high [*Enting and Mansbridge*, 1987; *Randerson et al.*, 2002]. The effect was small for Santiago de Compostela, Izaña and Dakar, and larger for New Jersey for the bomb peak period [*Randerson et al.*, 2002]. However, there is no information of the magnitude of this effect in autumn months for the North Atlantic region. The terrestrial biospheric flux may have contributed to the secondary troughs of the atmospheric ^{14}C records in the North Atlantic region, but in what portion is unknown.

[25] The similarity of the excess $\Delta^{14}C$ seasonal structures between the above records strongly suggests that wind systems over the North Atlantic region, rather than local weather and CO_2 free of ^{14}C derived from fossil fuel CO_2 sources within tens or hundreds of kilometers, are responsible for the presence of the secondary ^{14}C troughs discussed above. During the summer of the Northern Hemisphere, continental wind flows southward from northern Europe. This wind transfers excess ^{14}C from locations in high latitudes such as Fruholmen to Vermunt, Santiago de Compostela, Izaña and Dakar (see Figure 3). New Jersey receives excess ^{14}C from northern America by northwesterly wind, and from Europe and northwestern Africa by southwesterly wind, which originates from easterly wind across the Atlantic Ocean. A high-pressure zone over the North Atlantic Ocean creates a well mixed atmospheric zone for western Europe, northwestern Africa and the eastern United States through atmospheric circulation (Figure 3). During the winter of the Northern Hemisphere, the wind systems change. A reverse wind flows from the Atlantic Ocean to northern Europe, which inhibits transfer of excess ^{14}C to the south. Similarly, New Jersey is dominated by westerly winds during the winter time, which do not come from the northern high latitudes (Figure 3). In brief, the air masses over these sites are not dominated by northern high-latitudinal air during the winter. Note that the semiperiodic variation in the winter mode of atmospheric circulation across the North Atlantic region is known as the North Atlantic Oscillation (NAO) [*Hurrell et al.*, 2003]. As a result of the above seasonal wind patterns a very similar ^{14}C seasonal structure for western Europe, northwestern Africa and the eastern United States can be inferred. The two wind modes are dominated by two different air masses: high-latitudinal air with relatively high ^{14}C, and low-latitudinal air with relatively low ^{14}C, respectively. During the bomb peak, the difference between the two wind modes in terms of ^{14}C are significant, therefore any changes in the wind systems for a particular region can cause a significant change in atmospheric ^{14}C for that region. The secondary troughs found in September–December for the above atmospheric records (Vermunt, Santiago de Compostela, Izaña, Dakar and New Jersey) might be due to temporary reversal of wind system from high-latitudinal to low-latitudinal air mode. During the study period 1963–1966 A.D., the NAO was in negative phase with a more southerly track of westerly winds across the North Atlantic region [*Hurrell et al.*, 2003]. As a result, the midlatitudes in the North Atlantic region such as southwestern Europe, the Mediterranean, northwestern Africa and the eastern United States were strongly influenced by (low-latitudinal) westerly winds. Meanwhile, high latitudes in continental Europe such as Vermunt in Austria (47°N, 10°E) received little contribution from (low-latitudinal) westerly winds. This explains why there are much larger secondary troughs in September–December for the former sites compared to those in November–December for Vermunt for 1963–1966 A.D.

3.2. East Asia

[26] Another example of the influence of seasonal atmospheric circulation patterns on atmospheric ^{14}C can be seen for the midlatitude regions in east Asia and adjacent seas. During the summer season, unlike Europe and northwestern Africa, this part of the world mainly receives maritime air from the northern Pacific Ocean (Figure 3), which does not come from high latitudes and therefore has a ^{14}C level lower than that of high-latitudinal air during the bomb peak. During the winter season, this region receives high-latitudinal air from a high-pressure zone over north central Asia (Figure 3). During the bomb peak, this air mass carried large amounts

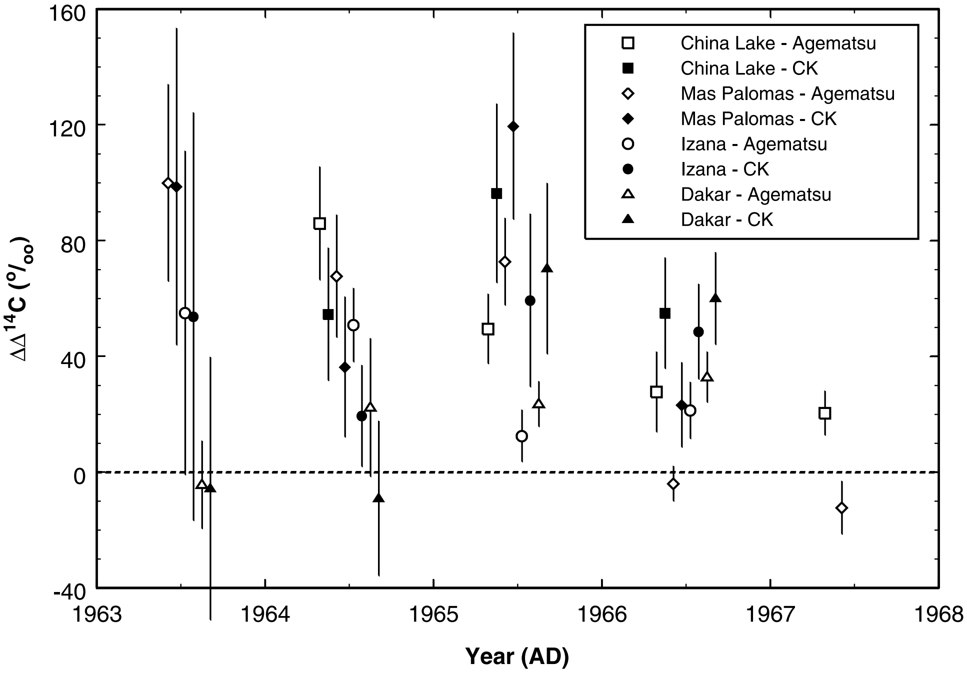


Figure 6. Differences in $\Delta^{14}C$ between sites in northern low latitudes to midlatitudes in the western United States and northwestern Africa and midlatitudes in east Asia for the growing season of tree rings. CK stands for Mounts Chiak and Kyeryong. All symbols should be plotted in the middle of each year. However, they are displayed in the diagram with small temporal offsets for reason of clarity. Error bars are 1σ. Note that the May–August ^{14}C data for China Lake used for the calculation of $\Delta^{14}C$ differences were original data reported by *Berger et al.* [1965] and *Berger and Libby* [1966, 1967, 1968] after correction for $\delta^{13}C$ (using an assumed value of −23.2‰, which is the average $\delta^{13}C$ value for this site for the period 1977–1983 A.D. [*Berger et al.*, 1987]) and decay correction.

of excess ^{14}C from high latitudes to these regions. Unfortunately, there is no atmospheric ^{14}C record available during the bomb peak for east Asia to see whether any difference exists between the atmospheric ^{14}C seasonal structure for this region and that for eastern America, western Europe and northwestern Africa. Only atmospheric ^{14}C data for the summer season derived from tree rings over Japan [*Muraki et al.*, 1998] and Korea [*Park et al.*, 2002] are available. Figure 6 shows differences in $\Delta^{14}C$ for tree rings from Japan and Korea compared to those for summer months (May–August) derived from atmospheric records in northern low latitudes to midlatitudes. The results show that the ^{14}C levels for summer months during the bomb peak for Japan and Korea (36°–37°N, 122°–138°E) are usually lower than those at similar latitudes in the western United States (China Lake, 36°N, 118°W) and even those at lower latitudes in northwestern Africa (Izaña, 28°N, 17°W; Mas Palomas, 28°N, 16°W; and Dakar, 15°N, 17°W). The magnitudes of the differences are 20–96‰ for China Lake versus east Asia for 1964–1967, 23–120‰ for Mas Palomas versus east Asia for 1963–1966 (except the difference between Mas Palomas and Agematsu is ~0‰ in 1966), 13–59‰ for Izaña versus east Asia for 1963–1966, and 23–70‰ for Dakar versus east Asia for 1964–1966 (except the difference between Dakar and Mounts Chiak and Kyeryong is small in 1964). The relatively low atmospheric ^{14}C during summer months for Japan and Korea for the bomb peak is due to the fact that these regions received low-latitudinal air containing relatively low ^{14}C, while other sites in low latitudes to midlatitudes in America and northwestern Africa received more northerly air masses containing higher ^{14}C. This is understandable in the light of the influence of regional atmospheric circulation on regional atmospheric ^{14}C. Note that there were no significant atmospheric $\Delta^{14}C$ anomalies between the above sites in the western United States, northwestern Africa and midlatitudes in Japan and Korea for the bomb pulse period, arising from oceanic and terrestrial biospheric exchanges [*Randerson et al.*, 2002].

4. Conclusion and Implications

[27] Published bomb ^{14}C data from tree rings and atmospheric CO_2 samples at different latitudes in the Northern Hemisphere during the bomb peak have been systematically analyzed. For the northern tropics, ^{14}C levels over India, Thailand, Vietnam and Ethiopia were significantly lower than those at similar latitudes in northwestern Africa, implying that the Asian summer monsoon and ITCZ position influenced atmospheric ^{14}C over the study area. This supports our previous finding that atmospheric ^{14}C over Thailand was influenced by the entrainment of Southern Hemisphere air masses in the southwest Asian monsoon during the latter part of the LIA and for 1938–1954 A.D. [*Hua et al.*, 2004a, 2004b].

[28] For the North Atlantic region, atmospheric records from western Europe, northwestern Africa and the northeastern United States were observed to have coincident variations, characterized by secondary ^{14}C troughs during September–December. These troughs might partly be due to a temporary reversal of wind systems from high-latitudinal to low-latitudinal air mode containing relatively lower ^{14}C levels during the bomb peak. Tree ring $\Delta^{14}C$ values for midlatitudes in Japan and Korea (east Asia) were lower than those at similar latitudes in the western United States and even lower than those at lower latitudes in northwestern Africa, because the eastern Asian sites received low-latitudinal air masses from the Pacific which contained relatively low levels of ^{14}C for the bomb peak, during the growing season of tree rings.

[29] These observations show that the spatial distribution of bomb ^{14}C during the bomb peak depended on atmospheric circulation and did not have a simple latitudinal dependence. Global atmospheric circulation is characterized by Ferrel cell circulation for midlatitudes to high latitudes, and Hadley cell and monsoon circulations for low latitudes to midlatitudes [*Linacre and Geerts*, 1997]. These circulations created 3 different zones for the Northern Hemisphere, separated more or less by the Ferrel cell–Hadley cell boundaries between zones 1 and 2, and the summer ITCZ between zones 2 and 3 [*Hua and Barbetti*, 2004].

[30] Intrahemispheric ^{14}C offsets for the Northern Hemisphere for the prenuclear era have been investigated by a number of researchers. The magnitudes of these ^{14}C offsets are about several per mil and their uncertainties are of similar levels for the prebomb period [*Damon et al.*, 1989; *Hua et al.*, 2004a], and for earlier periods [*Damon et al.*, 1996; *Stuiver and Braziunas*, 1998; *Kromer et al.*, 2001; *Hua et al.*, 2004b]. Because of measurement uncertainties it has sometimes been difficult to determine the magnitudes of intrahemispheric ^{14}C offsets. Bomb ^{14}C injected into the atmosphere during the late 1950s and early 1960s increased the contrast, in terms of ^{14}C, between different air masses, which helps to improve our understanding of the influence of atmospheric circulation on regional ^{14}C differences. This notion may provide a valuable guide to explaining why intrahemispheric ^{14}C offsets for the Northern Hemisphere appear to be quite variable through time, even between sites which differ little in latitude.

[31] **Acknowledgments.** We thank Stuart Hankin for helping with the preparation of figures. We also thank Nir Krakauer and two anonymous reviewers for critical and constructive comments. We gratefully acknowledge funding over many years from the Australian Institute of Nuclear Science and Engineering (AINSE), A. M. Macintosh, and the Australian Research Council (ARC).

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