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REVIEW OF RADIOCARBON DATA FROM ATMOSPHERIC AND TREE RING SAMPLES FOR THE PERIOD 1945–1997 AD

by

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SUMMARY

A summary of ^{14}C data from atmospheric sampling and measurements on wood from annual tree rings for the period 1945–1997 AD is presented and evaluated. Atmospheric records are characterized by different distributions of bomb-test ^{14}C between the Northern and Southern Hemispheres, latitude dependence, and seasonal fluctuations. Radiocarbon data from tree rings are summarised and plotted against atmospheric records from similar latitudes. In some cases, discrepancies are found. Possible reasons for this include: 1) the use of stored carbohydrate from the previous year, 2) different ^{14}C levels in the air around subcanopy trees due to respiration of CO_2, 3) regional and local effects of anthropogenic CO_2 and ^{14}C sources, 4) sampling of wood material too close to ring boundaries, and 5) insufficient pretreatment of tree ring samples for dating. But in cases where trees were carefully selected and the samples adequately pretreated, radiocarbon data from tree rings show excellent agreement with direct atmospheric sampling records.

Key words: Atmospheric radiocarbon, tree rings, nuclear detonation, air-sea exchange, atmospheric mixing

INTRODUCTION

A project to measure ^{14}C concentration in annual tree rings from Australia and Thailand in the bomb-test period is being carried out at the Australian Nuclear Science and Technology Organisation (ANSTO) and the University of Sydney. As a part of our project, the data from other studies of atmospheric radiocarbon and tree rings have been collected and evaluated as a background for our research. In this paper we present a summary of previously published data for the period 1945–1997, together with a brief description of our experiment.

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Atmospheric radiocarbon levels are usually expressed as $\Delta^{14}C$ values, in parts per thousand above or below the internationally accepted standard concentration. All values are adjusted for isotopic fractionation (using $\delta^{13}C$) and radioactive decay up to the time of measurement (from 1950 AD for the standard; and from the time of collection for atmospheric samples, or the time of formation for tree rings) (Nydal & Lovseth 1996; Hertelendi & Csongor 1982; and the Δ term of Stuiver & Polach 1977).

ATMOSPHERIC RADIOCARBON RECORDS

As a result of hundreds of atmospheric nuclear weapon explosions, starting in 1945, the concentration of ^{14}C in the atmosphere increased. As the amount of ^{14}C generated in the atmosphere is proportional to the effective yield of the nuclear detonation (which is a function of the height of burst; Enting 1982), the shape of atmospheric $\Delta^{14}C$ curves followed the magnitude of nuclear detonation (Fig. 1). There were three major periods of atmospheric nuclear explosions as shown in Figure 1:

1) Some atmospheric nuclear explosions in 1945–1958 (negligible injection of ^{14}C into the atmosphere between 1945–1951 AD; moderate amount between 1952 and 1958 AD): atmospheric $\Delta^{14}C$ increased in the troposphere and peaked at Vermunt, Austria (47° N) in 1959. The level of ^{14}C in the Northern Hemisphere decreased in 1960 and balanced with that in the Southern Hemisphere (Wellington, New Zealand at 41° 18' S) at the end of 1961, in the absence of atmospheric nuclear testing in the period 1959–1960.

2) There were many atmospheric nuclear tests in 1961–1962 (large amounts of ^{14}C injected to the atmosphere): the most dramatic increase occurred in the period 1962–1963, when the ^{14}C content in the northern troposphere nearly doubled. Since the Nuclear Test Ban Treaty of 1963, the $\Delta^{14}C$ level has gradually declined due to the CO_2 exchange between the atmosphere and the ocean and biosphere (Levin & Kromer 1997; Nydal & Gislefoss 1996; Manning et al. 1990).

3) Sporadic atmospheric testing in 1966–1980: atmospheric $\Delta^{14}C$ decreased exponentially, but there were some disturbances in atmospheric ^{14}C in the period 1968–1971 due to bomb tests mostly performed by China in the Northern Hemisphere and France in the South Pacific. For example, there was no decrease in atmospheric $\Delta^{14}C$ at Fruholmen (Norway, 71° 06' N) between the July 1969 peak of 576 ‰ and the August 1970 peak of 589 ‰ (Fig. 1 & 3); and at N'Djamena (Chad, 12° 08' N) between the July 1968 peak of 591 ‰ and the June 1969 peak of 594 ‰ (Fig. 3).

$\Delta^{14}C$ in the troposphere at different latitudes in the Northern and Southern Hemispheres are shown in Figure 2. Solid symbols represent stations in the Northern Hemisphere and open symbols represent samples collected in the Southern Hemisphere. A clear difference between the Hemispheres is found for the period 1959–1970 AD.

Most atmospheric nuclear tests were carried out at high- and mid-northern latitudes. The bomb-test ^{14}C produced around the fire ball was mostly injected into the stratosphere by the plume, then returned to the troposphere during spring and summer, when the tropopause height increased in the higher latitudes (Nydal & Gislefoss 1996). As the result of the exchange between the two layers of the atmosphere, and atmospheric mixing in the troposphere, there was a gradient in the ^{14}C content of the troposphere: high levels in the higher and lower levels in lower northern latitudes, and much lower levels in the Southern Hemisphere. As we can see from Figure 1, the highest level at Fruholmen, Norway (71° 06' N) is 1040 ‰ in late August 1963 (Nydal & Lovseth 1996) while the peak at Wellington, New Zealand (41° 18' S) is 695 ‰ in early July 1965 (Manning & Melhuish 1994). The maximum ^{14}C level of the bomb peak in the troposphere decreased from north to south, with a time delay of 1–2 years.

Figure 3 shows the $\Delta^{14}C$ of six stations at different latitudes in the Northern Hemisphere, from the far north (Fruholmen, Norway at 71° N) to tropical regions (Debre Zeit, Ethiopia at 8° N). Clear latitude dependence is found in the 1960s. A summary of the $\Delta^{14}C$ at six stations in the Southern Hemisphere, from tropical regions (Suva, Fiji at 18° S) to Antarctica (Scott Base at 78° S), is shown in Figure 4. A small latitude dependence is found (Manning et al. 1990). The reason for the low latitudinal gradient in the Southern Hemisphere is that the sources of bomb ^{14}C, which are mainly in the Northern Hemisphere, are far from the sampling sites (Manning et al. 1990), and once the ^{14}C excess is transported over the broad Inter-Tropical Convergence Zone, fast mixing within the Southern Hemisphere does not allow for a large gradient there. By the late 1970s the radiocarbon content of the entire troposphere was more or less homogeneous and decreasing (Fig. 1–4) by half over 17 yr (Manning et al. 1990; Dai et al. 1992). From 1980 onwards, atmospheric $\Delta^{14}C$ in Europe was slightly lower than that in the Southern Hemisphere (Fig. 1). This was due to a regional 'Suess' effect over Europe (from fossil fuel; Manning et al. 1990).

The seasonal fluctuations of ^{14}C in the Northern Hemisphere during the period 1963–1968 (Fig. 1–3) are due to at least five reasons, placed in decreasing order of significance (Enting & Mansbridge 1987):

1) The injection of ^{14}C from the stratosphere into the troposphere peaks during spring and summer as mentioned above;
2) there are seasonal variations in the rate of transfer of ^{14}C to the Southern Hemisphere;
3) ocean uptake of atmospheric CO_2 is largest in winter;
4) the largest release of CO_2 from the biosphere to the atmosphere is in autumn. This CO_2 will be of relatively low ^{14}C activity in the period immediately following testing and so the dilution is greatest in the autumn; and
5) the dilution of $^{14}CO_2$ by excess of dead CO_2 from fossil fuel combustion (mostly in the Northern Hemisphere) is largest in winter.

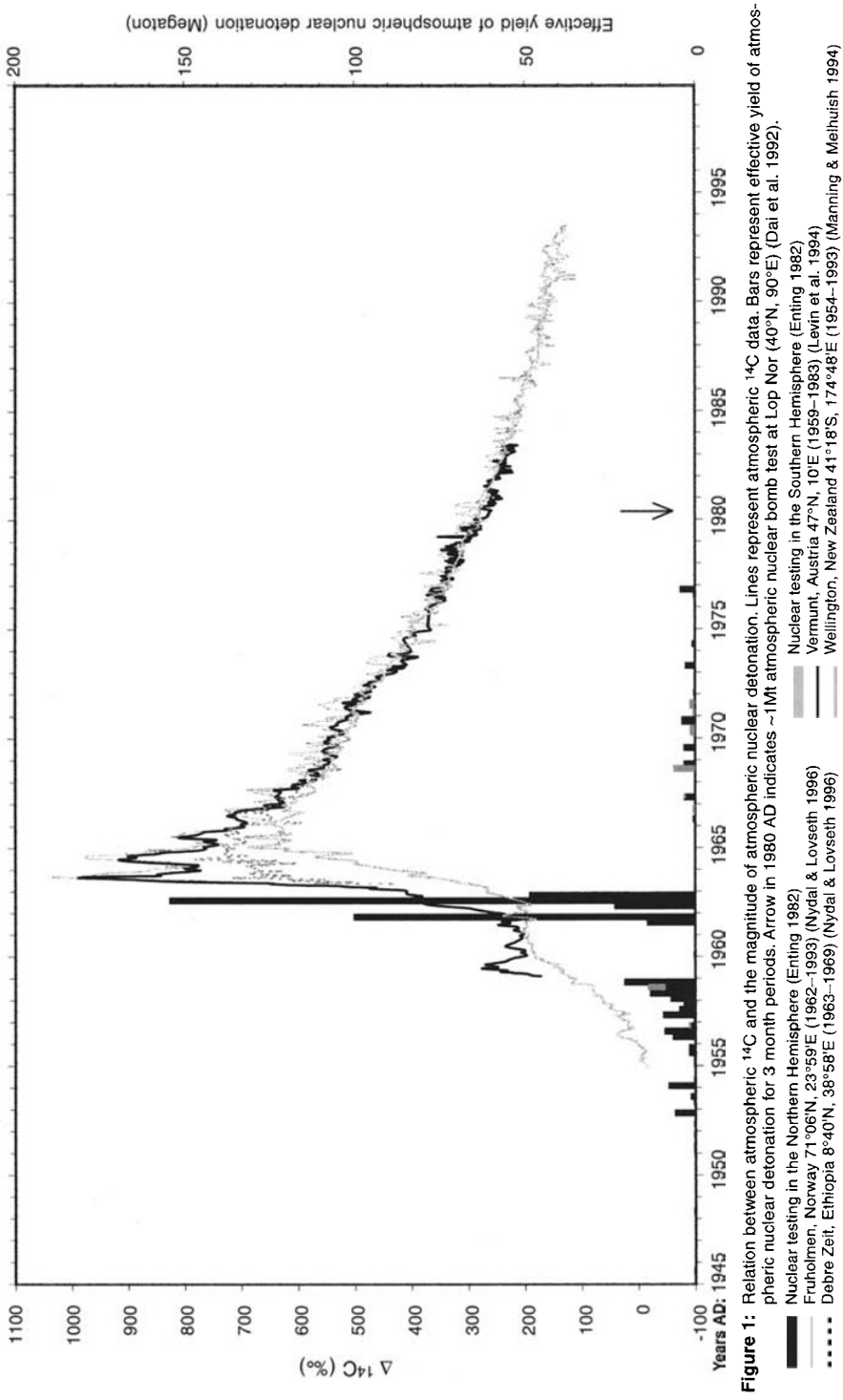

Figure 1: Relation between atmospheric ^{14}C and the magnitude of atmospheric nuclear detonation. Lines represent atmospheric ^{14}C data. Bars represent effective yield of atmospheric nuclear detonation for 3 month periods. Arrow in 1980 AD indicates ~1Mt atmospheric nuclear bomb test at Lop Nor (40°N, 90°E) (Dai et al. 1992).

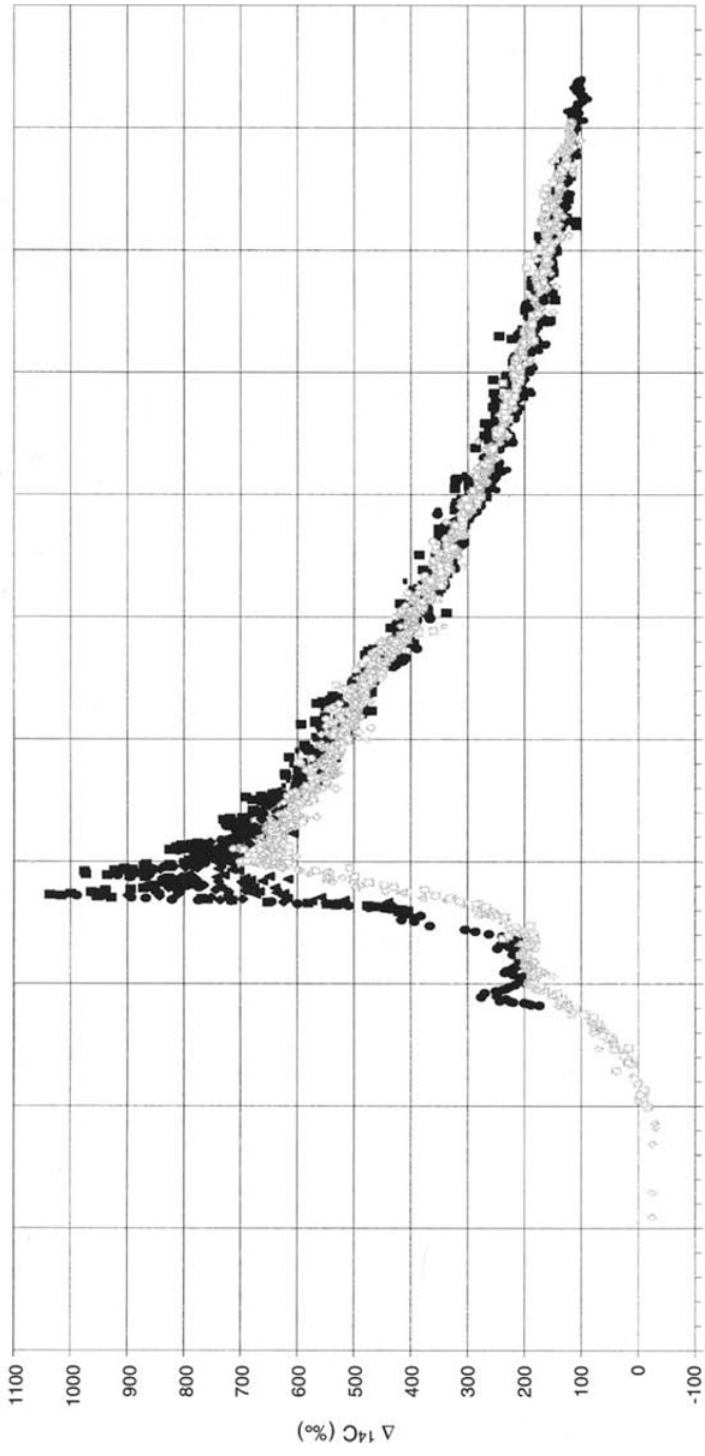

Figure 2: Atmospheric ^{14}C at different latitudes. Error bars are too small to be shown.

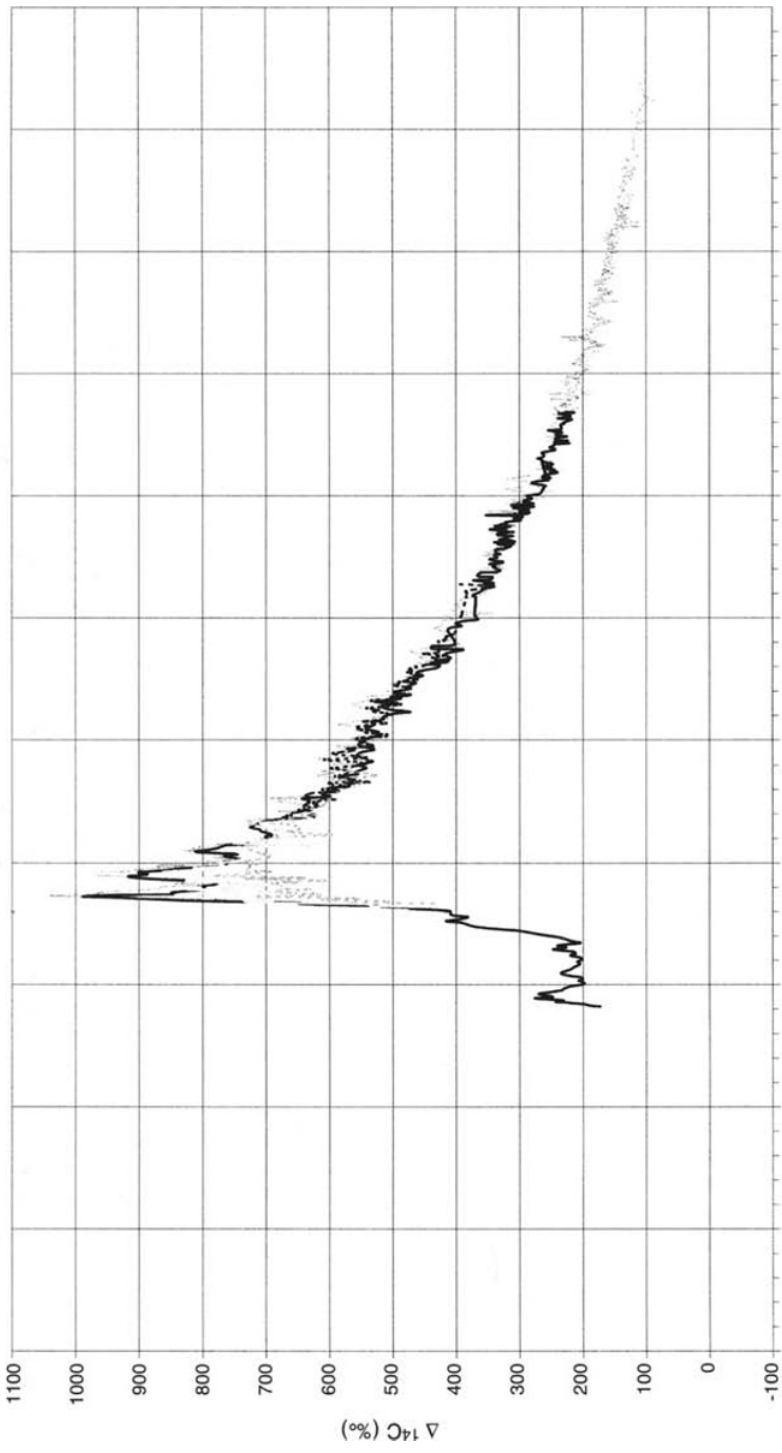

Figure 3: Atmospheric ^{14}C in the Northern Hemisphere at various latitudes. Error bars are too small to be shown.
Fruholmen, Norway 71°06'N, 23°59'E (1962–1993) (Nydal & Lovseth 1996)
Vermunt, Austria 47°N, 10'E (1959–1983) (Levin et al. 1994)
Izana, Canary Islands 28°22'N, 16°30'W (1963–1967) (Nydal & Lovseth 1996)
N'Djamena, Chad 12°08'N, 15°57'E (1966–1976) (Nydal & Lovseth 1996)
Debre Zeit, Ethiopia 8°04'N, 38°58'E (1963–1969) (Nydal & Lovseth 1996)
Schauinsland, Germany 48°N, 8°E (1976–1997) (Levin et al. 1994)

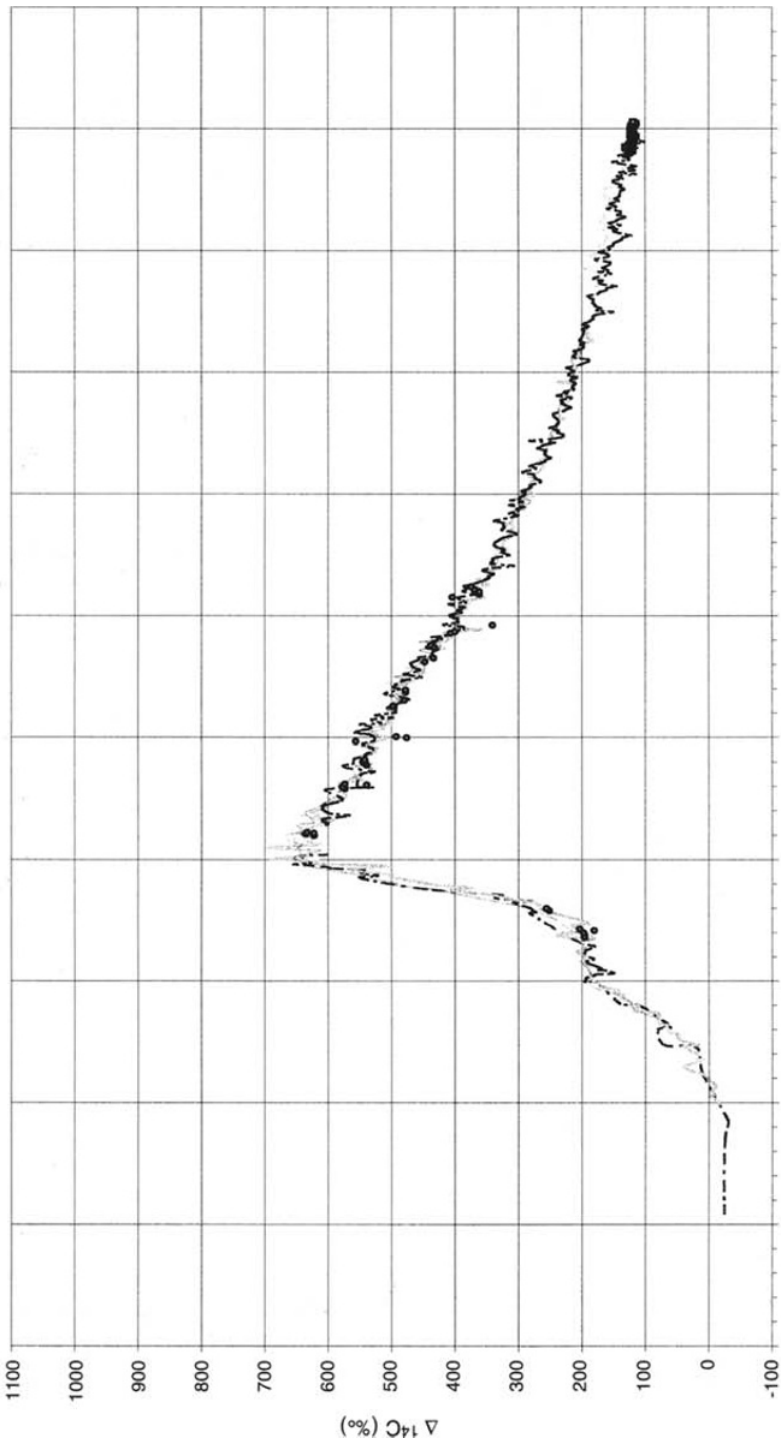

Figure 4: Atmospheric ^{14}C in the Southern Hemisphere at various latitudes. Error bars are too small to be shown.

Table 1. Summary of data of annual tree rings (13 groups) and leaves (1 group), including pretreatment method and the proportion for which there is a discrepancy between the ^{14}C content in tree rings or leaves and the atmosphere.

Data source & pretreatment method	**Tree species & (tree latitude vs atmospheric latitude)**	**No. annual rings measured (years 19xx AD)**	**No. measurements with discrepancies [(years 19xx AD) #]**	**Fig. No.**
Kolesnikov et al. (1970)+	Pine (60°N vs 71°N & 45°N)	10 (58–67)	No discrepancy	5
Dai & Fan (1986)	Spruce (68°N vs 71°N)	5 (63–67)	2 [(65–66) **H**]	5
Dai et al. (1992)	Spruce (47°N vs 47°N)	21 (61–67 & 70–83)	11 [(65–67 & 75–78) H & (79–82) **H**]	6a
AAA	White spruce (27°N vs 28°N)	7 (61–67)	2 [(66–67) H]	6a
Muraki et al. (1998) (Solvent extraction + Alkali + Acid)	Pine (36°N vs 28°N & 47°N)	10 (60–67, 69 & 95)	No discrepancy	6a
Nakamura et al. (1987a, b) AAA	Japanese cypress (*Chamaecyparis obtusa*) (35°N vs 28°N & 47°N)	38 (45–83)	6 [(61) **L**, (65–66) 1, (81–82) H & (83) h]	6a
Cain & Suess (1976)	Oak (41°N vs 47°N)	24 (45–68)	7 [(59) L, (60) 1 & (64–68) L]	6b
(Solvent extraction + Alkali + Acid)	Red oak (41°N vs 47°N)	26 (45–70)	No discrepancy	6b
McNeely (1994) Hot water rinse	Maple leaves (45°N vs 47°N)	33 (61–93)	23 [(67, 71–72 & 85–93) h, (76, 80 & 84) H & (68–69, 73–75 & 81–83) **H**]	6b
Eichinger et al. (1980)+	Poplar (48°N vs 47°N)	20 (58–77)	3 [(58–60) **L**]	6c
Hertelendi & Csongor (1982) (Hot water + $NaClO_2$ / HCl + Alkali + Acid)++	Acacia (*Robinia pseudoacacia*) (47°N vs 47°N)	28 (51–78)	5 [(65–67 & 69) L & (64) h]	6c
Levin et al. (1985)	Pine (*Pinus nigra*) (49°N vs 47°N)	17 (66–82)	**1** [(66) L]	6c
Levin & Kromer (1997) (AAA + Solvent extraction)	Spruce (*Picea abies*) (48°N vs 47°N & 48°N)	12 (74–85)	**1** [(74) h]	6c

(Table 1 continued)

Kozak et al. (1989) (Hot Na_2S + $NaClO_2$ / HCl)$^{+++}$	Spruce (48° vs 47°N)	14 (56, 59, 61, 63–66, 69, 72, 75, 78, 81, 84 & 86)	3 [(66) l, (75) H & (84) **H**]	6c
Willkomm & Erlenkeuser (1968)$^{+}$	Elm (*Ulmus*) (54°N vs 47°N)	16 (48–53, 54–64)	2 [(59–60) h]	6c
Worbes & Junk (1989)	Oak (*Quercus robur*) (51°N vs 47°N)	12 (64, 76–86)	2 [(64) **H** & (83) H]	6c
	Rourea sp. (3°S vs 18°S)	10 (51–59, 63)	2 [(58–59) l]	7b
AAA	*Swartzia laevicarpa* (3°S vs 41°S)	24 (58–81)	11 [(58–61 & 67–71) **L**, (75) L & (80) l]	7b
Murphy et al. (1997)	Teak (*Tectona grandis*) (23°N vs 28°N)	6 (55, 60, 63, 65, 70 & 80)	No discrepancy	7a
	Teak (*Tectona grandis*) (7°S vs 18°S)	6 (55, 60, 63, 65, 70 & 80)	1 [(63) **L**]	7b
Holocellulose (Solvent extraction + $NaClO_2$ / HCl)				
Kikata et al. (1992 & 1993)	Merkusii pine (*Pinus merkusii*) (11°N vs 8°N)	6 (62–67)$^{@}$	No discrepancy	7a
Yonenobu et al. (1995)	Teak (*Tectona grandis*) (7°S vs 18°S)	14 (57–70)$^{@}$	6 [(59, 62, 64 & 69–70) **L** & (63) L]	7b
Nakai & Nakurama (1989)	Parana pine (*Araucaria angustifolia*) (27°S vs 26°S)	10 (60–69)$^{@\#\#}$	3 [(60) **L**, (61) L & (69) h]	8
AAA	Lingue (*Percea lingue*) (34°S vs 41°S)	8 (61–68)$^{@\#\#}$	2 [(62 & 68) L]	8

Legend: AAA = Acid, Alkali, Acid; $^{+}$ = no pretreatment is stated; $^{++}$ = pretreated material is not alpha-cellulose as quoted by the authors (because solvent extraction is lacking); $^{+++}$ = pretreated material is not holocellulose as quoted by the authors (because solvent extraction is lacking).

$^{\#}$ (**H**, H & h): ^{14}C content in tree ring is much higher, higher and slightly higher than that in atmosphere respectively; (**L**, L & l): ^{14}C content in tree ring is much lower, lower and slightly lower than that in atmosphere respectively.

$^{\#\#}$: We have adjusted the dates of tree rings of Parana pine (27°S) and Lingue (38° 04' S). The time span of Parana pine given in Table 2 of Kikata et al. (1993) is 1961–1970. However, the peak in 1965 was plotted in the middle of the growing period for trees in the Southern Hemisphere (Dec. 1964–Jan. 1965) (Fig. 2 of Kikata et al. 1993 and Fig. 1 of Kikata et al. 1992). Therefore, the time span for Parana pine (27°S) should be 1960–1969, following normal dendrochronological conventions. A similar problem was found for Lingue (38° 04' S), so its time span is 1961–1968 rather than 1962–1969 as quoted in Table 2 of Kikata et al. (1993). The labelling of the two time series of Parana pine and Lingue is also transposed in Fig. 2 of Yonenobu et al. (1995).

$^{@}$: $\delta^{13}C$ values are not listed in the publications and corresponding $\Delta^{14}C$ values plotted in Fig. 7a, b & 8 are not adjusted for isotopic fractionation.

(text continued on page 270)

These five phenomena produce seasonal fluctuations in atmospheric $\Delta^{14}C$: summer maxima and winter minima. The large seasonal variations of ^{14}C found during 1963–1968 were caused mainly by the first phenomenon – the meteorological influx of ^{14}C from the stratosphere with concentrations 10 to 20 times higher during that early period (Nydal & Gislefoss 1996).

TREE-RING DATA

^{14}C data in annual tree rings, which have been published by thirteen groups of researchers, are presented in Table 1 to cover the whole range from the high latitudes in the Northern Hemisphere to the middle latitudes in the Southern Hemisphere. The issue raised here is "does the ^{14}C content of annual tree rings match the atmospheric radiocarbon level at the same latitude?" If not, why?

In order to compare the two types of data, we split them into four different groups of latitudes. The seven diagrams compare $\Delta^{14}C$ in tree rings and in the atmosphere at high latitudes in the Northern Hemisphere (Fig. 5), middle latitudes in the Northern Hemisphere (Fig. 6a, b, c), equatorial regions (Fig. 7a, b) and middle latitudes in the Southern Hemisphere (Fig. 8). In each graph we show atmospheric curves at 47° N (Vermunt, Austria) and at 41° S (Wellington, New Zealand) as reference curves for the Northern and Southern Hemispheres respectively, for ease of comparison. $\Delta^{14}C$ values for annual tree rings are plotted as points in the middle of the growing period: June–July of the current year for the Northern Hemisphere and December (current year)–January (next year) for the Southern Hemisphere.

The differences between ^{14}C contents in annual tree rings and in the atmosphere at the appropriate latitude are summarised in Table 1. The number of discrepancies for each group are 0/10 for Kolesnikov et al. (1970); 15/33 for Dai and Fan (1986), Dai et al. (1992); 0/10 for Muraki et al. (1998); 6/38 for Nakamura et al. (1987a, b); 7/50 for Cain and Suess (1976); 3/20 for Eichinger et al. (1980); 5/28 for Hertelendi and Csongor (1982); 2/29 for Levin et al. (1985), Levin and Kromer (1997); 3/14 for Kozak et al. (1989); 2/16 for Willkomm & Erlenkeuser (1968); 15/46 for Worbes and Junk (1989); 1/12 for Murphy et al. (1997); and 11/38 for Kikata et al. (1992, 1993), and Yonenobu et al. (1995). A set of data from maple leaves is also included in Table 1; there are 23 discrepancies in 33 years of data.

Possible reasons for these discrepancies are:

1) Trees used some carbohydrate reserves from the previous year to form the current year ring (Worbes & Junk 1989; Grootes et al. 1989a, b). This could cause lower ^{14}C concentrations in tree rings in comparison with those in the atmosphere for rings before the bomb peak (1963–1964 for the Northern Hemisphere and 1965 for

the Southern Hemisphere). This might, for example, explain the cases of rings 1958–1961 of *Swartzia laevicarpa* (3° S, Fig. 7b) (Worbes & Junk 1989) and rings 1960–1961 of Parana pine (27° S, Fig. 8). After the bomb peak, trees using stored carbohydrates would show higher ^{14}C concentrations in tree rings in comparison with those in the atmosphere. The case of rings 1965–1967 of spruce (47° N, Fig. 6a) may be an example. However, according to Grootes et al. (1989a, b), this effect should be small as the contribution of carbon from previous carbohydrate to the total carbon deposited as radial stem growth is less than 15% (and possibly 0%) (for the case of Sitka spruce grown in the Pacific coast of Washington).

A long series of maple leaves (1961–1993, McNeely 1994) in Ottawa (45° N) is also plotted in Figure 6b for comparison. Discrepancies were found: ^{14}C contents in leaves were clearly lower (before the bomb peak, 1961–1964) and higher (after the bomb peak, 1965–1993) than those in the atmosphere and in tree rings. The contribution of photosynthates from the previous year would be large for new season leaves and is a likely explanation for these discrepancies (Lowdon & Dyck 1974).

2) Trees in a subcanopy situation had a different radiocarbon content than that of the atmosphere, owing to CO_2 contribution from decomposition of material on and in the forest floor (Worbes & Junk 1989; Grootes et al. 1989a, b). This may cause the ^{14}C content in tree rings to differ from that in the atmosphere. The case of rings 1967–1971 of *Swartzia laevicarpa* (3° S, Fig. 7) can be explained by this reason (Worbes & Junk 1989). The contribution of this carbon to the total carbon deposited in tree rings is about 10–23% for the case of Sitka spruce (Grootes et al. 1989a, b).

3) (a) Local sources of 'dead' CO_2 (free of ^{14}C) from the combustion of fossil fuel may cause discrepancies. This could be the explanation of significantly depleted values of $\Delta^{14}C$ in rings 1945–1968 of oak in central New York city, compared to those in the atmosphere and in tree rings in a rural area at similar latitudes (41° N, Fig. 6b).

   (b) Local sources of ^{14}C from nuclear industries are another possibility, and can lead to higher ^{14}C concentrations in tree rings. The magnitude of the effect depends on the type and size of nuclear plants, on the distance between their locations and the tree sampling sites, and on local meteorological patterns. The effect was found to be significant around the Sellafield reactor/reprocessing plant in northern England (Otlet et al. 1992), intermediate around the Obrigheim power reactor in Germany (Levin et al. 1980), and slight around nuclear power plants in Sweden (Stenstrom et al. 1998) and Hungary (Uchrin et al. 1998).

4) Sampling of wood material too close to ring boundaries might also cause discrepancies when the ^{14}C content in earlier or later rings in the bomb pulse period are

significantly different from that in the current ring.

5) The pretreatment was not enough to remove mobile materials such as resins, sugars, waxes, oils, etc. which might cause discrepancies. Because they can migrate from earlier and/or later rings, the ^{14}C measured in a particular ring might be lower or higher than that of the atmosphere. In Table 1, the most common method of pretreatment was AAA, except for the studies by Muraki et al. (1998), Cain and Suess (1976), Levin et al. (1985), and Levin and Kromer (1997), which used solvent extraction in addition to AAA, and Murphy et al. (1997), which used holocellulose extraction. The AAA treatment does not remove all of the mobile materials, which are especially abundant in conifers (Head 1979; McPhail et al. 1983). Hertelendi and Csongor (1982) employed hot water and $NaClO_2$/HCl followed by AA (Alkali, Acid) to remove lignin and sugars but not resins, waxes, etc. Therefore the discrepancy of 5/28 may be due to the presence of mobile materials in pretreated wood. In a similar manner, the discrepancy of 3/14 for the case of Kozak et al. (1989) may be due to the lack of removal of mobile materials from wood in the pretreatment. It is probably no coincidence that the more complete pretreatment methods of Muraki et al. (1998), Cain and Suess (1976), Levin et al. (1985), Levin and Kromer (1997) and Murphy et al. (1997) have resulted in a high level of matching between tree ring and atmospheric radiocarbon data (10/10 for Muraki, 26/26 for Cain and Suess [red oak only], 27/29 for Levin and 11/12 for Murphy cases).

6) A more subtle effect may arise if the pretreatment method does not remove lignin, which may form within a ring later in the growing season (Fritts 1976); depending on the species, this might be up to several months after cellulose fibrils are formed. The AAA method, and even holocellulose extraction, do not completely remove lignin (Hoper et al. 1998), and in some circumstances it may be necessary to extract alpha-cellulose for determination of $\Delta^{14}C$ (Head 1979; Hoper et al. 1998; Tans et al. 1978), especially when heartwood is used for determination of atmospheric ^{14}C. Organic materials in outermost sapwood rings move across the heartwood – sapwood transition during the formation of heartwood. This contemporaneous carbon, amounting to as much as 15% of the total carbon in the ring, can only be removed from heartwood by isolating the pure cellulose in the wood (Cain & Suess 1976). In addition, the suitability of pure cellulose for the determination of atmospheric ^{14}C was supported by Grootes et al. (1989a, b), as they found the tree cellulose ^{14}C closely follows atmospheric $^{14}CO_2$ concentrations with a delay of 5 to 6 weeks.

Discrepancies can also be explained by combinations of the above factors. For example, discrepancies in the cases of rings 1958–1961 of *Swartzia laevicarpa* (3° S, Fig. 7), rings 1960–1961 of Parana pine (27° S, Fig. 8), and rings 1965–1967 of spruce (47° N, Fig. 6a) can be accounted for by the first factor – the use of stored carbohydrate from the previous year to form the current year ring in trees. These

cases might also be explained by the fifth factor – the lack of a proper pretreatment of tree rings – because the pretreatment employed in all three cases was AAA. The case of rings 1967–1971 of *Swartzia laevicarpa* (3° S, Fig. 7) is another example where discrepancies may be explained by a combination of the second factor – the difference of ^{14}C content between air around subcanopy trees and the regional atmosphere – and the fifth factor (AAA treatment was employed in this case).

The discrepancies also vary from species to species. Merkusii pine (11° N, Fig. 7) from 1962–1967 and Teak (7° S, Fig. 7) from 1957–1970 were prepared and measured by the same group of researchers (Kikata et al. 1992, 1993; Yonenobu et al. 1995). No discrepancy was found for Merkusii pine (highly resinous), while 6 out of 14 rings of Teak (not thought to be highly resinous) had $\Delta^{14}C$ lower than that of the atmosphere (Table 1). The case of ring 1964 AD of oak (51° N, Fig. 6c) is another example. Oak uses more stored materials from earlier years than do other species, so the ^{14}C content in 1964 AD ring of oak (51° N, Fig. 6c) is much higher than that in the atmosphere (Worbes & Junk 1989).

A small peak extending for several years around 1980, evident in tree rings from eastern China (Dai et al. 1992, Fig. 6a), maple leaves from eastern Canada (McNeely 1994, Fig. 6b) and, to a lesser degree, in tree rings from central Japan (Nakamura et al. 1987a, b, Fig. 6a), does not appear in atmospheric and tree-ring data from Europe at that time (Levin & Kromer 1997; Levin et al. 1985; Levin et al. 1994; Hertelendi & Csongor 1982; Kozak et al. 1989; Worbes & Junk 1989; Fig. 6c), nor in another short atmospheric data set from western USA (1977–1983, Berger et al. 1987). We must therefore conclude that some of the data sets are erroneous, or that the source of increased ^{14}C levels is somehow regional or local (such as nuclear industry – the third factor), and not injected into the stratosphere and circulated through the troposphere.

CONCLUSION

In order to avoid the problems – discrepancies between the ^{14}C content in tree rings and in the atmosphere – we should: 1) use canopy trees in an open forest situation, far from local sources of contamination, and not subcanopy trees, for determination of ^{14}C; 2) avoid species which use significant quantities of photosynthates from the previous year; 3) employ a good sampling strategy with proper cross-dating and verification of year of growth, with subsampling of wood material well clear of ring boundaries; 4) employ a more careful pretreatment to extract alpha-cellulose from wood, so that the results reflect atmospheric ^{14}C in the growing season; and 5) measure additional time series of various tree species. This will also allow us to determine if there are discrepancies from the use of stored materials from previous years to form the current ring in trees or from the pretreatment of wood for dating, or both. With these protocols, tree rings can be used to obtain high-quality data, from the tree-growing season, for regions where direct atmospheric sampling was not done in the 1940s–

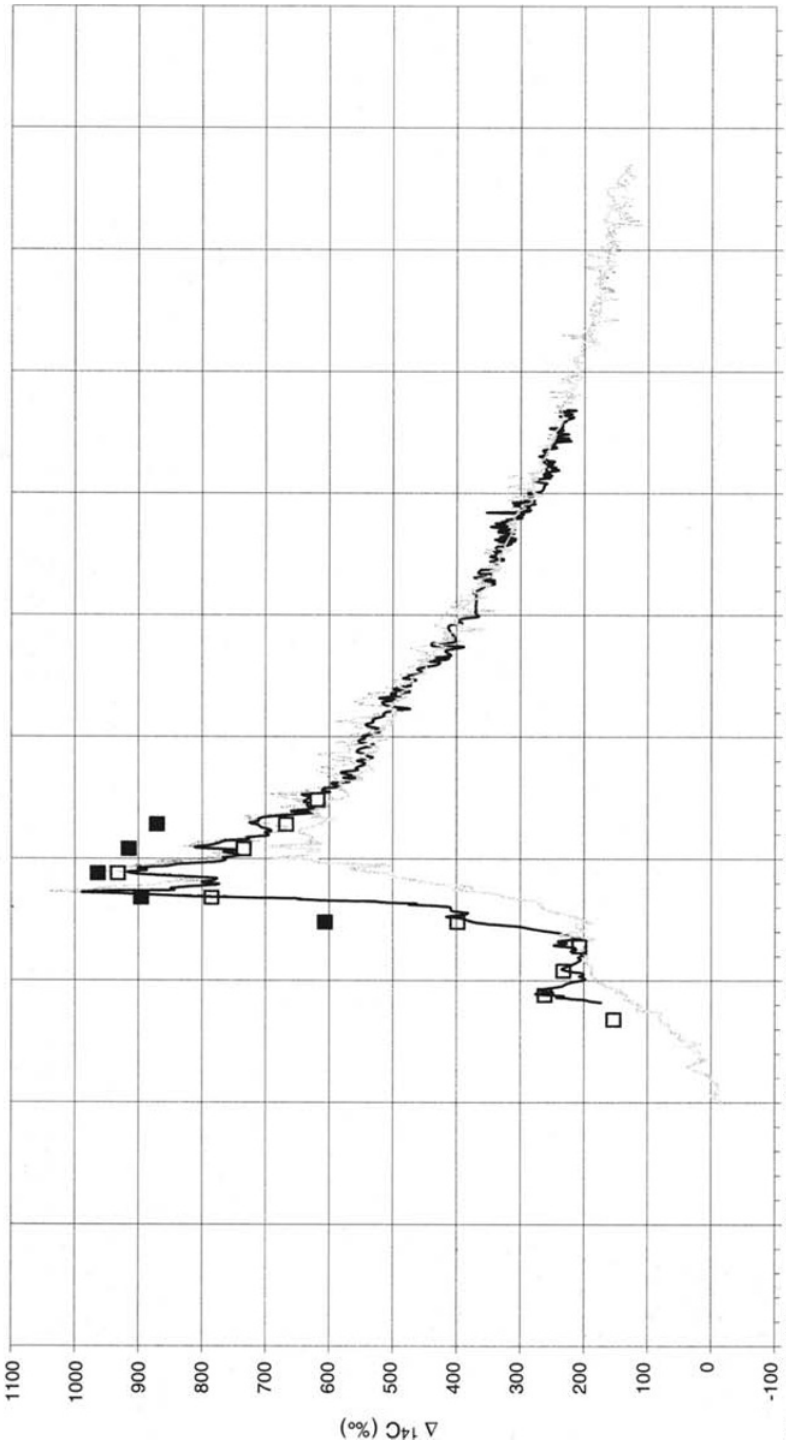

Figure 5: $\Delta^{14}C$ in the Northern Hemisphere at high latitudes. Lines and dots represent atmospheric and tree-ring data, respectively. Error bars are too small to be shown.

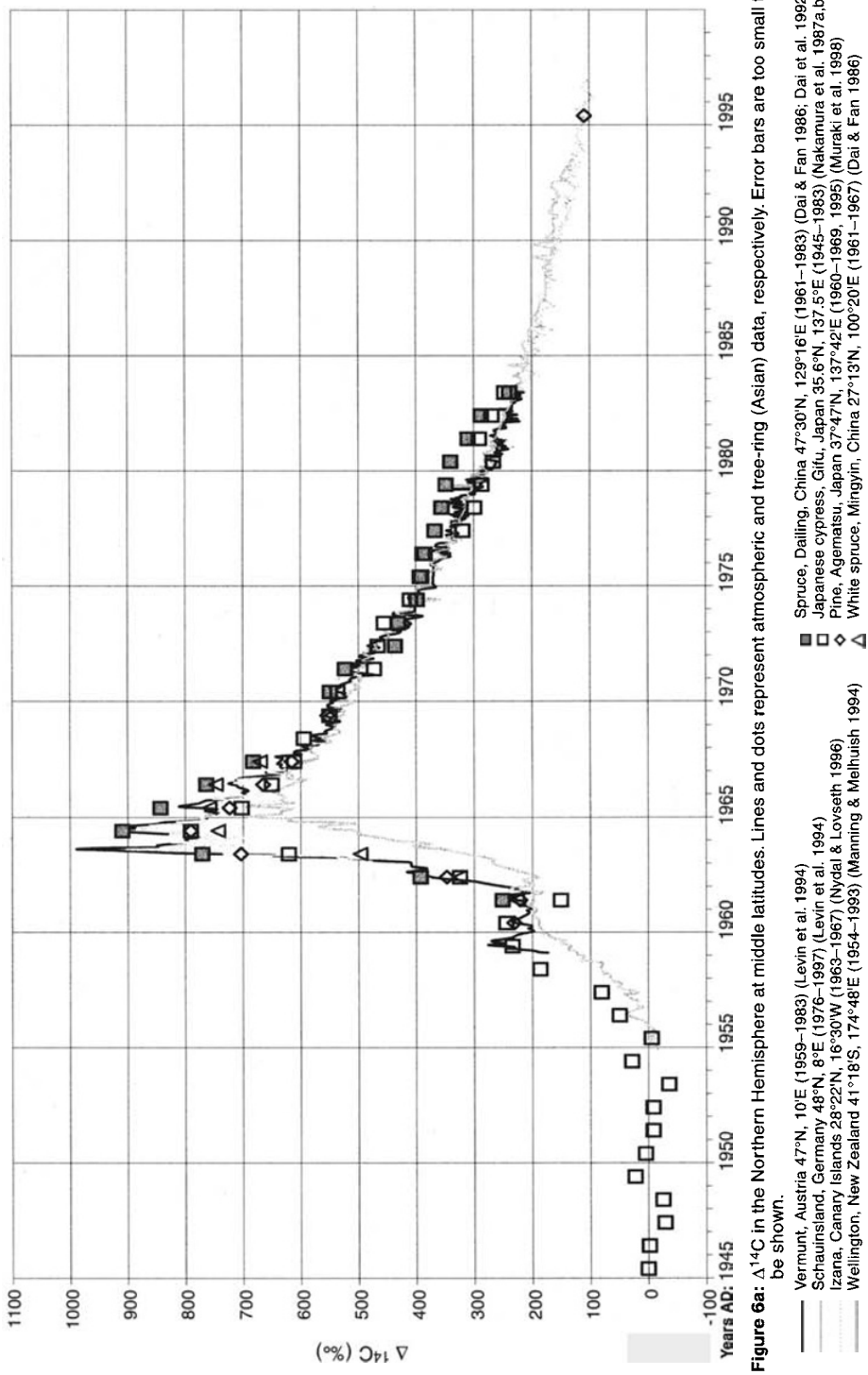

Figure 6a: $\Delta^{14}C$ in the Northern Hemisphere at middle latitudes. Lines and dots represent atmospheric and tree-ring (Asian) data, respectively. Error bars are too small to be shown.

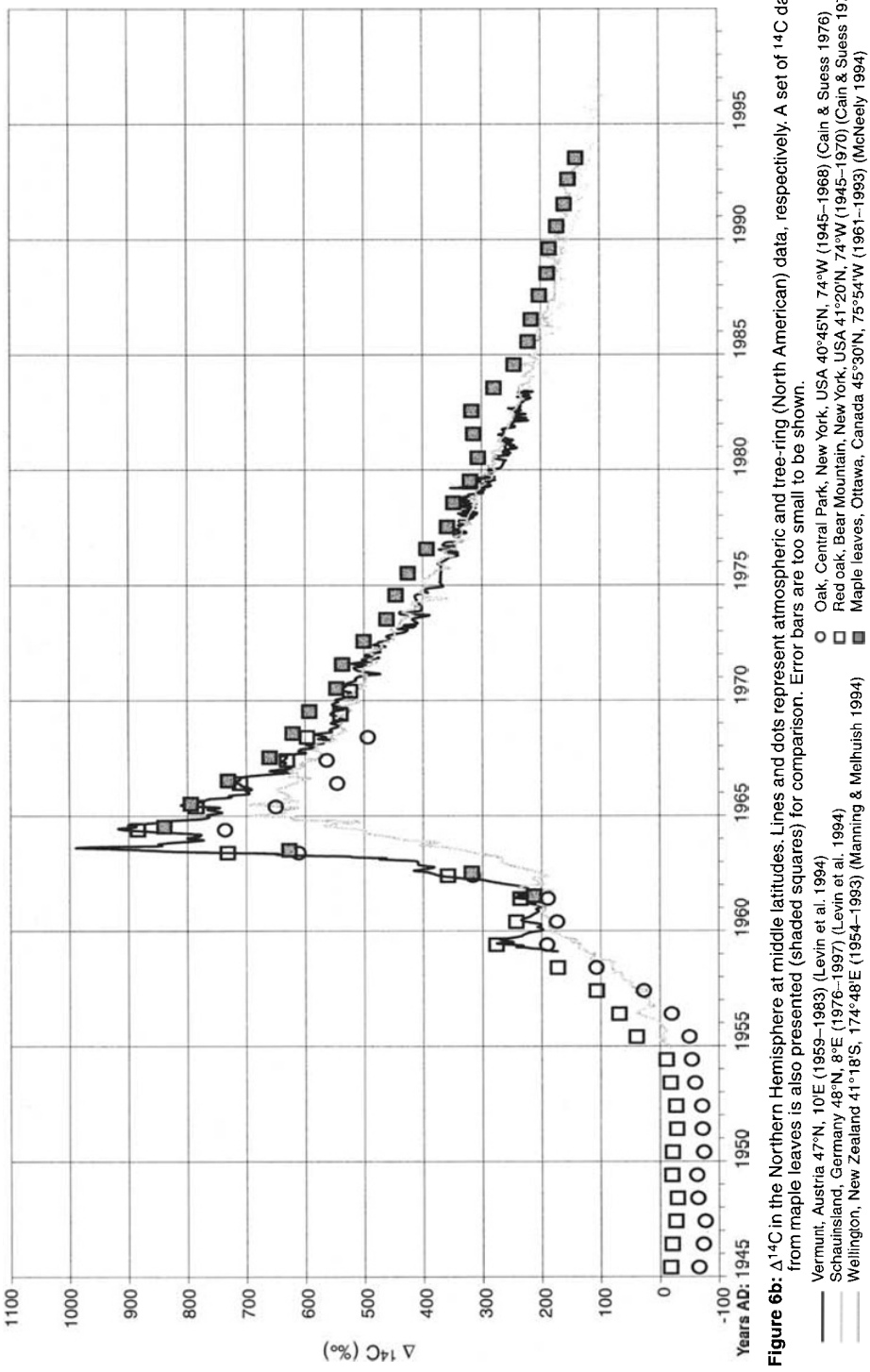

Figure 6b: $\Delta^{14}C$ in the Northern Hemisphere at middle latitudes. Lines and dots represent atmospheric and tree-ring (North American) data, respectively. A set of ^{14}C data from maple leaves is also presented (shaded squares) for comparison. Error bars are too small to be shown.

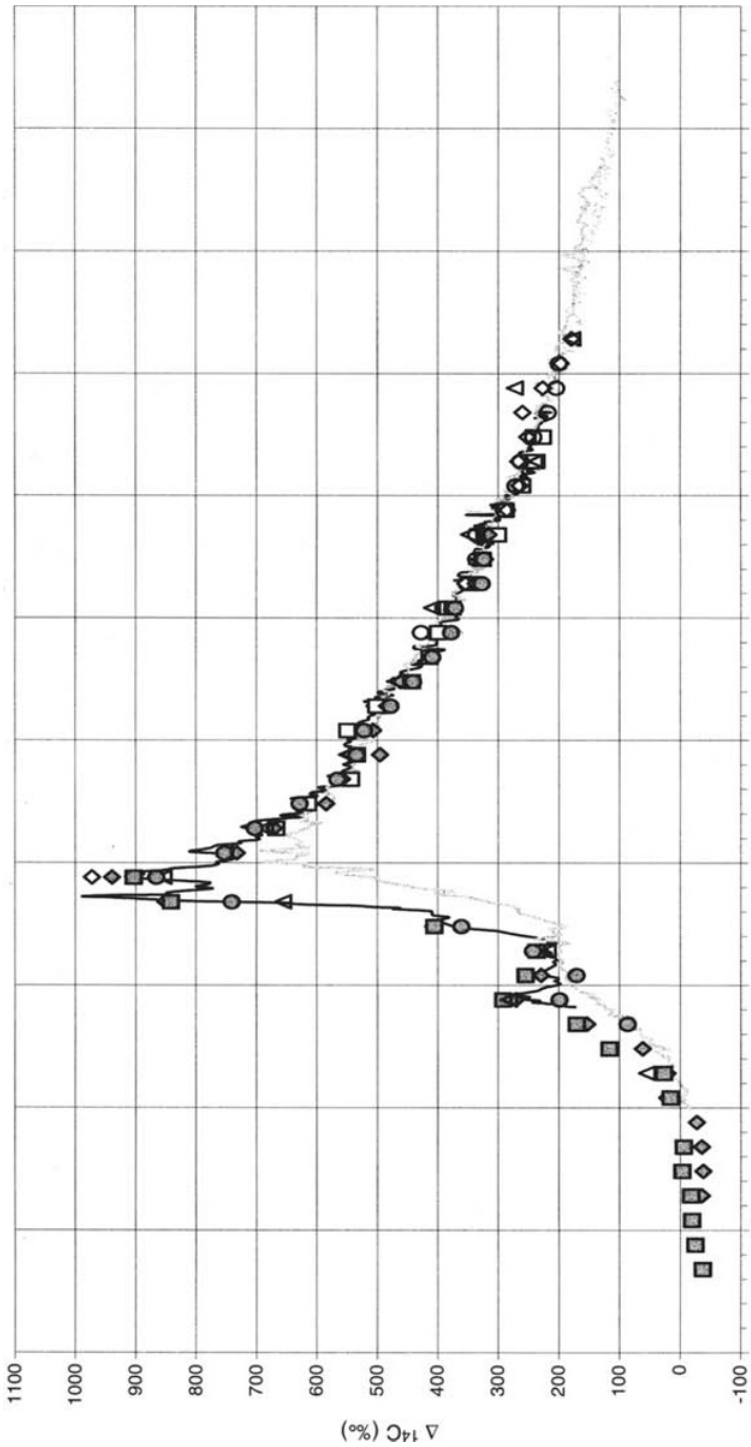

Figure 6c: $\Delta^{14}C$ in the Northern Hemisphere at middle latitudes. Lines and dots represent atmospheric and tree-ring (European) data, respectively. Error bars are too small to be shown.

- Vermunt, Austria 47°N, 10°E (1959–1983) (Levin et al. 1994)
- Wellington, New Zealand 41°18'S, 174°48'E (1954–1993) (Manning & Melhuish 1994)
- Schauinsland, Germany 48°N, 8°E (1976–1997) (Levin et al. 1994)
- Spruce, Schauinsland, Germany 48°N, 8°E (1974–1985) (Levin & Kromer 1997)
- Spruce, Matra Mt, Hungary 47°54'N, 19°55'E (1956–1986) (Kozak et al. 1989)
- Pine, Obrigheim, Germany 49°N, 9°E (1966–1982) (Levin et al. 1985)
- Oak, Solling, Germany 51°45'N, 9°30'E (1964, 1976–1986) (Worbes & Junk 1989)
- Acacia, Hungary 47°35'N, 21°35'E (1951–1978) (Hertelendi & Czongor 1982)
- Elm, Kiel, Germany 54°20'N, 10°7'E (1948–1964) (Willkomm & Erlenkeuser 1968)
- Poplar, Ulm, Germany 48°24'N, 10°E (1958–1977) (Eichinger et al. 1980)

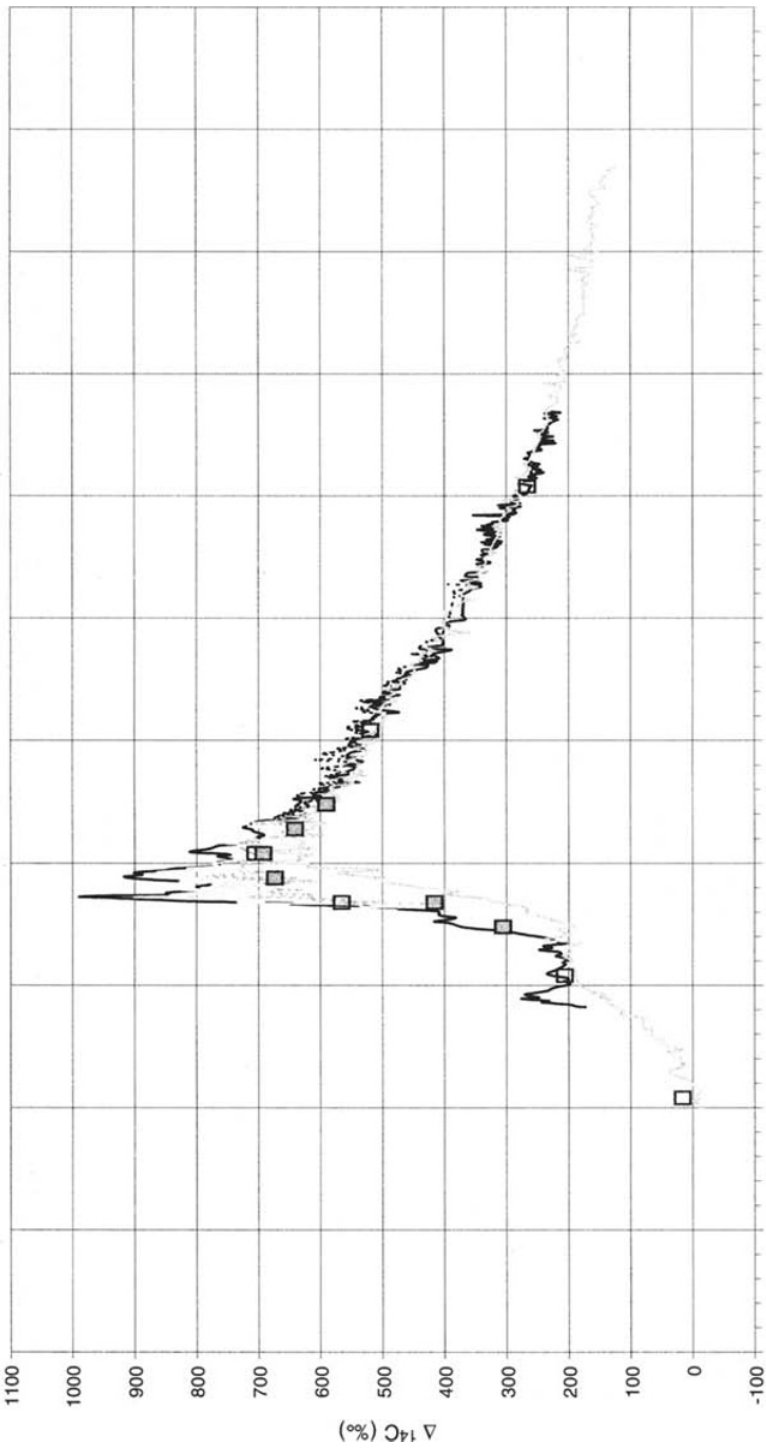

Figure 7a: $\Delta^{14}C$ in the Northern Equatorial Regions. Lines and dots represent atmospheric and tree-ring data, respectively. Error bars are too small to be shown.

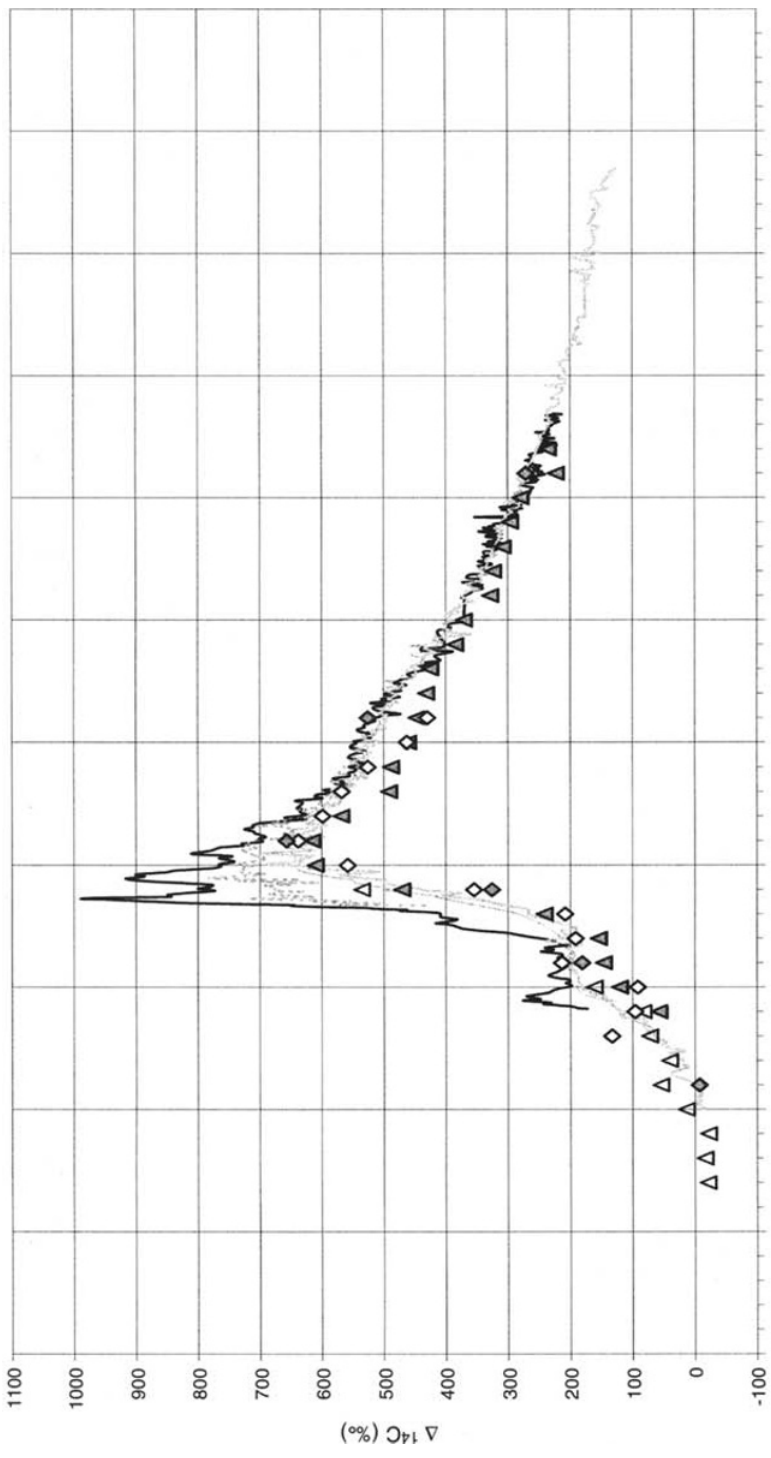

Figure 7b: $\Delta^{14}C$ in the Southern Equatorial Regions. Lines and dots represent atmospheric and tree-ring data, respectively. Error bars are too small to be shown.

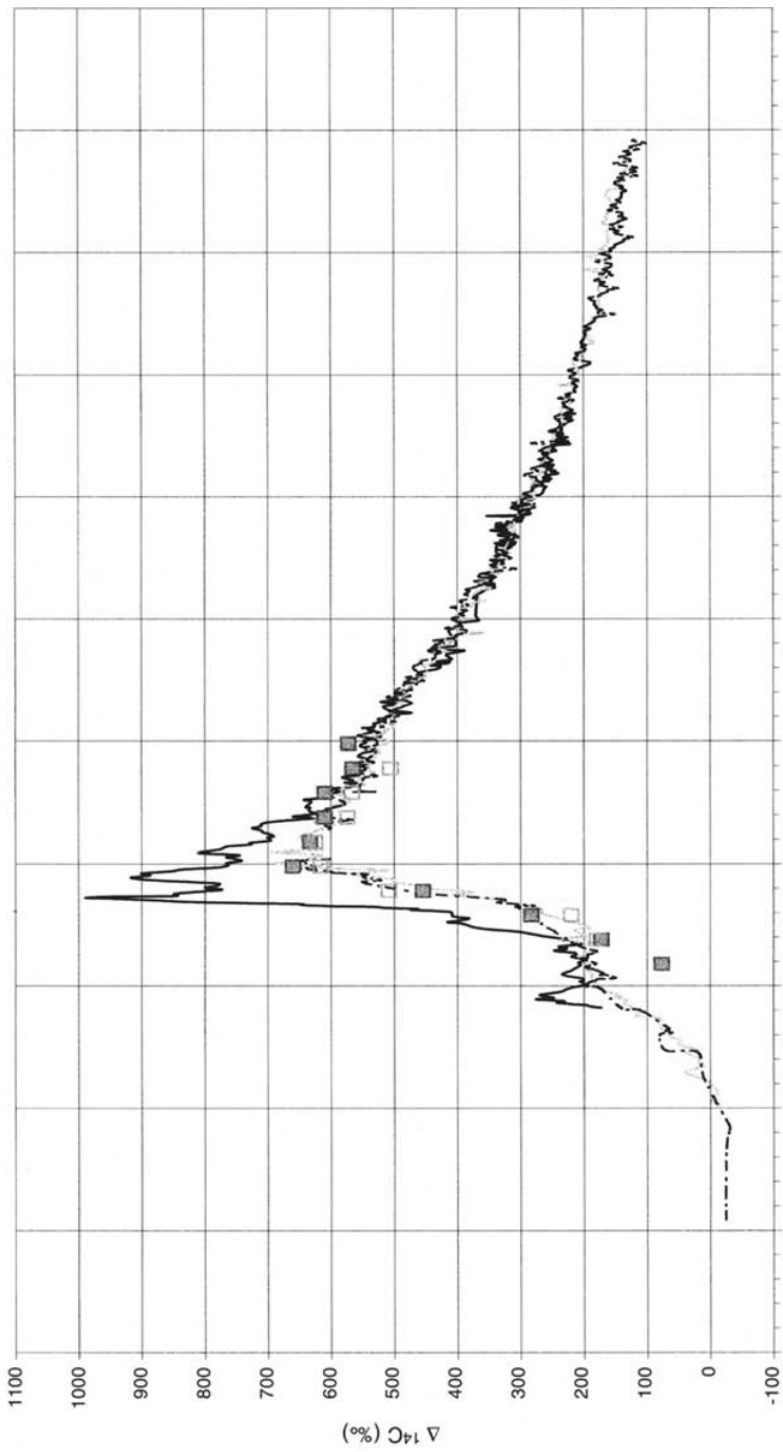

Figure 8: $\Delta^{14}C$ in the Southern Hemisphere at middle latitudes. Lines and dots represent atmospheric and tree-ring data, respectively. Error bars are too small to be shown.

1990s.

(text continued on page 281)

FURTHER RESEARCH

We are using two sets of tree rings at two different sites having different air mass trajectories to examine the atmospheric ^{14}C excess after 1955 resulting from atmospheric nuclear testing. One set is from a medium-sized *Pinus kesiya* with clear annual ring structure (Buckley et al. 1995; D'Arrigo et al. 1997) grown in northwestern Thailand at 19° N. The other is *Lagarostrobos franklinii* grown in northwestern Tasmania, Australia at 42° S (Buckley et al. 1997). Twenty-four consecutive single rings for each set from 1952 to 1975 AD will be measured by Accelerator Mass Spectrometry (AMS) to reconstruct and extend the bomb pulse curve. The time frame covers the background from 1952–1955 AD, the bomb peak period, and a decade after the bomb peak (1964–1975 AD). Because of very different levels of ^{14}C in each ring, our strategy for sampling is to avoid the ring boundary areas. The middle part of the earlywood of each ring, ~0.2 mm away from each boundary was selected. We have employed a careful method of pretreatment to extract alpha cellulose for dating (Head 1979). Finally, our measured data will be compared with published data in order to gain a better understanding of global carbon cycle and air-sea interactions.

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REFERENCES

Berger, R., T.B. Jackson, R. Michael & H.E. Suess. 1987. Radiocarbon content of tropospheric CO_2 at China Lake, California 1977–1983. Radiocarbon 29: 18–23.

Buckley, B.M., M. Barbetti, M. Watanasak, D.R. D'Arrigo, S. Boonchirdchoo & S. Sarutanon. 1995. Dendrochronological investigations in Thailand. IAWA J. 16: 393–409.

Buckley, B.M., E.R. Cook, M.J. Peterson & M. Barbetti. 1997. A changing temperature response with elevation for Lagarostrobos franklinii in Tasmania, Australia. Climatic Change 36: 477–498.

Cain, W.F. & H.E. Suess. 1976. Carbon 14 in tree rings. J. Geophys. Res. 81: 3688–3694.

Dai, K.M. & C.Y. Fan. 1986. Bomb produced ^{14}C content in tree rings grown at different latitudes. Radiocarbon 28: 346–349.

Dai, K.M., Y. Qian & C.Y. Fan. 1992. Bomb-produced ^{14}C in tree rings. Radiocarbon 34: 753–756.

D'Arrigo, R.D., M. Barbetti, M. Watanasak, B.M. Buckley, P. Krusic, S. Boonchirdchoo & S. Sarutanon. 1997. Progress in dendroclimatic studies of mountain pine in northern Thailand. IAWA J. 18: 433–444.

Eichinger, L., W. Rauert & M. Wolf. 1980. ^{14}C-Messungen an Weinen und Baumringen. Ber. Gesells. f. Strahlen- u. Umweltforsch. R 250: 105–118.

Enting, I.G. 1982. Nuclear weapons data for use in carbon cycle modelling. CSIRO Division of Atmospheric Physics Technical Paper No. 44. CSIRO, Melbourne.

Enting, I.G. & J.V. Mansbridge. 1987. Preliminary studies with a two-dimensional model using transport fields derived from a GCM. CSIRO Division of Atmospheric Physics Research Technical Paper No. 14. CSIRO, Melbourne.

Fritts, H.C. 1976. Tree rings and climate. Academic Press, London.

Grootes, P.M., G.W. Farwell, F.H. Schmidt, D.D. Leach & M. Stuiver. 1989a. Importance of biospheric CO_2 in a subcanopy atmosphere deduced from ^{14}C AMS measurements. Radiocarbon 31: 475–480.

Grootes, P.M., G.W. Farwell, F.H. Schmidt, D.D. Leach & M. Stuiver. 1989b. Rapid response of tree cellulose radiocarbon content to changes in atmospheric $^{14}CO_2$ concentration. Tellus 41B: 134–148.

Head, J. (ms.). 1979. Structure and chemical properties of fresh and degraded wood. MSc thesis. Australian National University, Canberra.

Hertelendi, E. & E. Csongor. 1982. Anthropogenic ^{14}C excess in the troposphere between 1951 and 1978 measured in tree rings. Radiochem. Radioanal. Letters 56: 103–110.

Hoper, S.T., F.G. McCormac, A.G. Hogg, T.F.G. Higham & M.J. Head. 1998. Evaluation of wood pretreatment on oak and cedar. Radiocarbon 40: 45–50.

Kikata, Y., H. Yonenobu, F. Morishita & Y. Hattori. 1992. ^{14}C concentrations in tree stems. Bull. Nagoya. Univ. Furukawa Mus. 8: 41–46.

Kikata, Y., H. Yonenobu, F. Morishita, Y. Hattori & S.N. Marsoem. 1993. ^{14}C concentrations in tree stems I. Mokuzai Gakkaishi 39: 333–337.

Kolesnikov, N.V., I.A. Gorshkova & Yu. F. Biryulin. 1970. Variation of the radiocarbon concentration in the atmosphere during the period 1957–1968 (according to dendrological data). Atmospheric and Oceanic Physics 6: 647–649.

Kozak, K., B. Obelic & N. Horvatincic. 1989. Tritium and ^{14}C in tree rings of the last three decades. Radiocarbon 31: 766–770.

Levin, I. & B. Kromer. 1997. Twenty years of atmospheric $^{14}CO_2$ observations at Schauinsland station, Germany. Radiocarbon 39: 205–218.

Levin, I., B. Kromer & R.J. Francey. 1996. Continuous measurements of ^{14}C in atmospheric CO_2 at Cape Grim. In: R.J. Francey, A.L. Dick & N. Derek (eds.), Baseline Atmospheric Program Australia 1994–1995: 106–107. CSIRO, Melbourne.

Levin, I., B. Kromer, H. Schoch-Fischer, M. Bruns, M. Munnich, D. Berdau, J.C. Vogel & K.O. Munnich. 1985. 25 years of tropospheric ^{14}C observations in Central Europe. Radiocarbon 27: 1–19.

Levin, I., B. Kromer, H. Schoch-Fischer, M. Bruns, M. Munnich, D. Berdau, J.C. Vogel & K.O. Munnich. 1994. $\Delta^{14}CO_2$ records from two sites in Central Europe. In: T.A. Boden, D.P. Kaiser, R.J. Sepanski & F.W. Stoss (eds.), Trends 93 – A compendium of data on global change: 203–222 and online updates (Online Trends). Carbon Dioxide Information Analysis Centre. Oak Ridge National Laboratory, Oak Ridge, TN. ORNL/CDIAC-65.

Levin, I., K.O. Munnich & W. Weiss. 1980. The effect of anthropogenic CO_2 and ^{14}C sources on the distribution of ^{14}C in the atmosphere. Radiocarbon 22: 379–391.

Lowdon, J.A. & W. Dyck. 1974. Seasonal variations in the isotope ratios of carbon in maple leaves and other plants. Can. J. Earth Sci. 11: 79–88.

Manning, M.R., D.C. Lowe, W.H. Melhuish, R.J. Sparks, G. Wallace, C.A.M. Brenninkmeijer

& R.C. McGrill. 1990. The use of radiocarbon measurements in atmospheric studies. Radiocarbon 32: 37–58.

Manning, M.R. & W.H. Melhuish. 1994. $\Delta^{14}CO_2$ record from Wellington. In: T.A. Boden, D.P. Kaiser, R.J. Sepanski & F.W. Stoss (eds.), Trends 93 – A compendium of data on global chang: 173–202 and online updates (Online Trends). Carbon Dioxide Information Analysis Centre. Oak Ridge National Laboratory, Oak Ridge, TN. ORNL/CDIAC-65.

McNeely, R. 1994. Long-term environmental monitoring of ^{14}C levels in the Ottawa region. Environment International 20: 675–679.

McPhail, S., M. Barbetti, R. Francey, T. Bird & J. Dolezal. 1983. ^{14}C Variations from Tasmanian trees – Preliminary results. Radiocarbon 25: 797–802.

Muraki, Y., G. Kocharov, T. Nishiyama, Y. Naruse, T. Murata, K. Masuda & Kh.A. Arslanov. 1998. The new Nagoya radiocarbon laboratory. Radiocarbon 40: 177–182.

Murphy, J.O., E.M. Lawson, D. Fink, M.A.C. Hotchkis, Q. Hua, G.E. Jacobsen, A.M. Smith & C. Tuniz. 1997. ^{14}C AMS measurements of the bomb pulse in N- and S-hemisphere tropical trees. Nucl. Instr and Meth. in Phys. Res. 123B: 447–450.

Nakai, N. & T. Nakamura. 1989. Direct detection of radiocarbon using Tandetron Accelerator Mass Spectrometer and the application to age determination. Bull. Nagoya Univ. Furukawa Mus. 5: 139–149.

Nakamura, T., N. Nakai, M. Kimura, S. Ohishi, Y. Hattori & Y. Kikata. 1987b. Variations in ^{14}C concentrations of tree rings (1945–1983). Chikyu-Kagaku (Geochemistry) 21: 7–12.

Nakamura, T., N. Nakai & S. Ohishi. 1987a. Applications of environmental ^{14}C measured by AMS as a carbon tracer. Nucl. Instr and Meth. in Phys. Res. 29B: 355–360.

Nydal, R. & J.S. Gislefoss. 1996. Further application of bomb ^{14}C as a tracer in the atmosphere and ocean. Radiocarbon 38: 389–406.

Nydal, R. & K. Lovseth. 1996. Carbon-14 measurement in atmospheric CO_2 from Northern and Southern Hemisphere sites, 1962–1993. Carbon Dioxide Information Analysis Center – World Data Center-A for Atmospheric Trace Gases – Oak Ridge National Laboratory, Tennessee.

Otlet, R.L., M.J. Fulker & A.J. Walker. Environmental impact of atmospheric carbon-14 emissions resulting from the nuclear energy cycle. In: R.E. Taylor, A. Long & R.S. Kra (eds.), Radiocarbon after four decades: An interdisciplinary perspective: 519–534. Springer-Verlag, New York.

Stenstrom, K., G. Skog, C. Thornberg, B. Erlandsson, R. Hellborg, S. Mattsson & P.Persson. 1998. ^{14}C levels in the vicinity of two Swedish nuclear power plants and at two 'clean-air' sites in southernmost Sweden. Radiocarbon 40: 433–438.

Stuiver, M. & H.A. Polach. 1977. Reporting of ^{14}C data. Radiocarbon 19: 353–363.

Tans, P.P., A.F.M. De Jong & W.G. Mook. 1978. Chemical pretreatment and radial flow of ^{14}C in tree rings. Nature 271: 234–235.

Uchrin, G., E. Hertelendi, G. Volent, O. Slavik, J. Moravek, I. Kobal & B. Vokal. 1998.^{14}C Measurements at PWR-type nuclear power plants in three middle European countries. Radiocarbon 40: 439–446.

Vogel, J.C. & M. Marais. 1971. Pretoria radiocarbon dates I. Radiocarbon 13: 378–394 and regular updates (J.C. Vogel, pers. comm.).

Willkomm, H. & H. Erlenkeuser. 1968. University of Kiel radiocarbon measurements III. Radiocarbon 10: 328–332.

Worbes, M. & W.J. Junk. 1989. Dating tropical trees by means of ^{14}C from bomb tests. Ecology 70: 503–507.

Yonenobu, H., F. Morishita, Y. Hattori, Y. Kikata & S.N. Marsoem. 1995. Growth rates of tropical species from the ^{14}C concentration variations in tree stems. In: Proceedings of the International Workshop on Asian and Pacific Dendrochronology, 4–9 March 1995, Tsukuba, Japan: 210–215.