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TWENTY YEARS OF ATMOSPHERIC $^{14}CO_2$ OBSERVATIONS AT SCHAUINSLAND STATION, GERMANY

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ABSTRACT. We present and discuss quasi-continuous long-term $^{14}CO_2$ observations from the continental background station Schauinsland (48°N, 8°E, 1205 m asl, Black Forest, southern Germany). The observed steady decline of atmospheric $^{14}CO_2$ from 1977 to 1996 can be described by a single exponential function with an e-folding time of (16.3 ± 0.2) yr. Summer means (May to August) in atmospheric $^{14}CO_2$ at Schauinsland compare within $\Delta^{14}C = \pm 4‰$ with measurements made on individual rings from a tree grown in the near vicinity of the Schauinsland site. Both data sets are slightly depleted by up to 5‰ if compared to maritime background measurements of atmospheric $^{14}CO_2$ made at Izaña, Tenerife. This is due to the influence of fossil fuel CO_2 emissions over the European continent as well as generally in mid latitudes of the Northern Hemisphere. $\delta^{13}C$ analyses from the Schauinsland samples show mean seasonal variations with an amplitude of ±0.4‰, caused by atmosphere-biosphere exchange, and a mean decrease from 1977 to 1996 of $\delta^{13}C = -0.017‰\ yr^{-1}$. This trend is mainly due to an increasing quantity of fossil fuel CO_2 in the atmosphere, depleted in $^{13}C/^{12}C$ ratio, and compares well to trends measured at other stations in mid-to-high northern latitudes.

INTRODUCTION

During atmospheric nuclear weapon testing in the 1950s and early 1960s, large amounts of radiocarbon were generated in the atmosphere. This artificial ^{14}C input caused a global increase of the $^{14}C/^{12}C$ ratio in atmospheric CO_2 by a factor of almost two in 1963 (see Fig. 1), leading to a substantial disequilibrium of ^{14}C between atmosphere, biosphere and surface ocean water. In the last 20 years, this atmospheric ^{14}C perturbation has been used extensively to investigate CO_2 cycling between the atmosphere and the rapidly exchanging ocean and biosphere reservoirs (*e.g.*, Stuiver 1980; Druffel and Suess 1983; Goudriaan 1992). More recent quantitative attempts to budget bomb ^{14}C in the global carbon system, however, led to evidence of a serious imbalance (Hesshaimer, Heimann and Levin 1994) that has still not been resolved. Nevertheless, constraints on exchange rates provided by bomb ^{14}C are largely strengthened with the length of the observational record of bomb ^{14}C decline in the atmosphere.

We here present our extended data set of atmospheric $^{14}CO_2$ observations from continental Europe for three main purposes. First, we want to make this record available to serve as an input function for global carbon cycle modeling. Second, for investigations of anthropogenic perturbations such as regional contamination by ^{14}C-free fossil fuel CO_2 emissions or by releases of $^{14}CO_2$ by nuclear power plants, the Schauinsland station can serve as an ideal reference, at least for Central Europe. Third, our record may be applied in dating young (post-bomb) organic materials where the exact time-dependent $^{14}CO_2$ level is needed as a reference.

^{14}C data of individual samples from the two Central European sites—Vermunt, Austrian Alps (47°N, 10°E, 1800 m asl) and Schauinsland, Black Forest, Germany (48°N, 8°E, 1205 m asl)—have already been published and provided in tabulated form through 1983 by Levin *et al.* (1985), together with some earlier Heidelberg $^{14}CO_2$ data from a number of other sites, also in the Southern Hemisphere (see also Fig. 1). The extended data set from Schauinsland station was discussed in detail by Levin, Graul and Trivett (1995) and the individual data were made available through the Carbon Dioxide Information Analysis Center (DIAC) (Levin *et al.* 1994). Here we will provide, in tabulated

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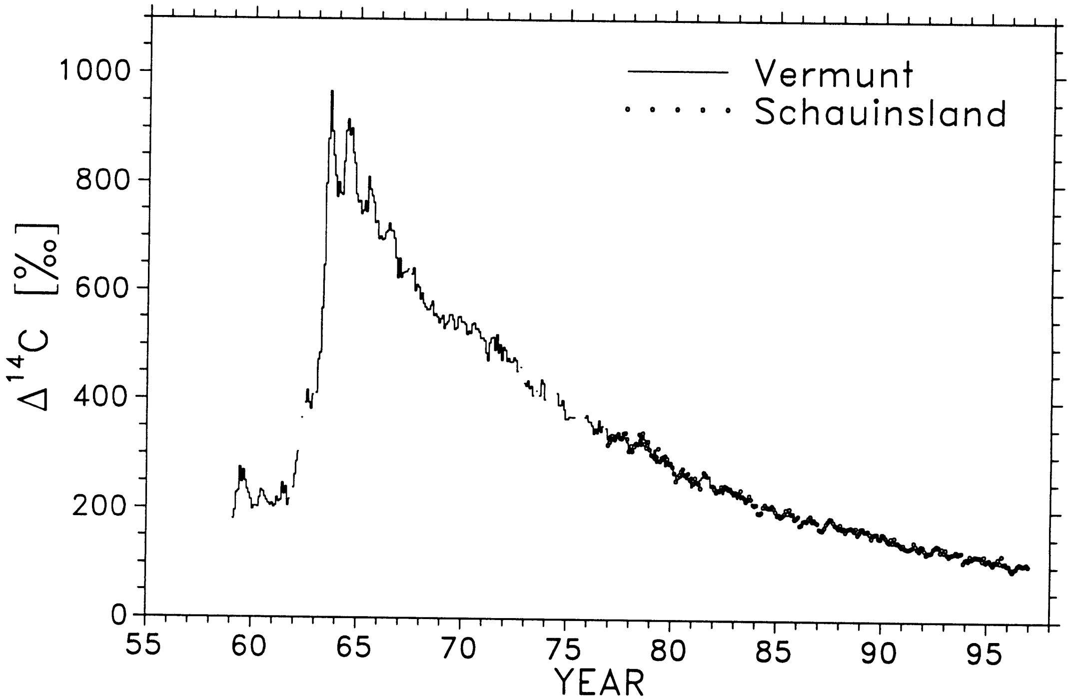

Fig. 1. Long-term observations of $\Delta^{14}C$ in atmospheric CO_2 in the Northern Hemisphere (Levin *et al.* 1985 with extended data from Table 1). The Vermunt record overlaps with Schauinsland data from 1977 to 1983. Shortly after the atmospheric test ban treaty in 1962, the $^{14}CO_2$ level in the Northern Hemisphere was twice as high as the natural equilibrium value. $\Delta^{14}C$ decreases thereafter due to equilibration with the world oceans and the terrestrial biosphere.

form (Table 1, Appendix), the Schauinsland data from 1983 onwards extended until the end of 1996 (monthly mean values as shown in Figs. 1 and 2 will be made available via FTP). In addition, as a service for dating purposes we present summer means together with ^{14}C data from wood samples of a tree grown in the immediate vicinity of the Schauinsland site (Table 2, Appendix).

SAMPLING SITE

The regional GAW (Global Atmosphere Watch of the World Meteorological Organisation) station Schauinsland (48°N, 8°E) is located close to the top of Schauinsland Mountain in the Black Forest, southern Germany, at an elevation of 1205 m asl. The observatory is run by the Federal Environmental Agency (UBA), Berlin. At a height of more than 1000 m above the Upper Rhine Valley, the station is usually above the ground-level inversion layer of the valley, but during daytime and particularly in the summer months contamination from Rhine Valley pollutant sources may regularly occur. Surrounding the station are pastures and forest areas. The station itself is set up in a remote building with electrical heating only. Occasional local contamination is possible only from local traffic of the station personnel. As was shown from continuous CO_2 measurements performed at the Schauinsland site by UBA (Levin, Graul and Trivett 1995), during moderate and strong winds the station samples air representative of mean atmospheric conditions over Western Europe at this elevation of *ca.* 1000 m asl.

Measurement Techniques

All air samples were collected from a ventilated intake stack at an elevation of *ca.* 7 m above local ground. CO_2 samples integrated over two weeks from *ca.* 15–20 m^3 of air were continuously collected by dynamic quantitative absorption in carbonate-free sodium hydroxide solution as described in detail by Levin, Münnich and Weiss (1980). ^{13}C analyses of the CO_2 were performed by mass spectrometry, ^{14}C analyses by high-precision proportional counting of the purified CO_2 sample (Schoch *et al.* 1980; Kromer and Münnich 1992). $\delta^{13}C$ values are given relative to the VPDB standard (Hut 1987); the overall precision of a single analysis is typically ±0.15‰. Conventionally $\delta^{13}C$-corrected $\Delta^{14}C$ data are given relative to NBS oxalic acid activity corrected for decay (Stuiver and Polach 1977); the precision of a single $\Delta^{14}C$ measurement is typically ±(3–5)‰. Tree-ring samples were pretreated following the procedure outlined by Kromer and Becker (1993), which is Soxhlet extraction followed by the A-A-A sequence.

Results and Discussion

Long-Term Trend of $\Delta^{14}CO_2$ in Central Europe

Since the nuclear test ban treaty in 1962, 35 years of atmospheric $^{14}CO_2$ observations, typical of well-mixed air over Central Europe, are now available (Fig. 1). They complement data sets by other groups performed at northern hemispheric background sites (Nydal and Lövseth 1996) as well as in polluted areas (Kuc 1989). $^{14}CO_2$ in the Northern Hemisphere was dominated in the early sixties by large seasonal variations that are caused by seasonal input of bomb ^{14}C-rich air from the stratosphere into the northern hemispheric troposphere (Hesshaimer and Levin, submitted). The subsequent bomb ^{14}C decline observed after 1963 mainly reflects the $^{14}CO_2$ exchange fluxes with the ocean and the biosphere, which are governed by the internal circulation dynamics within these two reservoirs. But anthropogenic CO_2 emissions also contribute to the observed $^{14}CO_2$ decline, whereas ^{14}C emissions from the nuclear industry slightly counteract (by <1.5‰ per year) these effects (Hesshaimer, Heimann and Levin 1994). Figure 1 shows the combined data sets from Vermunt and Schauinsland. As discussed earlier, during the period of overlapping samples (1977–1983), results from both stations agree very well within measurement accuracy (Levin *et al.* 1985).

$\Delta^{14}CO_2$ at Schauinsland Station

The complete record of monthly mean $\Delta^{14}CO_2$ data from the Schauinsland site is displayed in Figure 2. The $^{14}C/^{12}C$ ratio shows a steady and approximately exponential decrease from 1977 until today with a time constant of $\tau = (16.3 \pm 0.2)$ yr. Overlying this trend is a seasonality with minimum values occurring during the winter months. ^{14}C analyses of individual tree rings (*Picea abies*) from 1974 to 1985, collected in the near vicinity of the Schauinsland station, are also displayed in Figure 2. The tree-ring ^{14}C data closely match the summer values of the air samples averaged over the months May to August. Both data sets closely follow the upper envelope of the continuous atmospheric record, and are assumed to be representative for the respective growing seasons in Western and Central Europe.

The ^{14}C background level in mid latitudes of the Northern Hemisphere can be derived from observations at the GAW station Izaña, Tenerife (28°N, 16°W, 2376 m asl) and from the High Alpine Research Station Jungfraujoch in the Swiss Alps (47°N, 8°E, 3454 m asl). At these sites, quasi-continuous $^{14}CO_2$ samples have been measured beginning in 1984 and 1986, respectively (Levin *et al.* 1992, and unpublished Heidelberg data). In the period of 1986 to 1995 the Schauinsland $^{14}CO_2$ level during the summer months (May–August) is on average lower by $\Delta^{14}C = (1.8 \pm 0.8)$‰ if compared

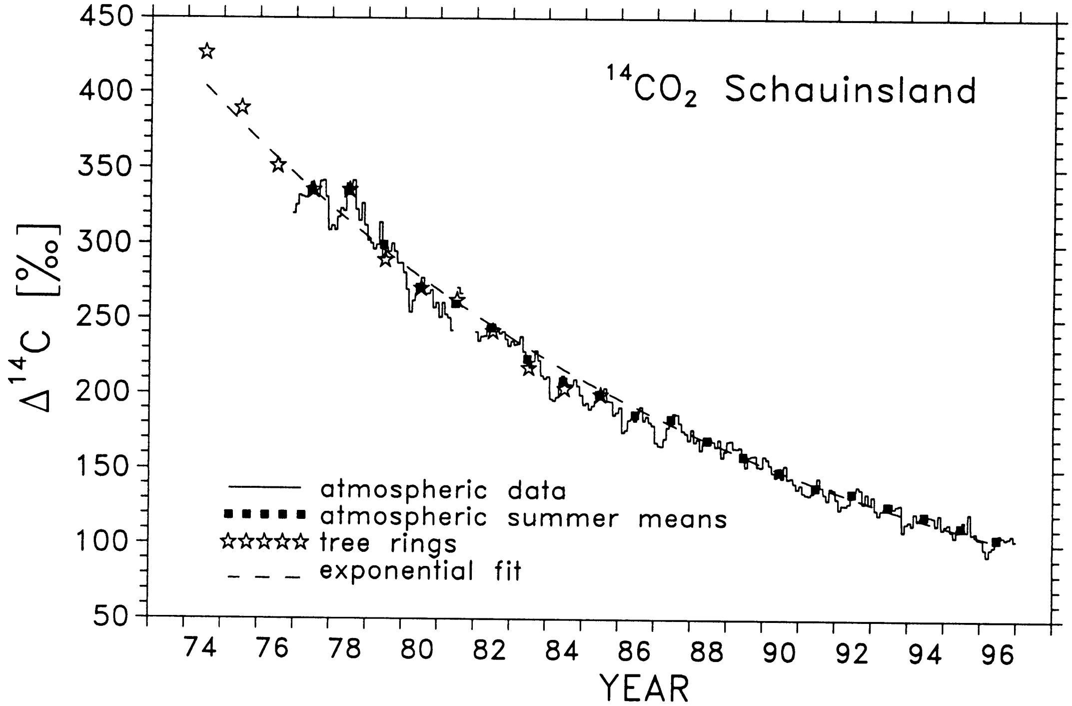

Fig. 2. Monthly mean $\Delta^{14}C$ in atmospheric CO_2 at Schauinsland (histogram) compared to values from individual rings of a tree grown close to the Schauinsland site. For overlapping years, the tree ring and summer mean (May–August) atmospheric values agree within ±4‰. -- = an exponential fit through the combined summer mean and tree ring values described by the function $\Delta^{14}C$ (t) = 417 • exp(-t/16.0); t = years after 1974.

to Jungfraujoch, and by $\Delta^{14}C$ = (4.3 ± 0.6)‰ if compared to Izaña. The difference between the two continental sites and Izaña is partly caused by the general continental pileup of fossil fuel CO_2 in Central Europe. However, due to fast atmospheric mixing in the west wind belt, mid northern latitudes (contributing >80% of global CO_2 emissions from fossil fuels (Rotty 1983)) may be generally influenced by fossil fuel CO_2 even over the Atlantic ocean.

As described previously (Levin, Graul and Trivett 1995), the regular seasonal variations after 1982, when all atmospheric tests stopped, have been attributed to seasonally varying contributions of fossil fuel CO_2 at the Schauinsland site. After extension of our observational $^{14}CO_2$ network to maritime clean-air stations, however, significant seasonal variations were observed at all northern hemispheric sites with *ca.* 5–8‰ higher $\Delta^{14}C$ values in late summer compared to early spring (Levin *et al.* 1992 and unpublished Heidelberg data). We are therefore confident that only about half of the seasonal amplitude observed at Schauinsland is caused by regional fossil fuel CO_2 contamination. The remaining part can be traced back to stratosphere-troposphere exchange ($\Delta^{14}C$ = 1–2‰), as well as to atmosphere-biosphere exchange through isotopic fractionation and disequilibrium effects (Hesshaimer 1997).

$\delta^{13}CO_2$ at Schauinsland Station

As a by-product of the $^{14}CO_2$ analyses of our large-volume CO_2 samples, the stable isotope ratio $^{13}C/^{12}C$ in CO_2 was obtained at Schauinsland during the period of 1977 to 1996 (Fig. 3). A large seasonal cycle with a mean amplitude of $\delta^{13}C$ = ±0.4‰ is observed, closely anti-correlated with atmo-

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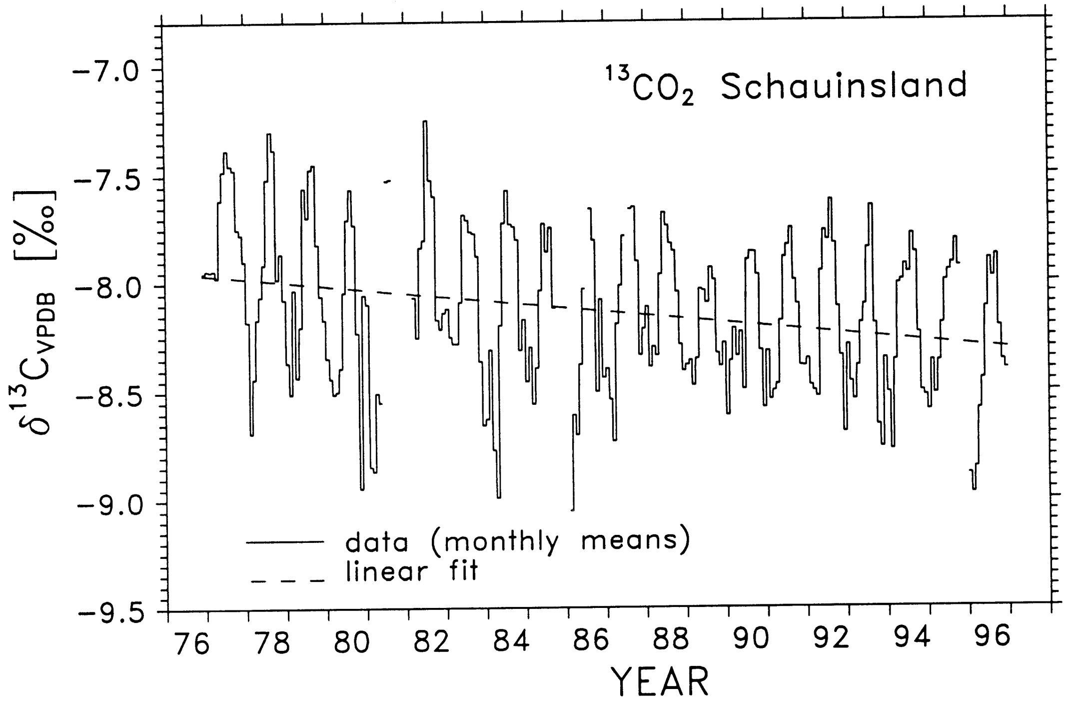

Fig. 3. Monthly mean values of $\delta^{13}C$ in atmospheric CO_2 at the Schauinsland site; -- a linear fit through the monthly data showing a mean decreasing trend of 0.017‰ yr^{-1}.

spheric CO_2 concentration (Levin, Graul and Trivett 1995). The mean $\delta^{13}C$ decreases from 1977 to 1996 by 0.017‰ yr^{-1}, comparable to trends observed at maritime background stations (Keeling *et al.* 1995). One may question the reliability of atmospheric $^{13}CO_2$ data derived from these samples as they may be partly fractionated during purification over charcoal. Therefore, we compared $\delta^{13}C$ results obtained by the chemical absorption method used here with those from samples specifically collected in glass flasks for stable isotopic analysis. CO_2 from whole air samples (*ca.* 100 ml of air) was trapped cryogenically (Finnigan, Bremen, MT-Box) and measured online with our MAT 252 mass spectrometer. These samples showed a systematic shift of +0.2‰ (after correction for N_2O) compared to the data presented here, which may partly be due to the different sampling and analysis techniques, and also to smaller regional source CO_2 contamination of the flask samples that were selectively collected during high wind speed situations. This contamination may arise from anthropogenic as well as from local biospheric CO_2 emissions, both depleted in $\delta^{13}C$. From the comparison of $\Delta^{14}C$ and $\delta^{13}C$ records, it is worth mentioning that both the seasonal amplitude of $^{14}CO_2$ and the seasonal amplitude of $^{13}CO_2$ vary from year to year. Particularly small seasonal $\delta^{13}C$ variations are observed in the years 1988 to 1990 when we also found only very small wintertime ^{14}C depletions (see Fig. 2). These years are characterized by relatively less severe winters associated with frequent maritime air mass flow in Western and Central Europe.

Conclusion

Atmospheric $^{14}CO_2$ in continental Europe shows an exponential decline with an e-folding time of *ca.* 16 yr, very similar to what is observed globally. During the growing season, May to August, the influence from continental fossil fuel CO_2 sources at Schauinsland, *ca.* 1000 m asl, causes a $\Delta^{14}C$

depletion on the order of ≤5‰. During winter, this depletion is about twice as large. Schauinsland observations can therefore serve well as a reference for regional atmospheric $^{14}CO_2$ studies and also for dating of modern organic material or groundwater in Central Europe. If corrected for the small anthropogenic effect, our Schauinsland record can be used as an input function for global carbon cycle modeling studies.

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Appendix: Data from Schauinsland Station and Tree-Ring Samples

Table 1. Δ^{14}C in atmospheric CO_2 at Schauinsland, Germany (48°N, 8°E, 1205 m asl). * = δ^{13}C values fractionated during sampling.

Lab code	Sample no.	Sampling period (dd/mm/yy–dd/mm/yy)	Δ^{14}C (‰)	δ^{13}C (‰)
Hd-8662	Sch-156	11/06/84 – 26/06/84	216 ± 3	−7.76
Hd-5789	Sch-157	25/06/84 – 09/07/84	208 ± 4	−7.53
Hd-8774	Sch-158	09/07/84 – 23/07/84	206 ± 2	−7.55
Hd-8776	Sch-159	23/07/84 – 06/08/84	211 ± 4	−7.65
Hd-8790	Sch-160	06/08/84 – 20/08/84	203 ± 4	−7.76
Hd-8909	Sch-161	20/08/84 – 03/09/84	209 ± 3	−6.52*
Hd-8857	Sch-162	07/09/84 – 24/09/84	209 ± 4	−7.80
Hd-8936	Sch-163	24/09/84 – 04/10/84	198 ± 13	−7.59
Hd-8937	Sch-164	04/10/84 – 15/10/84	206 ± 4	−7.79
Hd-8946	Sch-165	15/10/84 – 30/10/84	216 ± 4	−7.86
Hd-9008	Sch-166	30/10/84 – 13/11/84	205 ± 2	−8.10*
Hd-9014	Sch-167	13/11/84 – 26/11/84	207 ± 2	−8.31
Hd-9015	Sch-168	26/11/84 – 10/12/84	203 ± 2	−8.33*
Hd-9111	Sch-169	10/12/84 – 21/12/84	206 ± 4	−7.92
Hd-9112	Sch-170	21/12/84 – 14/01/85	190 ± 4	−8.43
Hd-9122	Sch-171	14/01/85 – 28/01/85	194 ± 4	−8.52
Hd-9162	Sch-172	28/01/85 – 11/02/85	199 ± 4	−8.27
Hd-9364	Sch-173	11/02/85 – 25/02/85	192 ± 3	−8.29
Hd-9365	Sch-174	25/02/85 – 06/03/85	190 ± 5	−8.40
Hd-9366	Sch-175	06/03/85 – 18/03/85	184 ± 4	−8.70
Hd-9367	Sch-176	18/03/85 – 02/04/85	195 ± 4	−8.49
Hd-9368	Sch-177	02/04/85 – 15/04/85	198 ± 4	−8.22
Hd-9387	Sch-178	15/04/85 – 29/04/85	186 ± 4	−8.55
Hd-9528	Sch-179	29/04/85 – 13/05/85	194 ± 4	−8.32
Hd-9529	Sch-180	13/05/85 – 28/05/85	196 ± 4	−7.88
Hd-9530	Sch-181	28/05/85 – 10/06/85	200 ± 4	−7.72
Hd-9531	Sch-182	10/06/85 – 24/06/85	203 ± 4	−7.58
Hd-9648	Sch-183	24/06/85 – 08/07/85	205 ± 4	−8.05
Hd-9649	Sch-184	08/07/85 – 22/07/85	191 ± 4	−7.86
Hd-9650	Sch-185	22/07/85 – 05/08/85	199 ± 4	−7.71
Hd-9651	Sch-186	05/08/85 – 19/08/85	208 ± 4	−7.62
Hd-9829	Sch-187	19/08/85 – 02/09/85	202 ± 4	−7.90
Hd-9828	Sch-188	02/09/85 – 16/09/85	193 ± 4	−8.14
Hd-9830	Sch-189	16/09/85 – 30/09/85	197 ± 4	−8.33*
Hd-9858	Sch-190	30/09/85 – 14/10/85	204 ± 4	−8.45*
Hd-9974	Sch-191	14/10/85 – 28/10/85	187 ± 4	−8.70*
Hd-9975	Sch-192	28/10/85 – 18/11/85	195 ± 4	−8.14*

TABLE 1. (Continued)

Lab code	Sample no.	Sampling period (dd/mm/yy–dd/mm/yy)	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
Hd-9976	Sch-193	18/11/85 – 02/12/85	174 ± 4	−9.48*
Hd-9977	Sch-194	02/12/85 – 16/12/85	188 ± 4	−9.04*
Hd-9997	Sch-195	16/12/85 – 13/01/86	189 ± 4	−5.97*
Hd-10123	Sch-196	13/01/86 – 27/01/86	192 ± 4	−9.66*
Hd-10124	Sch-197	27/01/86 – 03/02/86	195 ± 4	−10.32*
Hd-10125	Sch-198	03/02/86 – 17/02/86	163 ± 4	−9.58*
Hd-10126	Sch-199	17/02/86 – 03/03/86	184 ± 4	−9.05
Hd-10181	Sch-200	03/03/86 – 17/03/86	161 ± 4	−8.96
Hd-10182	Sch-201	17/03/86 – 01/04/86	189 ± 4	−8.20
Hd-10183	Sch-202	01/04/86 – 14/04/86	174 ± 4	−9.02
Hd-10222	Sch-203	14/04/86 – 28/04/86	191 ± 4	−8.50
Hd-10398	Sch-204	28/04/86 – 12/05/86	182 ± 4	−8.26
Hd-10399	Sch-205	12/05/86 – 26/05/86	186 ± 5	−8.47
Hd-10400	Sch-206	26/05/86 – 09/06/86	182 ± 2	−8.89*
Hd-10401	Sch-207	09/06/86 – 23/06/86	180 ± 5	−8.03
Hd-10406	Sch-208	23/06/86 – 07/07/86	186 ± 5	−8.21*
Hd-10490	Sch-209	07/07/86 – 21/07/86	186 ± 2	−7.83*
Hd-10491	Sch-210	21/07/86 – 04/08/86	192 ± 2	−7.75*
Hd-10494	Sch-211	04/08/86 – 18/08/86	193 ± 4	−7.66
Hd-10562	Sch-212	18/08/86 – 02/09/86	190 ± 2	−8.72*
Hd-10563	Sch-213	02/09/86 – 15/09/86	184 ± 4	−7.77
Hd-10564	Sch-214	15/09/86 – 29/09/86	180 ± 4	−7.84
Hd-10583	Sch-215	29/09/86 – 13/10/86	182 ± 3	−8.62*
Hd-10661	Sch-216	13/10/86 – 27/10/86	188 ± 3	−8.50
Hd-10662	Sch-217	27/10/86 – 10/11/86	185 ± 2	−8.61*
Hd-10673	Sch-218	10/11/86 – 24/11/86	180 ± 3	−8.64*
Hd-10710	Sch-219	24/11/86 – 08/12/86	179 ± 3	−8.08
Hd-10711	Sch-220	08/12/86 – 22/12/86	178 ± 2	−8.64*
Hd-10724	Sch-221	22/12/86 – 02/01/87	181 ± 3	−8.71
Hd-10801	Sch-222	02/01/87 – 19/01/87	161 ± 2	−8.98*
Hd-10802	Sch-223	19/01/87 – 02/02/87	176 ± 3	−8.36
Hd-10813	Sch-224	02/02/87 – 16/02/87	173 ± 3	−8.27
Hd-10868	Sch-225	16/02/87 – 02/03/87	157 ± 2	−8.85
Hd-10869	Sch-226	02/03/87 – 16/03/87	167 ± 3	−9.31
Hd-10875	Sch-227	16/03/87 – 30/03/87	164 ± 4	−8.18
Hd-10876	Sch-228	30/03/87 – 13/04/87	172 ± 4	−8.38
Hd-11028	Sch-229	13/04/87 – 27/04/87	169 ± 3	−8.03
Hd-11029	Sch-230	27/04/87 – 11/05/87	172 ± 2	−8.18
Hd-11032	Sch-231	11/05/87 – 25/05/87	180 ± 2	−7.98
Hd-11033	Sch-232	25/05/87 – 09/06/87	182 ± 3	−7.83
Hd-11037	Sch-233	09/06/87 – 22/06/87	180 ± 3	−7.76
Hd-11253	Sch-234	22/06/87 – 13/07/87	186 ± 4	−7.92*
Hd-11254	Sch-235	13/07/87 – 03/08/87	188 ± 4	−7.90*
Hd-11255	Sch-236	03/08/87 – 17/08/87	189 ± 4	−7.59
Hd-11256	Sch-237	17/08/87 – 03/09/87	184 ± 4	−7.73
Hd-11258	Sch-238	03/09/87 – 14/09/87	183 ± 4	−7.56
Hd-11259	Sch-239	14/09/87 – 28/09/87	180 ± 4	−7.68
Hd-11369	Sch-240	28/09/87 – 12/10/87	174 ± 5	−7.81

TABLE 1. (Continued)

Lab code	Sample no.	Sampling period (dd/mm/yy–dd/mm/yy)	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
Hd-11370	Sch-241	12/10/87 – 26/10/87	178 ± 4	−7.84
Hd-11371	Sch-242	26/10/87 – 09/11/87	173 ± 4	−8.25
Hd-11372	Sch-243	09/11/87 – 23/11/87	180 ± 4	−8.26
Hd-11438	Sch-244	23/11/87 – 07/12/87	164 ± 4	−8.56
Hd-11439	Sch-245	07/12/87 – 21/12/87	168 ± 4	−8.21
Hd-11442	Sch-246	21/12/87 – 04/01/88	175 ± 3	−8.01
Hd-11539	Sch-248	18/01/88 – 01/02/88	177 ± 4	−8.14
Hd-11540	Sch-249	01/02/88 – 15/02/88	168 ± 4	−8.29
Hd-11549	Sch-250	15/02/88 – 29/02/88	168 ± 4	−8.49
Hd-11620	Sch-252	14/03/88 – 28/03/88	174 ± 4	−8.25
Hd-11621	Sch-253	28/03/88 – 11/04/88	156 ± 3	−8.47
Hd-11630	Sch-254	11/04/88 – 25/04/88	167 ± 3	−8.32
Hd-11701	Sch-255	25/04/88 – 09/05/88	165 ± 4	−8.13
Hd-11702	Sch-256	09/05/88 – 24/05/88	168 ± 5	−7.91
Hd-11703	Sch-257	24/05/88 – 06/06/88	174 ± 4	−7.89
Hd-11767	Sch-258	10/06/88 – 20/06/88	174 ± 4	−7.51
Hd-11867	Sch-259	20/06/88 – 04/07/88	163 ± 4	−7.73
Hd-11868	Sch-260	04/07/88 – 18/07/88	172 ± 4	−7.75
Hd-11879	Sch-261	18/07/88 – 01/08/88	171 ± 4	−7.73
Hd-11880	Sch-262	01/08/88 – 15/08/88	167 ± 4	−7.77
Hd-12010	Sch-263	15/08/88 – 29/08/88	171 ± 3	−7.88
Hd-12013	Sch-264	29/08/88 – 12/09/88	165 ± 5	−7.81
Hd-12020	Sch-265	12/09/88 – 26/09/88	163 ± 5	−7.85
Hd-12021	Sch-266	26/09/88 – 10/10/88	171 ± 4	−8.00
Hd-12101	Sch-267	10/10/88 – 24/10/88	173 ± 4	−7.94
Hd-12102	Sch-268	24/10/88 – 07/11/88	163 ± 4	−8.29
Hd-12162	Sch-269	07/11/88 – 21/11/88	158 ± 4	−8.26
Hd-12128	Sch-270	21/11/88 – 05/12/88	155 ± 3	−8.32
Hd-12131	Sch-271	05/12/88 – 19/12/88	160 ± 5	−8.52
Hd-12132	Sch-272	19/12/88 – 02/01/89	167 ± 5	−8.31
Hd-12277	Sch-273	02/01/89 – 16/01/89	167 ± 5	−8.50
Hd-12278	Sch-274	16/01/89 – 30/01/89	171 ± 4	−8.27
Hd-12283	Sch-275	30/01/89 – 13/02/89	165 ± 4	−8.34
Hd-12284	Sch-276	13/02/89 – 27/02/89	173 ± 4	−8.33
Hd-12525	Sch-277	27/02/89 – 13/03/89	170 ± 5	−8.75
Hd-12638	Sch-278	13/03/89 – 28/03/89	159 ± 3	−8.29
Hd-12648	Sch-279	28/03/89 – 10/04/89	165 ± 4	−8.28
Hd-12654	Sch-280	10/04/89 – 24/04/89	164 ± 4	−8.39
Hd-12661	Sch-281	24/04/89 – 08/05/89	162 ± 5	−8.37
Hd-12978	Sch-282	08/05/89 – 22/05/89	168 ± 5	−7.89
Hd-12979	Sch-283	22/05/89 – 05/06/89	162 ± 3	−7.98
Hd-13019	Sch-284	05/06/89 – 19/06/89	163 ± 3	−8.09
Hd-13027	Sch-285	19/06/89 – 03/07/89	155 ± 5	−8.00
Hd-12844	Sch-286	03/07/89 – 17/07/89	154 ± 3	−8.18
Hd-12845	Sch-287	17/07/89 – 31/07/89	149 ± 4	−8.03
Hd-12910	Sch-288	31/07/89 – 14/08/89	161 ± 3	−8.01
Hd-12911	Sch-289	14/08/89 – 28/08/89	157 ± 5	−7.90
Hd-12913	Sch-290	28/08/89 – 11/09/89	154 ± 4	−7.80

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TABLE 1. (Continued)

Lab code	Sample no.	Sampling period (dd/mm/yy–dd/mm/yy)	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
Hd-12914	Sch-291	12/09/89 – 25/09/89	162 ± 4	–7.99
Hd-12929	Sch-292	25/09/89 – 09/10/89	162 ± 3	–8.35
Hd-12930	Sch-293	09/10/89 – 23/10/89	159 ± 3	–8.23
Hd-13040	Sch-294	23/10/89 – 06/11/89	157 ± 3	–8.46
Hd-12993	Sch-295	06/11/89 – 20/11/89	152 ± 3	–8.39
Hd-12994	Sch-296	20/11/89 – 04/12/89	150 ± 3	–8.34
Hd-13076	Sch-297	04/12/89 – 18/12/89	149 ± 3	–8.36
Hd-13077	Sch-298	18/12/89 – 02/01/90	155 ± 3	–8.18
Hd-13085	Sch-299	02/01/90 – 15/01/90	159 ± 3	–8.93
Hd-13182	Sch-300	15/01/90 – 29/01/90	161 ± 5	–8.39
Hd-13183	Sch-301	29/01/90 – 12/02/90	150 ± 4	–8.44
Hd-13213	Sch-302	12/02/90 – 26/02/90	162 ± 2	–8.35
Hd-13214	Sch-303	26/02/90 – 12/03/90	162 ± 2	–8.08
Hd-13194	Sch-304	12/03/90 – 26/03/90	150 ± 4	–8.09
Hd-13215	Sch-305	26/03/90 – 09/04/90	144 ± 3	–8.79
Hd-13256	Sch-306	09/04/90 – 23/04/90	151 ± 4	–8.20
Hd-13257	Sch-307	23/04/90 – 07/05/90	148 ± 4	–8.10
Hd-13345	Sch-308	07/05/90 – 21/05/90	142 ± 4	–8.09
Hd-13264	Sch-309	21/05/90 – 05/06/90	148 ± 4	–8.49
Hd-13265	Sch-310	05/06/90 – 25/06/90	154 ± 3	–8.15*
Hd-13392	Sch-311	25/06/90 – 16/07/90	146 ± 3	–8.50*
Hd-13418	Sch-312	16/07/90 – 30/07/90	143 ± 5	–7.89
Hd-13444	Sch-313	30/07/90 – 06/08/90	145 ± 5	–7.97
Hd-13407	Sch-314	13/08/90 – 28/08/90	157 ± 4	–7.80
Hd-13408	Sch-315	28/08/90 – 10/09/90	148 ± 3	–7.96
Hd-13445	Sch-316	10/09/90 – 24/09/90	139 ± 6	–7.81
Hd-13612	Sch-317	24/09/90 – 08/10/90	149 ± 5	–7.84
Hd-13622	Sch-318	08/10/90 – 22/10/90	142 ± 6	–7.87
Hd-13634	Sch-319	22/10/90 – 05/11/90	145 ± 5	–8.21
Hd-13635	Sch-320	05/11/90 – 19/11/90	146 ± 4	–8.20
Hd-13647	Sch-321	19/11/90 – 03/12/90	134 ± 5	–8.49
Hd-13582	Sch-322	03/12/90 – 17/12/90	138 ± 5	–8.62
Hd-13709	Sch-323	17/12/90 – 31/12/90	137 ± 5	–8.58
Hd-13680	Sch-324	31/12/90 – 14/01/91	142 ± 5	–8.08
Hd-13685	Sch-325	14/01/91 – 28/01/91	139 ± 3	–8.48
Hd-13731	Sch-326	28/01/91 – 11/02/91	138 ± 3	–8.57
Hd-13808	Sch-327	11/02/91 – 25/02/91	134 ± 4	–8.56
Hd-13809	Sch-328	25/02/91 – 11/03/91	130 ± 5	–8.36
Hd-13845	Sch-329	11/03/91 – 25/03/91	137 ± 5	–8.49
Hd-13846	Sch-330	25/03/91 – 08/04/91	132 ± 5	–8.73
Hd-13872	Sch-331	08/04/91 – 22/04/91	135 ± 6	–8.42
Hd-13987	Sch-332	22/04/91 – 06/05/91	131 ± 4	–8.32
Hd-13988	Sch-333	06/05/91 – 21/05/91	132 ± 5	–8.25
Hd-13999	Sch-334	21/05/91 – 03/06/91	140 ± 5	–8.01
Hd-14005	Sch-335	03/06/91 – 17/06/91	130 ± 3	–8.00
Hd-14012	Sch-336	17/06/91 – 01/07/91	141 ± 5	–7.75
Hd-14070	Sch-337	01/07/91 – 17/07/91	146 ± 5	–7.81
Hd-14115	Sch-338	17/07/91 – 29/07/91	145 ± 4	–7.88

TABLE 1. (Continued)

Lab code	Sample no.	Sampling period (dd/mm/yy–dd/mm/yy)	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
Hd-14102	Sch-339	29/07/91 – 12/08/91	134 ± 3	–7.76
Hd-14183	Sch-340	12/08/91 – 26/08/91	142 ± 4	–7.76
Hd-14217	Sch-341	26/08/91 – 09/09/91	140 ± 6	–7.72
Hd-14260	Sch-342	09/09/91 – 23/09/91	128 ± 4	–8.06
Hd-14227	Sch-343	23/09/91 – 07/10/91	138 ± 3	–8.17
Hd-14202	Sch-344	07/10/91 – 21/10/91	124 ± 6	–8.15
Hd-14239	Sch-345	21/10/91 – 04/11/91	133 ± 6	–8.00
Hd-14378	Sch-346	04/11/91 – 18/11/91	144 ± 4	–8.44
Hd-14381	Sch-347	18/11/91 – 02/12/91	133 ± 5	–8.42
Hd-14388	Sch-348	02/12/91 – 16/12/91	141 ± 4	–8.33
Hd-14441	Sch-349	16/12/91 – 30/12/91	134 ± 3	–8.46
Hd-14504	Sch-350	30/12/91 – 13/01/92	134 ± 4	–8.17
Hd-14505	Sch-351	13/01/92 – 27/01/92	130 ± 4	–8.45
Hd-14467	Sch-352	27/01/92 – 10/02/92	125 ± 5	–8.54
Hd-14728	Sch-353	10/02/92 – 24/02/92	123 ± 4	–8.45
Hd-14729	Sch-354	24/02/92 – 09/03/92	124 ± 4	–8.42
Hd-14578	Sch-355	09/03/92 – 23/03/92	127 ± 5	–8.40
Hd-14607	Sch-356	23/03/92 – 06/04/92	129 ± 4	–8.73
Hd-14858	Sch-357	06/04/92 – 21/04/92	131 ± 4	–8.48
Hd-14859	Sch-358	21/04/92 – 04/05/92	118 ± 4	–8.48
Hd-14869	Sch-359	04/05/92 – 18/05/92	124 ± 4	–7.98
Hd-14870	Sch-360	18/05/92 – 01/06/92	134 ± 5	–7.79
Hd-14875	Sch-361	01/06/92 – 15/06/92	136 ± 5	–7.75
Hd-14876	Sch-362	15/06/92 – 29/06/92	133 ± 4	–7.77
Hd-15100	Sch-363	29/06/92 – 13/07/92	132 ± 5	–7.83
Hd-15056	Sch-364	13/07/92 – 27/07/92	141 ± 4	–7.81
Hd-15150	Sch-365	27/07/92 – 10/08/92	134 ± 5	–7.74
Hd-15202	Sch-366	10/08/92 – 24/08/92	141 ± 3	–7.56
Hd-15153	Sch-367	24/08/92 – 07/09/92	142 ± 3	–7.59
Hd-15170	Sch-369	21/09/92 – 05/10/92	134 ± 5	–7.98
Hd-15204	Sch-370	05/10/92 – 19/10/92	123 ± 4	–8.29
Hd-15232	Sch-371	19/10/92 – 02/11/92	131 ± 3	–7.97
Hd-15155	Sch-372	02/11/92 – 16/11/92	135 ± 3	–8.38
Hd-15176	Sch-373	16/11/92 – 30/11/92	134 ± 5	–8.32
Hd-15211	Sch-374	30/11/92 – 14/12/92	134 ± 4	–8.65
Hd-15212	Sch-375	14/12/92 – 28/12/92	122 ± 3	–8.79
Hd-15229	Sch-376	28/12/92 – 11/01/93	125 ± 3	–8.46
Hd-15238	Sch-377	11/01/93 – 25/01/93	142 ± 6	–8.17
Hd-15245	Sch-378	25/01/93 – 08/02/93	124 ± 3	–8.27
Hd-15588	Sch-379	08/02/93 – 22/02/93	127 ± 4	–8.44
Hd-15589	Sch-380	22/02/93 – 08/03/93	116 ± 3	–8.70
Hd-15622	Sch-381	08/03/93 – 22/03/93	120 ± 5	–8.48
Hd-15635	Sch-382	22/03/93 – 05/04/93	124 ± 4	–8.50
Hd-15641	Sch-383	05/04/93 – 19/04/93	122 ± 5	–8.41
Hd-15648	Sch-384	19/04/93 – 03/05/93	125 ± 4	–8.31
Hd-15991	Sch-385	03/05/93 – 17/05/93	115 ± 3	–8.17
Hd-15992	Sch-386	17/05/93 – 01/06/93	134 ± 5	–8.19
Hd-16123	Sch-387	01/06/93 – 14/06/93	116 ± 4	–8.22

TABLE 1. (Continued)

Lab code	Sample no.	Sampling period (dd/mm/yy–dd/mm/yy)	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
Hd-16124	Sch-388	14/06/93 – 28/06/93	134 ± 5	–8.03
Hd-16131	Sch-389	28/06/93 – 12/07/93	125 ± 3	–7.93
Hd-16132	Sch-390	12/07/93 – 26/07/93	131 ± 3	–7.83
Hd-16158	Sch-391	26/07/93 – 09/08/93	130 ± 5	–7.72
Hd-16157	Sch-392	09/08/93 – 23/08/93	125 ± 6	–7.48
Hd-16165	Sch-393	23/08/93 – 06/09/93	119 ± 4	–7.86
Hd-16168	Sch-394	06/09/93 – 20/09/93	130 ± 4	–8.40
Hd-16136	Sch-395	20/09/93 – 04/10/93	125 ± 3	–8.09
Hd-16150	Sch-396	04/10/93 – 18/10/93	133 ± 4	–8.54
Hd-16169	Sch-397	18/10/93 – 02/11/93	121 ± 4	–8.92
Hd-16143	Sch-398	02/11/93 – 15/11/93	107 ± 4	–8.62
Hd-16151	Sch-399	15/11/93 – 29/11/93	109 ± 5	–8.93
Hd-16144	Sch-400	29/11/93 – 15/12/93	119 ± 4	–8.31
Hd-16381	Sch-401	15/12/93 – 27/12/93	109 ± 3	–8.38
Hd-16396	Sch-402	27/12/93 – 10/01/94	114 ± 4	–8.40
Hd-16397	Sch-403	10/01/94 – 24/01/94	117 ± 4	–8.44
Hd-16404	Sch-404	24/01/94 – 07/02/94	116 ± 4	–8.76
Hd-16685	Sch-405	07/02/94 – 21/02/94	109 ± 4	–8.97
Hd-16760	Sch-406	21/02/94 – 07/03/94	119 ± 5	–8.42
Hd-16676	Sch-407	07/03/94 – 21/03/94	125 ± 4	–8.34
Hd-16765	Sch-408	21/03/94 – 06/04/94	118 ± 4	–8.35
Hd-16677	Sch-409	06/04/94 – 18/04/94	112 ± 5	–8.73
Hd-16678	Sch-410	18/04/94 – 02/05/94	120 ± 4	–7.18
Hd-16601	Sch-411	02/05/94 – 16/05/94	120 ± 5	–8.08
Hd-16766	Sch-412	16/05/94 – 30/05/94	117 ± 5	–8.00
Hd-16684	Sch-413	30/05/94 – 13/06/94	120 ± 4	–7.87
Hd-16672	Sch-414	13/06/94 – 27/06/94	120 ± 4	–7.93
Hd-16736	Sch-415	27/06/94 – 11/07/94	123 ± 5	–8.06
Hd-16840	Sch-416	11/07/94 – 25/07/94	117 ± 3	–7.91
Hd-16775	Sch-417	25/07/94 – 08/08/94	120 ± 5	–7.90
Hd-16776	Sch-418	08/08/94 – 22/08/94	118 ± 5	–7.81
Hd-16785	Sch-419	22/08/94 – 05/09/94	121 ± 5	–7.65
Hd-16970	Sch-421	19/09/94 – 04/10/94	118 ± 5	–7.95
Hd-16969	Sch-422	04/10/94 – 17/10/94	112 ± 4	–8.34
Hd-16985	Sch-423	17/10/94 – 31/10/94	113 ± 5	–8.19
Hd-17006	Sch-424	31/10/94 – 14/11/94	122 ± 4	–8.62
Hd-16984	Sch-425	14/11/94 – 28/11/94	120 ± 4	–8.41
Hd-17094	Sch-426	28/11/94 – 12/12/94	116 ± 3	–8.37
Hd-17106	Sch-427	12/12/94 – 27/12/94	108 ± 4	–8.49
Hd-17107	Sch-428	27/12/94 – 09/01/95	108 ± 4	–8.99
Hd-17118	Sch-429	09/01/95 – 23/01/95	116 ± 5	–8.50
Hd-17119	Sch-430	23/01/95 – 06/02/95	114 ± 5	–8.34
Hd-17087	Sch-431	06/02/95 – 20/02/95	114 ± 3	–8.30
Hd-17175	Sch-432	20/02/95 – 06/03/95	115 ± 5	–8.48
Hd-17176	Sch-433	06/06/95 – 20/03/95	116 ± 4	–8.43
Hd-17336	Sch-434	20/03/95 – 03/04/95	107 ± 5	–8.53
Hd-17356	Sch-435	03/04/95 – 18/04/95	114 ± 4	–8.30
Hd-17360	Sch-436	18/04/95 – 02/05/95	95 ± 4	–8.41

TABLE 1. (Continued)

Lab code	Sample no.	Sampling period (dd/mm/yy–dd/mm/yy)	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
Hd-17370	Sch-437	02/05/95 – 15/05/95	114 ± 5	–8.21
Hd-17562	Sch-438	15/05/95 – 29/05/95	112 ± 4	–8.18
Hd-17553	Sch-439	29/05/95 – 12/06/95	106 ± 5	–8.06
Hd-17547	Sch-440	12/06/95 – 26/06/95	112 ± 4	–7.99
Hd-17652	Sch-441	26/06/95 – 10/07/95	118 ± 4	–7.86
Hd-17842	Sch-442	10/07/95 – 24/07/95	116 ± 4	–7.99
Hd-17843	Sch-443	24/07/95 – 07/08/95	114 ± 5	–7.99
Hd-17850	Sch-444	07/08/95 – 21/08/95	103 ± 5	–7.88
Hd-17831	Sch-445	21/08/95 – 04/09/95	115 ± 5	–7.92
Hd-17851	Sch-446	04/09/95 – 18/09/95	125 ± 5	–7.77
Hd-17818	Sch-447	18/09/95 – 02/10/95	119 ± 4	–7.82
Hd-17976	Sch-448	02/10/95 – 16/10/95	107 ± 4	–7.51
Hd-17947	Sch-449	16/10/95 – 30/10/95	107 ± 5	–8.36
Hd-18158	Sch-450	30/10/95 – 20/11/95	110 ± 5	–7.81*
Hd-18175	Sch-451	20/11/95 – 11/12/95	106 ± 4	–9.40*
Hd-18192	Sch-452	11/12/95 – 27/12/95	103 ± 4	–8.69*
Hd-18118	Sch-453	27/12/95 – 22/01/96	108 ± 4	–9.53*
Hd-18170	Sch-454	22/01/96 – 05/02/96	97 ± 5	–8.89
Hd-18105	Sch-455	05/02/96 – 19/02/96	97 ± 5	–9.07
Hd-18333	Sch-456	19/02/96 – 04/03/96	99 ± 4	–8.88
Hd-18346	Sch-457	04/03/96 – 18/03/96	88 ± 4	–9.09
Hd-18326	Sch-458	18/03/96 – 01/04/96	97 ± 3	–8.61
Hd-18327	Sch-459	01/04/96 – 15/04/96	94 ± 4	–8.82
Hd-18329	Sch-460	15/04/96 – 29/04/96	102 ± 4	–8.35
Hd-18321	Sch-461	29/04/96 – 13/05/96	95 ± 4	–8.55
Hd-18507	Sch-462	13/05/96 – 28/05/96	103 ± 3	–8.45
Hd-18506	Sch-463	28/05/96 – 10/06/96	100 ± 3	–8.07
Hd-18485	Sch-464	10/06/96 – 25/06/96	106 ± 3	–8.14
Hd-18486	Sch-465	25/06/96 – 08/07/96	109 ± 3	–8.15
Hd-18516	Sch-466	08/07/96 – 22/07/96	107 ± 3	–7.76
Hd-18669	Sch-467	31/07/96 – 19/08/96	100 ± 3	–8.03
Hd-18655	Sch-468	19/08/96 – 02/09/96	114 ± 4	–7.91
Hd-18685	Sch-469	02/09/96 – 16/09/96	105 ± 3	–7.80
Hd-18675	Sch-470	16/09/96 – 30/09/96	103 ± 3	–7.94
Hd-18692	Sch-471	30/09/96 – 14/10/96	103 ± 3	–8.32
Hd-18769	Sch-472	14/10/96 – 28/10/96	106 ± 3	–8.10
Hd-18779	Sch-473	28/10/96 – 11/11/96	109 ± 3	–8.20
Hd-18764	Sch-474	11/11/96 – 25/11/96	108 ± 3	–8.51
Hd-18809	Sch-475	25/11/96 – 09/12/96	101 ± 6	–8.29
Hd-19003	Sch-476	09/12/96 – 23/12/96	105 ± 3	–8.45
Hd-18994	Sch-477	23/12/96 – 13/01/96	99 ± 3	–8.76

TABLE 2. $\Delta^{14}C$ in individual tree rings (*Picea abies*) grown in the near vicinity of Schauinsland. Summer means (May–August) from atmospheric samples are also reported for comparison.

	Tree rings			Atmosphere
Year	Lab code	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)	$\Delta^{14}C$ (‰)
1974	Hd-10088	427 ± 3	–25.75	
1975	Hd-10087	390 ± 3	–26.02	
1976	Hd-10074	352 ± 3	–25.43	
1977	Hd-10075	336 ± 3	–24.42	335.3 ± 3.4
1978	Hd-10076	336 ± 3	–24.37	334.9 ± 7.9
1979	Hd-10077	290 ± 3	–25.76	299.4 ± 8.8
1980	Hd-10052	271 ± 3	–25.71	271.1 ± 4.6
1981	Hd-10051	263 ± 3	–26.40	260.0 ± 12.8
1982	Hd-10049	242 ± 3	–25.76	244.1 ± 1.7
1983	Hd-10045	218 ± 2	–24.52	223.4 ± 3.9
1984	Hd-10040	204 ± 2	–24.99	208.7 ± 2.2
1985	Hd-10039	200 ± 3	–25.65	199.8 ± 3.7
1986				186.4 ± 3.7
1987				183.4 ± 3.8
1988				169.4 ± 0.8
1989				158.7 ± 4.5
1990				148.6 ± 3.7
1991				138.4 ± 3.8
1992				134.4 ± 4.2
1993				125.9 ± 1.6
1994				119.5 ± 0.6
1995				111.9 ± 2.5
1996				104.3 ± 3.0