Wheeler[23] terms the sign change of a spinor under 2π rotation its 'entanglement relation', referring to the model of a macroscopic object tied (by elastic strings, ribbons or a strip of material) to its surroundings. Under 2π-rotations the strings become irremediably entangled, but after 4π-rotation they may be topologically restored to the untangled state without rotating the object. This 'entanglement relation' underlies a variety of popularized demonstrations of spinor properties. Dirac was fond of twisting his belt to show it. A triangular lecture-hall model has been described[21], but perhaps the most beautiful demonstration is that of the Phillipine Binasuan dance[24]. The causal

connections of interaction may be less surprising than those of correlation (as in recent implementations of the Einstein-Rosen-Podolsky 'paradox'[25]), but even if they turn out to be pictured or controlled more easily, they are no less real.

1. Aharonov, Y. & Suskind, L. Phys. Rev. 158, 1237-1238 (1967).
2. Bernstein, H. J. Phys. Rev. Lett. 18, 1102-1103 (1967).
3. Rauch, H. et al. Phys. Lett. 54A, 425-427 (1975).
4. Rauch, H., Wilfing, A., Bauspiess, W. & Bonse, U. Z. Phys. B29, 281-284 (1978).
5. Rauch, H. & Zeilinger, A. Hadr. J. 4, 1280-1294 (1981).
6. Werner, S. A., Colella, R., Overhauser, A. W. & Eagen, C. F. Phys. Rev. Lett. 35, 1053-1056 (1975).
7. Klein, A. G. & Opat, G. I. Phys. Rev. Lett. 37, 238-240 (1976).
8. Klempt, E. Phys. Rev. D13, 3125-3129 (1976).
9. Stoll, M. E., Vega, A. J. & Vaughan, R. W. Phys. Rev. A16, 1521-1524 (1977).
10. Stoll, M. E., Wolff, E. K. & Mehring, M. Phys. Rev. A17, 1561-1567 (1978).
11. Kaiser, R. Can. J. Phys. 56, 1321-1332 (1978).
12. Zeilinger, A. Nature 294, 544-546 (1981).
13. Byrne, J. Nature 275, 189-191 (1978).
14. Bernstein, H. J. & Zeilinger, A. Phys. Lett. 75A, 169-172 (1980).
15. Klein, A. G. & Werner, S. A. Rep. Prog. Phys. 46, 259-335 (1983).
16. Price, R. H. Am. J. Phys. 50, 300-329 (1982).
17. Bernstein, H. J. in Neutron Interferometry (ed. Bonse, U. & Rauch, H.) 231-240 (Oxford University Press, 1979).
18. Moore, G. T. Am. J. Phys. 38, 1177-1180 (1970).
19. Mezei, F. in Neutron Interferometry, 265-272 (Oxford University Press, 1979).
20. Balcar, E. in Neutron Interferometry, 252-264 (Oxford University Press, 1979).
21. Bernstein, H. J. Scientific Research (NY) 4, 32-33 (1969).
22. Eder, G. & Zeilinger, A. Nuovo Cimento B34, 76-90 (1976).
23. Wheeler, J. A. Einsteins Vision, 69-75 and Fig. 70 (Springer, Berlin, 1968).
24. Bernstein, H. J. & Phillips, A. V. Scient. Am. 245(7), 121-137 (1981).
25. Aspect, A. et al. Phys. Rev. Lett. 49, 1804-1807 (1982).

Evidence from polar ice cores for the increase in atmospheric CO₂ in the past two centuries

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Precise and continuous measurements of atmospheric CO_2 concentration were first begun in 1958 and show a clear increase from 315 parts per million by volume (p.p.m.v.)[1] then to 345 p.p.m.v. now. A detailed knowledge of the CO_2 increase since preindustrial time is a prerequisite for understanding several aspects of the role of CO_2, such as the contribution of biomass burning to the CO_2 increase and the sensitivity of climate to the CO_2 concentration in the atmosphere. Estimates of the preindustrial CO_2 concentration are in the range 250–290 p.p.m.v. (ref. 2), but the precise level then and the time dependence of the increase to the present levels remain obscure. The most reliable assessment of the ancient atmospheric CO_2 concentration is derived from measurements of air occluded in ice cores. An ice core from Siple Station (West Antartica) that allows determination of the enclosed gas concentration with very good time resolution has recently become available. We report here measurements of this core which now allow us to trace the development of the atmospheric CO_2 from a period overlapping the Mauna Loa record back over the past two centuries.

Air bubbles are a characteristic feature of natural ice. At locations with mean air temperatures well below the freezing point, ice is formed by sintering of dry firn. At the firn–ice transition, the pore volume becomes separated as isolated bubbles having no further interaction with the atmosphere. The transition process is slow, so that the sampling of air into isolated bubbles spans at least several years, the duration depending mainly on the accumulation rate and the mean annual temperature.

Measurements were made on a 200-m ice core drilled in the Antarctic summer 1983/84 at Siple Station (75°55′ S, 83°55′ W) by the Polar Ice Coring Office in Nebraska and our institute. The mean annual air temperature is −24 °C and the annual accumulation rate is $500 \, kg \, m^{-2}$. Only one clearly identifiable melt layer of irregular thickness (2–10 mm) was observed in the entire core at 7 m below surface (m.b.s.). Counting the seasonal variations of the electrical conductivity of the ice core allowed us to date the core over the past 200 yr with an accuracy of
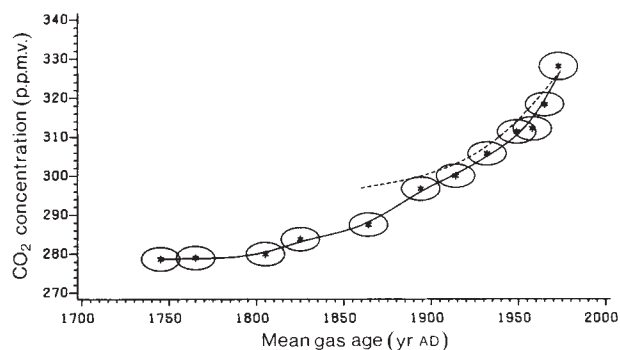

Fig. 1 Measured mean CO_2 concentration plotted against the estimated mean gas age. The horizontal axis of the ellipses indicates the close-off time interval of 22 yr. The uncertainties of the concentration measurements are twice the standard deviation of the mean value, but not lower than the precision of 1% of the measurement. The dotted line represents the model-calculated back extrapolation of the atmospheric CO_2 concentration, assuming only CO_2 input from fossil fuel[2].

Methods. After crushing the ice samples, the gas expands over a cold trap, condensing the water vapour at −80 °C into the absorption cell. Care was taken to minimize the effect of selective transport of CO_2 by water vapour. This effect was discovered while testing the system, when a difference in the measured CO_2 concentration was observed depending on whether the valve between the crusher and the cold trap was open or closed during the crushing process. With the valve closed, the concentrations were systematically higher by ~15 p.p.m.v. Crushing the ice with the valve open causes small ice particles to be transported to warm surfaces, where they immediately evaporate, leading to a considerable amount of water vapour in the system. The part of this water vapour not immediately condensed in the cold trap expands into the absorption cell and then flows slowly back into the cold trap, as indicated by the pressure monitoring in the absorption cell, which leads to a decreased CO_2 concentration in the absorption cell. When the valve is closed, the ice particles created during the crushing process are kept back in the cold crusher and less water vapour reaches the absorption cell after the valve is opened, leading to smaller deviations of the CO_2 concentration. Also, for this slightly modified measuring procedure, the measurements could be controlled by crushing pure gas-free single-crystal ice surrounded by the same amount of standard gas as contained in an average ice sample. With this procedure, small positive deviations in the CO_2 concentration, attributable to water vapour flow from the crusher into the cold trap, can be determined and the measurements corrected accordingly. This correction is below 1% for 10-cm³ samples and below 4% for 1-cm³ samples. All Siple samples were measured using the modified procedure and each fourth measurement was a calibration run.

Table 1 Mean CO_2 concentration in air extracted from ice samples

Siple Station

Depth below surface (m)	Samples measured	Ice from (yr AD)	Air enclosed (yr AD)	CO₂ concentration Extracted air (p.p.m.v.)	Atmosphere (p.p.m.v.)
68.2–68.6	8	1891	1962–1983	328 ± 3.5	328
72.4–72.7	11	1883	1954–1976	318 ± 3.0	321
76.2–76.6	11	1876	1947–1969	312 ± 3.0	315
82.0–83.0	28	1867	1938–1960	311 ± 3.0	
92.0–93.0	25	1850	1921–1943	306 ± 3.0	
102.0–103.0	26	1832	1903–1925	300 ± 3.0	
111.0–112.0	26	1812	1883–1905	297 ± 3.0	
128.0–129.0	47	1782	1842–1864	288 ± 3.0	
147.0–147.2	10	1743	1814–1836	284 ± 3.0	
162.0–162.3	9	1723	1794–1819	280 ± 3.0	
177.0–177.3	10	1683	1754–1776	279 ± 3.0	
187.0–187.3	10	1663	1734–1756	279 ± 3.0	

South Pole

	IRLS/LES			IRLS	LES
139.0–141.0	12/5	820	1660–1880	278 ± 3.0	280 ± 3.0
160.0–162.0	12/5	610	1450–1670	281 ± 3.0	283 ± 3.0
205.0–207.0	8/4	110	950–1170	279 ± 4.0	280 ± 3.0

LES, Large extraction system.

±2 yr (ref. 3). Based on porosity measurements, the time lag between the mean age of the gas and the age of the ice was determined to be 95 yr and the duration of the close-off process to be 22 yr (ref. 4). These values are, of course, evaluated for one particular core representing the present situation (1983), assuming a homogeneous enclosure process and not taking into account the sealing effect of observed impermeable layers.

The gases from ice samples were extracted by a dry-extraction system, in which bubbles are crushed mechanically to release the trapped gases, and then analysed for CO_2 by infrared laser absorption spectroscopy (IRLS) or by gas chromatography (GC). The combination of a needle crusher and IRLS allows us to analyse samples of ice as small as 1 g (ref. 5) (see Fig. 1 legend). During the past 2 yr, a new large dry-extraction device for ice samples up to 1 kg has been built and extensively tested[6]. The ice samples are ground in vacuo and the gases collected on a cold finger (20 K) and later analysed by both GC and IRLS. Applying both procedures to neighbouring ice samples gave the same concentration to within 1%. The measurements using the needle crusher, published previously[5,7,8], were performed using a slightly modified procedure and exhibited generally lower CO_2 concentrations by 15 p.p.m.v. (see Fig. 1 legend). In 1982 an intercalibration study with the Grenoble laboratory[7] was performed using the small crusher with the older measuring procedure. Based on our new results, the agreement of the intercalibration must be viewed as a discrepancy, which we will try to resolve in the near future with a new intercalibration series.

Figure 1 shows the mean CO_2 concentration of the 12 depth intervals, measured on the Siple core, plotted against the mean gas age. To extend our data series further back in time, a few samples from the South Pole were also measured (and cross-checked with the large extraction system). These values are listed in Table 1.

Five depth intervals between 82 and 130 m.b.s. of the Siple core were measured with a high spatial resolution. Figure 2a–e shows the individual data series together with the $\delta^{18}O$ results. A seasonal structure could not be identified, indicating that the CO_2 concentration in the bubbles of the Siple core is homogeneously distributed and is independent of the seasonality of the surrounding ice.

Based on earlier studies of ice samples from locations in Greenland, we expected to measure enhanced CO_2 concentration in the extracted gas, especially in summer layers[8,9] from locations with a mean annual air temperature above −25 °C. For comparable mean gas ages, the CO_2 concentration measured

on samples from the South Pole (−50 °C) and Siple Station (−24 °C) do not differ within the error limit. Therefore, we conclude that the CO_2 level in the bubbles of the Siple core is not influenced by a temperature effect. This is further supported by the lack of seasonal variations in the five data series measured with high spatial resolution.

Between 68 and 69 m.b.s., the porosity measurements showed two layers with bubble volumes above 80 cm³ kg$_{ice}^{-1}$, which correspond to complete enclosure. This is clearly above the 40 cm³ kg$_{ice}^{-1}$ expected from a smooth homogeneous enclosure process. These impermeable layers have a sealing effect if they are imbedded in firn with reduced permeability[10]. Because of the layers between 68 and 69 m.b.s., the air below is already completely isolated, about 7 m above the depth obtained, assuming a homogeneous enclosure. Consequently, for this core, the difference between ice and mean gas age is only 80–85 yr instead of 95 yr as estimated previously[4].

The enclosure of air occurs in winter layers at shallower depth than in summer layers, which could lead to a seasonal modulation of the CO_2 concentration in the bubbles during a period of changing atmospheric CO_2 concentration in case of a smooth and homogeneous enclosure process. The abovementioned lack of a seasonal variation in the series from 82 m.b.s. therefore further supports the sealing effect of such layers.

If the 328 p.p.m. measured at a depth of 68.5 m.b.s. is matched with the atmospheric South Pole record[11], the mean gas age is 10 yr, corresponding to a difference between mean gas age and ice age of 82 yr, which lies in the above estimated range. This difference is used in calculating the mean gas age for all depths. That the CO_2 concentration measured on the subsequent samples from 72.5 and 76.5 m.b.s. corresponds with the atmospheric South Pole record justifies this age determination and indicates, also, that impermeable layers at a depth of ~68 m.b.s. occur frequently.

There remains the question of the duration of the gas-enclosure time in such an impermeable layer. Assuming, as a first approximation, a constant and continuous enclosure rate ending at the time of core drilling (t_2 = AD1983), the beginning (t_1) can be calculated from the expression $(t_2 - t_1)C_{bubbles} = \int_{t_1}^{t_2} C_{atm}(t)\,dt$, where $C_{atm}(t) = a + b(t - 1958) + c(t - 1958)^2$ (with t in yr AD). The atmospheric concentration, C_{atm}, is taken from a quadratic fit of the measured CO_2 data in the atmosphere. The time t_1 so determined is 1963, so that the duration of the close-off is 20 yr, in good agreement with the estimate of Schwander and Stauffer[4].
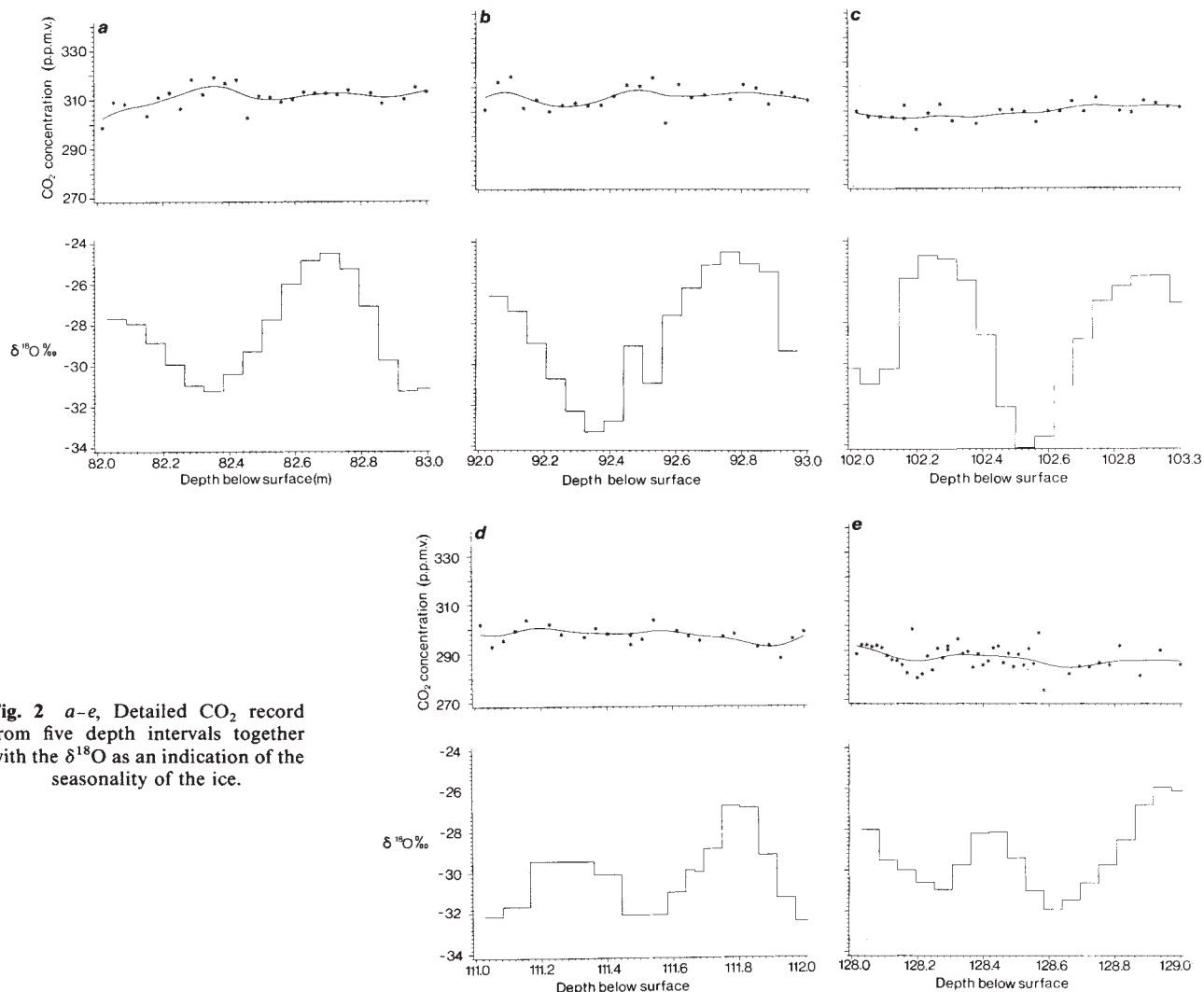
Fig. 2 a-e, Detailed CO_2 record from five depth intervals together with the $\delta^{18}O$ as an indication of the seasonality of the ice.

We would have more difficulties explaining values lower by 15 p.p.m.v., as obtained, for example by the open-valve method. This would require a 20-40-yr-older mean gas age in the bubbles, depending on the development of the atmospheric CO_2 concentration. A very much prolonged close-off interval, a possibility which we exclude[4], or an incomplete mixing of air in the firn would be required. Also, the atmospheric CO_2 concentration would have to be increased between 1930 and 1960 by ~1 p.p.m.v. yr^{-1}, a similar rate to that of the past 20 yr.

Based on these results, we conclude that the atmospheric CO_2 concentration around 1750 was 280 ± 5 p.p.m.v. and has increased since, essentially because of human factors, by 22.5% to 345 p.p.m.v. in 1984. Back extrapolation of the Mauna Loa curve, on the basis of the fossil fuel input data obtained by Rotty[12] and assuming zero CO_2 input from biomass burning, suggests a preindustrial level of 297 p.p.m.v. (ref. 2). The difference of 17 p.p.m.v. between this calculated value and that

measured indicates a significant contribution from biomass burning, even during the past century when the contribution from fossil fuel consumption was still small. The CO_2 increase in the Siple core will be studied in more detail using $\delta^{13}C$ measurements. Preliminary $\delta^{13}C$ results on an ice core from the South Pole are indeed very promising[13]. The CO_2/air $^{13}C/^{12}C$ record on the Siple ice core and the $^{14}C/^{12}C$ Suess effect from tree rings, together with the fossil fuel input-time history, will then enable us to separate the CO_2 inputs from those attributable to fossil fuels and biomass. It will also allow us to determine the effective airborne fraction for the combined input and to check the validity of the present carbon-cycle models.

1. Keeling, C. D. et al. Tellus 28, 538-551 (1976).
2. WMO Rep. 10 (World Meteorological Organization, Geneva, 1983).
3. Schwander, J. thesis, Univ. Bern (1984).
4. Schwander, J. & Stauffer, B. Nature 311, 45-47 (1984).
5. Zumbrunn, R., Neftel, A. & Oeschger, H. Earth planet. Sci. Lett. 60, 318-324 (1982).
6. Moor, E. & Stauffer, B. J. Glaciol. (in the press).
7. Barnola, J. M., Raynaud, D., Neftel, A. & Oeschger, H. Nature 303, 410-412 (1983).
8. Neftel, A., Oeschger, H., Schwander, J., Stauffer, B. & Zumbrunn, R. Nature 295, 220-223 (1982).
9. Stauffer, B. et al. Ann. Glaciol. 5, 160-164 (1984).
10. Stauffer, B. et al. Ann. Glaciol. (in the press).
11. Keeling, C. D. et al. Tellus 28, 552-564 (1976).
12. Rotty, R. M. in Carbon Cycle Modelling Scope 16 (ed. Bolin, B.) 121-126 (Wiley, New York, 1981).
13. Friedli, H. et al. Geophys. Res. Lett. 11, 1145-1148 (1984).