

## REPORTS

$$H_W = \sigma_w / (dM_S)$$

where  $\sigma_w$  is domain wall energy per unit area.

$$H_0 = 8M_S(1 + d^2/t^2)^{1/2} E(k, \pi/2) - 8dM_S/t$$

where

$$E(k, \pi/2) = \int_0^{\pi/2} (1 - k^2 \cos^2 \theta)^{1/2} d\theta$$

and  $k = (1 + t^2/d^2)^{-1/2}$ .  $H_0$  is given by the magnetic field from the rectangular parallelepiped magnet with magnetization of  $M_S$  and dimension of  $d \times d \times t$ .

11. Supported by Core Research for Evolutional Science and Technology (CREST) of the Japan Science and Technology Corporation (JST) and by The New Energy and Industrial Technology Development Organization (NEDO), Japan.

11 March 1999; accepted 11 May 1999

# Century-Scale Shifts in Early Holocene Atmospheric CO<sub>2</sub> Concentration

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The inverse relation between atmospheric carbon dioxide concentration and stomatal frequency in tree leaves provides an accurate method for detecting and quantifying century-scale carbon dioxide fluctuations. Stomatal frequency signatures of fossil birch leaves reflect an abrupt carbon dioxide increase at the beginning of the Holocene. A succeeding carbon dioxide decline matches the Preboreal Oscillation, a 150-year cooling pulse that occurred about 300 years after the onset of the Holocene. In contrast to conventional ice core estimates of 270 to 280 parts per million by volume (ppmv), the stomatal frequency signal suggests that early Holocene carbon dioxide concentrations were well above 300 ppmv.

The records of the relation of greenhouse gases to Quaternary climate change come largely from ice cores from Antarctica and Greenland. Trends in the atmospheric CO<sub>2</sub> amount parallel those of the temperature inferred from the isotopic compositions of oxygen ( $\delta^{18}\text{O}$ ) and hydrogen ( $\delta\text{D}$ ) during the past 250,000 years, showing that variation in greenhouse gas concentrations is an important factor in long-term glacial-interglacial climate evolution (1). Carbon dioxide data from ice cores also seem to correlate with millennial-scale temperature changes (2). However, a correlation of atmospheric CO<sub>2</sub> amounts to century-scale climate shifts in the Holocene (3, 4) is still unclear. Most of the Holocene ice core records from Antarctica do not have adequate temporal resolution (5). In Greenland ice, the Holocene CO<sub>2</sub> concentrations are generally considered to be influenced by postdepositional enrichment (6). Because of the apparent inadequacies and controversies in the CO<sub>2</sub> records derived from ice sheets, alternative methods have to be developed to improve the accuracy of detecting and

quantifying possible short-term shifts in the Holocene atmospheric CO<sub>2</sub> regime. Here, we provide a century-scale record of early Holocene atmospheric CO<sub>2</sub> amounts, based on a stomatal frequency analysis of leaves that were buried in peat deposits.

An analysis of herbarium material collected over the past 200 years and controlled growth experiments under preindustrial CO<sub>2</sub> amounts (7, 8) has shown that, for Northern Hemisphere tree species, stomatal frequency decreases linearly as atmospheric CO<sub>2</sub> concentration increases. A near-annual analysis of a 40-year record of the buried leaves of a solitary growing birch (*Betula pendula*) has illustrated that deciduous trees are equipped with a plastic phenotype, capable of a lifetime adjustment of stomatal frequency to an increase in anthropogenic CO<sub>2</sub> (9).

Stomatal frequency is conventionally expressed in terms of stomatal density and stomatal index (SI) (10). In contrast to stomatal density, SI expresses frequency changes independently of variation in epidermal cell size and therefore is the more sensitive parameter for detecting stomatal frequency response to changes in CO<sub>2</sub> concentration (11). The effects of intrinsic variation in SI values within and among leaves of an individual tree species (11, 12) can be accounted for analytically, allowing the replication of temporal trends of mean SI values (9, 13). At least for European tree birches (*B. pendula* and *B. pubescens*), field studies and controlled-envi-

attributed to the change in the magnetostatic energy of the domain caused by the relation of the temperature-dependent  $M_C$  to the stability of the formation of the bubble domain (1). The elongated domain is transformed into the collapse of the bubble domain as  $M_C$  is decreased (Fig. 1, D and F), because  $H_W$  increases and  $H_D$  decreases with decreased  $M_C$ , leading to the instability of the domain as a result of decreasing magnetostatic energy. For the formation of the bubble domain at 72 K, the important factor is not only the value of  $M_C$  but also the thin domain originating in the large anisotropic crystalline structure. If the domain is thicker, the elongated domain will appear because  $H_D$  increases with increasing  $t$ . The balance between the appropriate value of  $M_C$  and the thickness of the domain is preserved for the stable formation of the bubble domain at 72 K.

At 87 K, a steep structure appears with a larger absolute value of  $B_z$  than that at lower temperatures (Fig. 1G). In this temperature range, the magnetic moments nearly lie in the MnO<sub>2</sub> plane. Therefore, the structure represents a stray field arising from the boundaries of magnetic domains with in-plane magnetization. At 97 K, the  $B_z$  value of the observed structure is increased because of further tilting of the magnetic moments toward the MnO<sub>2</sub> plane (Fig. 1H). The size of the domain is nonuniform.

The observed close-packed bubble domain in the structure is promising for high-density magnetic recording. Removing the need for an external magnetic field for the generation of the domain provides a cost incentive for the downsizing of the device. In terms of its practical application, the bubble domain structure obtained in this study is still limited in temperature range; moreover, the size of the domain is not small enough. Further optimization of the material should be achieved through raising the temperature of the stable formation of the smaller bubble domains, either by finding a new composition for the layered structure or by fabricating an artificial layer structure.

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10.  $H_W$  and  $H_D$  are defined as follows:

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ronment experiments show that CO<sub>2</sub>-induced trends in mean SI values are not substantially disturbed by influences of environmental factors such as light, temperature, and nutrient supply (14). Calibrated against the Mauna Loa record of CO<sub>2</sub> increase (15), mean SI values for individual tree species may be empirically modeled as a function of changing atmospheric CO<sub>2</sub> concentrations (16). Response curves can be efficiently applied in the quantification of time-series data on stomatal frequency derived from fossil leaves of extant tree species. Long-term stomatal frequency changes in fossil leaves correlate with general glacial-interglacial CO<sub>2</sub> dynamics (17) and have been used to estimate atmospheric CO<sub>2</sub> concentrations in the late Miocene, Pliocene, and early Pleistocene (8).

We studied leaf material from a peat section that was temporarily exposed at the Borchert archaeological site near Denekamp, northeastern Netherlands (18). The sequence covers part of the Late Glacial (Younger Dryas) and the Holocene. We collected leaves of European tree birches (*B. pendula*

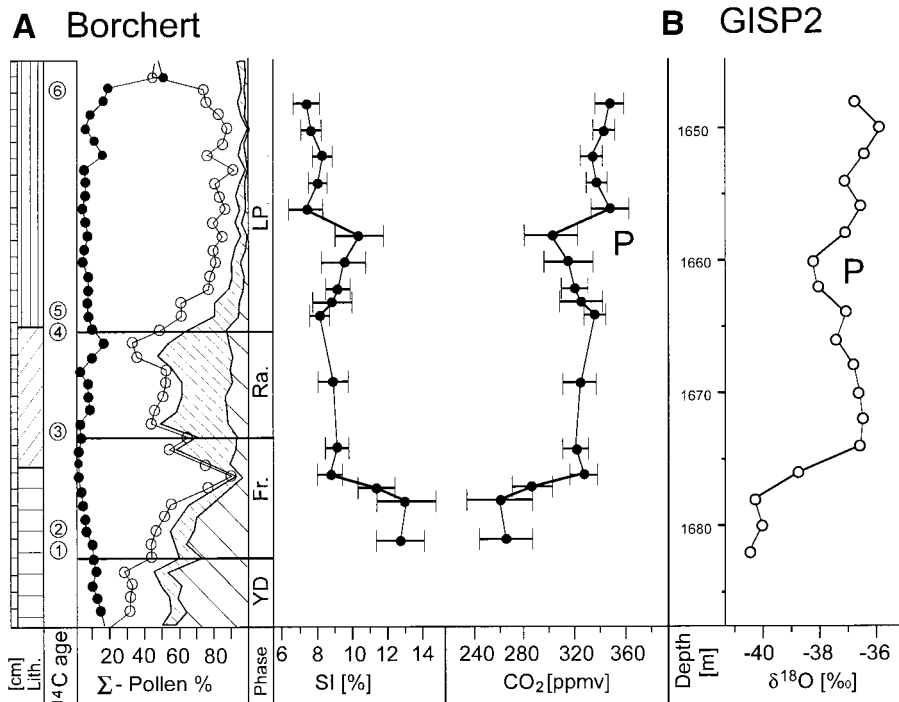
and *B. pubescens*) from 16 horizons of the early Holocene (Preboreal) part of the section. Regionally, the Preboreal is subdivided into the Friesland phase, the Rammelbeek phase, and the Late Preboreal (Fig. 1A). Characterized by the spread of tree birches, the Friesland phase marks the rapid expansion of woodland at the beginning of the Holocene. Six <sup>14</sup>C dates suggest an average sampling resolution of 40 to 50 years.

Leaves of *B. pendula* and *B. pubescens* display essentially similar SI patterns (16). In stomatal frequency analysis, therefore, the mixed fossil assemblage of leaves of tree birches from the Borchert section can be treated as a single category (Fig. 1A). We used the rate of historical CO<sub>2</sub> responsiveness of tree birches (Fig. 2) to derive a Preboreal atmospheric CO<sub>2</sub> record based on the mean SI values for the fossil leaf remains. In the Friesland phase, inferred CO<sub>2</sub> concentrations of 265 ± 21 and 260 ± 25 parts per million by volume (ppmv) are followed by a rapid rise to 327 ± 10 ppmv and a more gradual increase to a maximum of 336 ± 8 ppmv in the early part of the Late

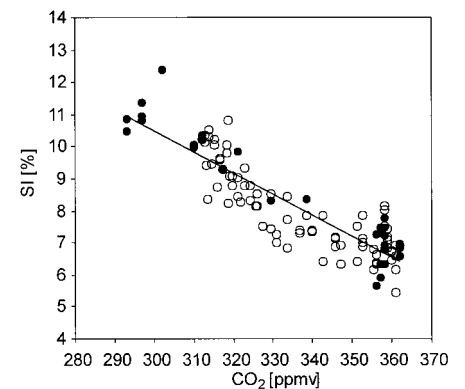
Preboreal. Then, there is a continuous CO<sub>2</sub> decline to a minimum of 301 ± 21 ppmv, followed by a sharp increase to 348 ± 14 ppmv. In the uppermost part of the studied interval, CO<sub>2</sub> concentrations stabilize again to values between 333 ± 8 and 347 ± 11 ppmv.

The initial decrease of the SI in the Friesland phase suggests that atmospheric CO<sub>2</sub> concentrations rose by ~65 ppmv in less than a century. The CO<sub>2</sub> increase occurred during prominent environmental changes, which are reflected in the lithology and the palynological record (18). Basal gyttja formation is followed by a rapid hydrosere succession at the formerly open water site at this time. Regional woodland expansion is reflected by an increase in *Betula* pollen and the occurrence of *Betula* macrofossils. Both the CO<sub>2</sub> increase and the environmental changes at this site correlate with the global climate amelioration at the beginning of the Holocene.

Because of the lack of leaf material from the lowermost part of the section, the onset of the CO<sub>2</sub> rise could not be exactly determined. Yet, the general timing of the rise is in agreement with the CO<sub>2</sub> record from the Antarctic Byrd ice core (19), where the Younger Dryas–Holocene transition is defined by a sudden CO<sub>2</sub> increase from 260 to 280 ppmv. In our SI-based reconstruction, the magnitude of the rise is higher, resulting in CO<sub>2</sub> concentrations well above 300 ppmv. There is a clear covariation (Fig. 1B) between the reconstructed CO<sub>2</sub> increase and the rapid positive δ<sup>18</sup>O shift that



**Fig. 1. (A)** Mean SI values (±1σ) for *B. pendula* and *B. pubescens* from the early Holocene part of the Borchert section (Netherlands; 52.23°N, 7.00°E) and reconstructed CO<sub>2</sub> concentrations. The scale of the section is in centimeters. Three lithological (Lith.) units can be recognized (18): a basal gyttja (=), succeeded by *Drepanocladus* peat (//), which is subsequently overlain by *Sphagnum* peat (||). Six conventional <sup>14</sup>C dates (in years before the present) are available (indicated by circled numbers): 1, 10,070 ± 90; 2, 9930 ± 45; 3, 9685 ± 90; 4, 9770 ± 90; 5, 9730 ± 50; and 6, 9380 ± 80. Summary pollen diagram includes arboreal pollen (white area) with *Pinus* (●) and with *Betula* (○) and nonarboreal pollen with Gramineae (\\) and with Cyperaceae, upland herbs, and Ericales (\\). Regional climatic phases after (18): YD, Younger Dryas; Fr., Friesland phase; Ra., Rammelbeek phase; and LP, Late Preboreal. For analytical method, see (13). Quantification of CO<sub>2</sub> concentrations according to the rate of historical CO<sub>2</sub> responsiveness of European tree birches (Fig. 2). P indicates the reconstructed position of the Preboreal Oscillation. **(B)** δ<sup>18</sup>O profile for the Younger Dryas–Holocene transition in the Greenland GISP2 ice core, after (20); P denotes the δ<sup>18</sup>O-inferred cooling of the Preboreal Oscillation, starting at ~11,300 calendar years before the present (3).



**Fig. 2.** Relation of mean SI for *B. pendula* (○) and *B. pubescens* (●) to the global atmospheric CO<sub>2</sub> increase in the period from 1896 to 1998. The historical training set for the European tree birches consists of 105 samples, originating from presently accumulating peat (9) supplemented by herbarium and field material. For analytical method, see (13). Mean historical CO<sub>2</sub> concentrations are derived from Mauna Loa monitoring (15) and Antarctic shallow ice core data (24). Mean SI values show a linear decrease from 11% at 290 ppmv to 6.4% at 360 ppmv CO<sub>2</sub> [*n* = 105; slope = -0.065; goodness-of-fit linear model: *R*<sup>2</sup> = 0.78, *R*<sup>2</sup><sub>adj</sub> = 0.78; analysis of variance results *F*(1, 103) = 384.97 (*P* < 0.000); statistics performed with SPSS 7.5 for Windows (Statistical Product and Service Solutions, Chicago, Illinois)].

characterizes the onset of Holocene warming in high-resolution isotope records from Greenland ice (20).

About three centuries after the initiation of Holocene warming, a  $\delta^{18}\text{O}$  minimum in Greenland ice reflects a short cooling event (Fig. 1B). A 150-year climate deterioration has also been deduced from numerous terrestrial and marine biorecords (21). Although exact dating of the non-ice core records is hampered by the occurrence of  $^{14}\text{C}$ -age plateaus during the early Holocene, multiproxy analysis suggests that all reported events collectively reflect the Preboreal Oscillation (3). In the Borchert section, the reconstructed  $\text{CO}_2$  values drop from  $\sim 340$  to  $\sim 300$  ppmv at this time (Fig. 1A). A relation between  $\text{CO}_2$  dynamics and the Preboreal Oscillation had been suspected on the basis of an abrupt rise in the early Holocene  $\Delta^{14}\text{C}$  curve inferred from German pine dendrochronology (3, 22), but this could not be confirmed by ice core data.

Our results falsify the concept of relatively stabilized Holocene  $\text{CO}_2$  concentrations of 270 to 280 ppmv until the industrial revolution. SI-based  $\text{CO}_2$  reconstructions may even suggest that, during the early Holocene, atmospheric  $\text{CO}_2$  concentrations that were  $>300$  ppmv could have been the rule rather than the exception (23).

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- leaves per horizon. Seven digitized images (field area,  $0.035 \text{ mm}^2$ ) per leaf were analyzed (standard deviations are constant after seven counts).
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25. We thank B. Aaby, R. Below, and P. J. C. Kuiper for stimulating discussions and for constructive comments on the topic. Supported by the Deutsche Forschungsgemeinschaft (DFG) and the Council for Earth and Life Sciences (ALW) of the Netherlands Organization for Scientific Research (NWO). This is contribution 990501 from the Netherlands Research School of Sedimentary Geology and contribution 510 from the University of Florida Contributions to Paleobiology.

5 March 1999; accepted 17 May 1999

## Contribution of Disturbance to Increasing Seasonal Amplitude of Atmospheric $\text{CO}_2$

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Recent increases in the seasonal amplitude of atmospheric carbon dioxide ( $\text{CO}_2$ ) at high latitudes suggest a widespread biospheric response to high-latitude warming. The seasonal amplitude of net ecosystem carbon exchange by northern Siberian ecosystems is shown to be greater in disturbed than undisturbed sites, due to increased summer influx and increased winter efflux. Increased disturbance could therefore contribute significantly to the amplified seasonal cycle of atmospheric carbon dioxide at high latitudes. Warm temperatures reduced summer carbon influx, suggesting that high-latitude warming, if it occurred, would be unlikely to increase seasonal amplitude of carbon exchange.

Explaining recent changes in the global environment is a scientific challenge with important political and economic implications. Although increases in concentrations of greenhouse gases such as  $\text{CO}_2$  and  $\text{CH}_4$  have clear anthropogenic origins (1), the causes of the observed increased

seasonal amplitude of atmospheric  $\text{CO}_2$  are less clear. The increased amplitude is most pronounced at arctic and subarctic  $\text{CO}_2$  monitoring stations (2) and largely reflects terrestrial carbon exchange at high latitudes (3). Two hypotheses have been advanced to explain this pattern: (i) The recent increase in March-April temperatures in high-latitude continental regions of North America and Siberia (4) could advance snowmelt and increase the length of the growing season (2), causing an increase in productivity and net ecosystem carbon gain (5); or (ii) temperature-driven increases in summer carbon gain balanced by increased winter respiration could enhance the seasonal amplitude of atmo-

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