

# Atmospheric CO<sub>2</sub> fluctuations during the last millennium reconstructed by stomatal frequency analysis of *Tsuga heterophylla* needles

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## ABSTRACT

**A stomatal frequency record based on buried *Tsuga heterophylla* needles reveals significant centennial-scale atmospheric CO<sub>2</sub> fluctuations during the last millennium. The record includes four CO<sub>2</sub> minima of 260–275 ppmv (ca. A.D. 860 and A.D. 1150, and less prominently, ca. A.D. 1600 and 1800). Alternating CO<sub>2</sub> maxima of 300–320 ppmv are present at A.D. 1000, A.D. 1300, and ca. A.D. 1700. These CO<sub>2</sub> fluctuations parallel global terrestrial air temperature changes, as well as oceanic surface temperature fluctuations in the North Atlantic. The results obtained in this study corroborate the notion of a continuous coupling of the preindustrial atmospheric CO<sub>2</sub> regime and climate.**

**Keywords:** carbon dioxide, paleoclimate, Holocene, conifers, climate controls, paleoecology.

## INTRODUCTION

Analysis of the coupling between atmospheric CO<sub>2</sub> levels and climate change under natural conditions in the past is crucial in the forecasting of the impact of anthropogenically produced CO<sub>2</sub> on the coupled climate-carbon cycle system. At present, the notion of covarying atmospheric CO<sub>2</sub> levels and climate is reinforced by the predictions in climate models of (1) a strong rise in global mean temperature as a result of excessive CO<sub>2</sub> emissions (1–7 °C; Houghton et al., 2001), and (2) significant feedback effects of these CO<sub>2</sub>-induced climate changes on the carbon exchange between oceanic, atmospheric, and terrestrial reservoirs (Plattner et al., 2001).

However, it seems as if the role of CO<sub>2</sub> variability in past climate change depends on the time scale studied. The clear coupling between CO<sub>2</sub> and temperature on both glacial-interglacial and millennial scales detected in Antarctic ice cores (Fischer et al., 1999; EPICA Community Members, 2004) is in marked contrast with apparently stable CO<sub>2</sub> levels of the Holocene that reflect neither prolonged climate shifts, such as the late-Holocene gradual cooling, nor centennial-scale cooling events, such as the Preboreal Oscillation and the 8.2 ka event (Indermühle et al., 1999).

It is intriguing that only ice-core records of the last millennium support centennial-scale coupling of CO<sub>2</sub> and Holocene climate; CO<sub>2</sub> changes of 5–15 ppmv are broadly concurrent with Northern Hemisphere climate shifts (Barnola et al., 1995; Etheridge et al., 1996; Indermühle et al., 1999). Unfortunately, data from different coring localities disagree about timing and magnitude of the changes. To resolve these discrepancies and to corroborate the concept that centennial-scale CO<sub>2</sub> variations are involved in late Holocene climate change, stomatal frequency analysis (Woodward, 1987; Royer, 2001) of tree leaves buried in peat and lake deposits provides an alternative method for detecting and quantifying short-term CO<sub>2</sub> fluctuations. The inverse relationship between the number of stomata (pores on plant leaves through which gas exchange occurs) and ambient CO<sub>2</sub> enables the reconstruction of past CO<sub>2</sub> levels with a potentially annual resolution (e.g., Wagner et al., 1996). Stomatal data increasingly substantiate a much more

dynamic Holocene CO<sub>2</sub> evolution than suggested by ice-core data. Coupled to centennial-scale cooling events, CO<sub>2</sub> changes of 20–50 ppmv occur synchronously in records from Europe and North America (Wagner et al., 2004).

So far the available stomatal and ice-core records over the last millennium have insufficient temporal resolution and chronological inaccuracies, and have not provided a picture of CO<sub>2</sub> dynamics that can be correlated with available high-resolution proxy temperature records (Rundgren and Beerling, 1999; Gerber et al., 2003). In this study we present a well-dated, high-resolution CO<sub>2</sub> record from North America, spanning the period A.D. 800–2000. In order to investigate the role of CO<sub>2</sub> as a forcing factor in preindustrial climate change, CO<sub>2</sub> data are compared with selected temperature records.

## MATERIAL AND METHODS

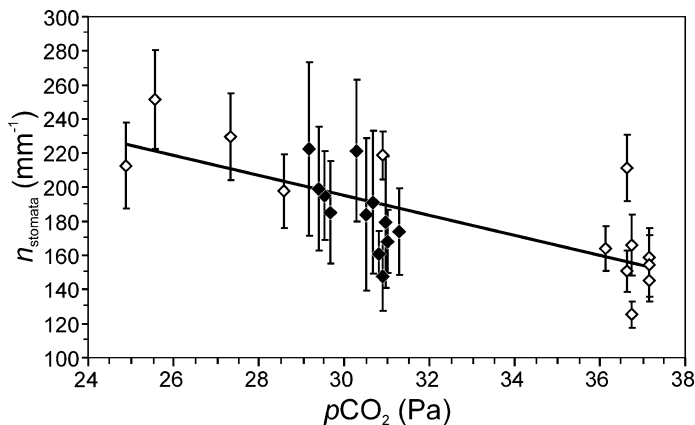
Buried needles of the conifer *Tsuga heterophylla* were recovered from a 91 cm sediment core from Jay Bath, a shallow pond at Mount Rainier (Washington State; 46°46'N, 121°46'W; 1311 m above sea level). The core was cut into 1-cm-thick sediment samples, 3–5 identifiable needles were processed, and stomatal frequency was measured according to the quantification strategy of Kouwenberg et al. (2003).

Quantification of preindustrial CO<sub>2</sub> levels is based on the response rate of *Tsuga heterophylla* needles to the CO<sub>2</sub> increase over the last century (Fig. 1). Needles incorporated in the calibration set originate from contrasting altitudes of the Northwest Pacific region. Because the difference in air pressure with altitude affects CO<sub>2</sub> partial pressure, and thus the amount of CO<sub>2</sub> available for gas exchange, CO<sub>2</sub> levels are expressed as partial pressures (*p*CO<sub>2</sub>) instead of mixing ratios. The stomatal frequency change in the calibration set is not attributable to changes in local precipitation and temperature or needle maturity (Kouwenberg et al., 2003, 2004). Reconstructed CO<sub>2</sub> values were calculated by using a classical linear regression, which allows a better performance at the extremes and with slight extrapolation than inverse linear regression (Osborne, 1991). To allow quantitative comparison with other CO<sub>2</sub> records, reconstructed CO<sub>2</sub> levels for the last millennium are expressed as mixing ratios (Jones, 1992; Kouwenberg, 2004).

Age vs. depth relationships for the sediment core (Fig. 2) were determined by fitting a fourth-order polynomial through a series of five accelerator mass spectrometry <sup>14</sup>C dates and one tephra layer at 21 cm from the A.D. 1481 Mount St. Helens eruption (Mullineaux, 1996). In this way an overall averaged mean sedimentation rate of 1 cm per 27 yr is obtained for the core.

## RESULTS

The reconstructed CO<sub>2</sub> mixing ratios at Jay Bath since A.D. 800 are presented in Figure 3. Preindustrial CO<sub>2</sub> values fluctuated around a long-term average of 280–290 ppmv culminating after A.D. 1850 in a sharp rise from 280 ppmv to 370 ppmv. The CO<sub>2</sub> record is characterized by high-frequency variability. Focusing on centennial-scale changes (the three-point moving average), the record shows prominent CO<sub>2</sub> minima of ~260 ppmv ca. A.D. 860 and A.D. 1150, and modest



**Figure 1.** Response of stomatal parameters of *Tsuga heterophylla* to  $p\text{CO}_2$  increase from 24 to 38 Pa.  $\text{CO}_2$  partial pressure was calculated as  $\text{CO}_2$  mixing ratio (in parts per million by volume) times barometric air pressure  $P_B$  (Pa). Barometric air pressure was estimated according to Jones (1992) as  $P_B = 101.325/e^{(z/29.3)/T}$ , where  $z$  is altitude above sea level and  $T$  air temperature in kelvin (estimated from mean annual temperature at closest weather station, corrected by temperature lapse rate in case of significant altitudinal difference between site and station).  $\text{CO}_2$  mixing ratios of 290–315 ppmv were derived from shallow Antarctic ice cores (Neftel et al., 1985); mixing ratios of 315–368 ppmv are annual means from Mauna Loa monitoring (Keeling and Whorf, 2002). Black diamonds—needles from living trees and short peat core next to Jay Bath (Mount Rainier, Washington); open diamonds—modern and herbarium needles from other localities. Error bars indicate  $\pm 1$  standard error. Solid line indicates best fit in classical linear regression analysis (linear model showed best fit over nonlinear models);  $n_{\text{stomata}}$  is number of stomata per millimeter needle length (same as TSDL as defined in Kouwenberg et al., 2003);  $n_{\text{stomata}} = -5.8581 \times p\text{CO}_2 + 371.14$ ;  $r^2 = 0.5124$  ( $P < 0.001$ ). This calibration data set plotted against  $\text{CO}_2$  mixing ratios was previously published in Kouwenberg et al. (2003).

minima of 275–280 ppmv occurred ca. A.D. 1600 and A.D. 1800. In between, ca. A.D. 1000, ca. A.D. 1300, and ca. A.D. 1700,  $\text{CO}_2$  maxima of 300 ppmv, 320 ppmv, and 300 ppmv, respectively, were estimated.

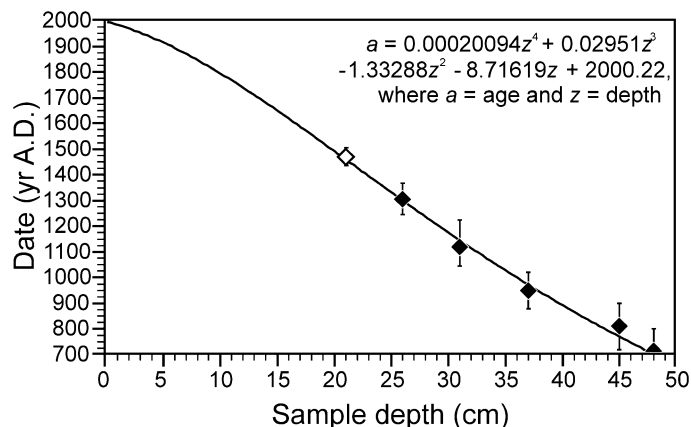
## DISCUSSION

### Comparison with Other $\text{CO}_2$ Records

Due to inherent stomatal frequency variation in tree leaves (Kürschner et al., 1997), the calculated raw proxy  $\text{CO}_2$  data show considerable scatter. Yet the three-point average matches the industrial  $\text{CO}_2$  rise after A.D. 1800 very well. The last six data points, which are completely independent from the calibration data set, clearly reproduce the  $\text{CO}_2$  increase from 280 to 370 ppmv, evident from shallow Antarctic ice cores (Neftel et al., 1985) and instrumental measurements at Mauna Loa since A.D. 1958 (Keeling and Whorf, 2002). This accurate correspondence corroborates the reliability of the older reconstructed  $\text{CO}_2$  fluctuations.

The presence of short-term  $\text{CO}_2$  variability in the last millennium is supported by stomatal records from Europe. Despite much lower chronological accuracy, a comparable pattern of centennial-scale fluctuations was recognized in stomatal frequency data of *Salix* leaves from Sweden (Rundgren and Beerling, 1999). A very high-resolution record based on *Quercus* leaves from the Netherlands, spanning the first half of the last millennium, shows a remarkably similar decrease in  $\text{CO}_2$  between A.D. 1000 and A.D. 1200, a sharp increase of  $\pm 34$  ppmv until A.D. 1300, and a slow decrease until A.D. 1500 (Wagner et al., 2004).

The observed  $\text{CO}_2$  fluctuations in the Jay Bath record, up to 50 ppmv, exceed those measured in Antarctic ice cores (Fig. 3). The most prominent feature in the stomatal  $\text{CO}_2$  reconstruction, the  $\text{CO}_2$  mini-



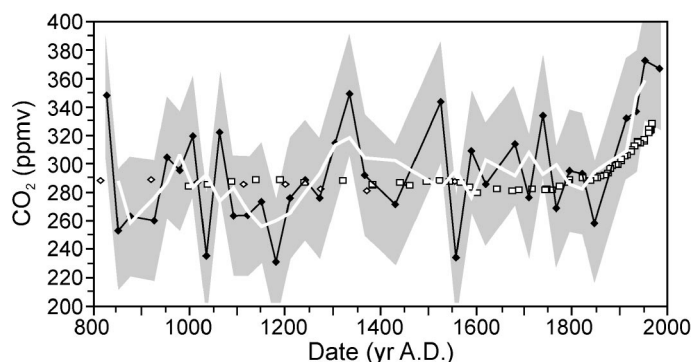
**Figure 2.** Age ( $a$ ) vs. depth ( $z$ ) diagram for Jay Bath core. White diamond—tephra layer from A.D. 1481 Mount St. Helens eruption (Mullineaux, 1996). Black diamonds— $^{14}\text{C}$  accelerator mass spectrometer dates converted to calendar ages by using Oxcal 3.8 (Bronk Ramsey, 1995) and INTCAL98 calibration data (Stuiver et al., 1998). Black line shows fourth-order polynomial providing best fit ( $r^2 = 0.9977$ ). Error bars indicate  $2\sigma$  (95.4%) probability ranges.

mum ca. A.D. 1200 and a subsequent rise during that century, is also reflected in the South Pole and D47 ice cores, albeit with a smaller amplitude maximum of 12 ppmv (Siegenthaler et al., 1988; Barnola et al., 1995). The Law Dome ice core temporal resolution in the earlier part of the record is insufficient to have captured this short-term  $\text{CO}_2$  event (Etheridge et al., 1996).

The discrepancies between the ice-core and stomatal reconstructions may partially be explained by varying age distributions of the air in the bubbles because of the enclosure time in the firn-ice transition zone. This effect creates a site-specific smoothing of the signal (decades for Dome Summit South [DSS], Law Dome, even more for ice cores at low accumulation sites), as well as a difference in age between the air and surrounding ice, hampering the construction of well-constrained time scales (Trudinger et al., 2003). However, the large amplitude of the  $\text{CO}_2$  signal in the Jay Bath record, compared to the ice cores and other stomatal reconstructions, may be influenced by the relatively low amount of needles in the low- $\text{CO}_2$  part of the calibration data set.

### Sources of Carbon

To explain substantial centennial-scale  $\text{CO}_2$  shifts during the last millennium, a mechanism must be found to produce such rapid changes



**Figure 3.** Reconstructed  $\text{CO}_2$  mixing ratios based on stomatal frequency counts on *Tsuga heterophylla* needles for A.D. 800–2000. Black line connects means of 3–5 needles per sample; thick white line shows three-point moving average. Gray area indicates confidence interval of  $\pm 1$  root mean standard error. White squares— $\text{CO}_2$  data from Law Dome ice core (Etheridge et al., 1996); white diamonds— $\text{CO}_2$  data from Taylor Dome ice core (Indermühle et al., 1999).

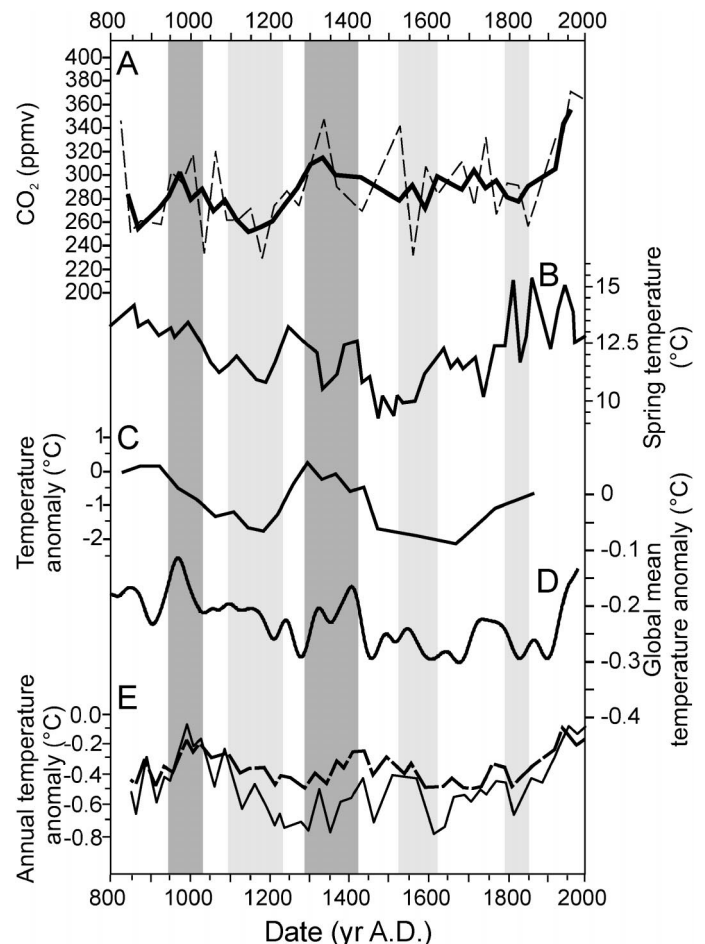
in carbon cycle dynamics. Ruddiman (2003) argued that decadal- to centennial-scale CO<sub>2</sub> anomalies of 4–10 ppmv observed in ice-core records could be the result of widespread reforestation of agricultural areas in Europe and Asia, which were abandoned after outbreaks of epidemic diseases. However, although natural or anthropogenic changes in the terrestrial biosphere are likely to have played a role in short-term carbon cycle dynamics, the amount of carbon needed to cause a shift of 50 ppmv would far exceed the size of potential carbon sources and sinks in the terrestrial biosphere (Ruddiman, 2003). Moreover, the lack of high-resolution land-cover reconstructions for the last millennium does not permit a detailed comparison between centennial-scale changes in land cover and the CO<sub>2</sub> record.

The oceans, on the other hand, may be a much larger potential source or sink for atmospheric carbon (Raven and Falkowski, 1999). Exchange of CO<sub>2</sub> between atmosphere and ocean is highly dependent on physicochemical factors affecting the solubility of CO<sub>2</sub> in the ocean surface waters, such as alkalinity, dissolved inorganic carbon content, salinity, and temperature (Takahashi et al., 1993). Involvement of sea-surface temperature (SST) changes in the production and depletion of atmospheric CO<sub>2</sub> is strongly suggested by the apparent synchronicity between the timing of CO<sub>2</sub> maxima and minima in the stomata-based record (within uncertainty limits), and changes in North Atlantic Ocean SST as recorded offshore the Mid-Atlantic United States (Cronin et al., 2003) (Fig. 4B), as well as offshore West Africa (DeMenocal et al., 2000) (Fig. 4C). Provided that SST fluctuations at these sites reflect SST changes in the entire North Atlantic, temperature-driven changes in the CO<sub>2</sub> flux between ocean surface waters and atmosphere may be invoked as a plausible mechanism to explain at least a substantial part of the reconstructed CO<sub>2</sub> variations over the last millennium. For example, a modeled cooling of 2.7 °C in the Atlantic Ocean (north of lat 20°N) during the Younger Dryas produced a local *p*CO<sub>2</sub> decrease of 2.7 Pa (Marchal et al., 1999). Apart from SST change, climate-induced changes in salinity, carbonate chemistry, circulation, and marine biological productivity in the Atlantic or the other ocean basins, such as the Southern Ocean, may also have affected atmospheric CO<sub>2</sub> levels (Sarmiento and Orr, 1991; Marchal et al., 1998; Plattner et al., 2001).

### CO<sub>2</sub> and Air Temperature

The CO<sub>2</sub> changes as detected in the stomatal records should be expected to significantly affect radiative forcing and thus global air temperatures. However, atmospheric CO<sub>2</sub> levels are influenced by temperature-induced changes in biospheric and marine feedback systems. In order to investigate the potential of rapid CO<sub>2</sub> changes as a forcing factor in climate over the last millennium, the CO<sub>2</sub> record is compared to global temperatures. Some remarkable correlations are revealed between the stomata-based CO<sub>2</sub> record and a multiproxy-based reconstruction of global mean temperature (Mann and Jones, 2003), particularly with respect to the timing of the warm periods and the CO<sub>2</sub> maxima ca. A.D. 950 and A.D. 1300 (Fig. 4D). The overall picture suggests a clear covariation between CO<sub>2</sub> and global temperature. Note, however, that the Southern Hemisphere contribution to the global mean temperature estimation is based on limited proxy data.

A causative link between CO<sub>2</sub> and terrestrial air temperature would be most prominent in Northern Hemisphere records, because models of the spatial distribution of a future temperature rise due to anthropogenic CO<sub>2</sub> emission indicate that the greatest warming is expected to occur at high latitudes on the Northern Hemisphere continents (Houghton et al., 2001). Similar centennial-scale fluctuations can be recognized in Northern Hemisphere air temperature records based on extratropical tree-ring records (Briffa, 2000; Esper et al., 2002) (Fig. 4E). In model sensitivity studies on the evolution of CO<sub>2</sub> and Northern Hemisphere temperature in response to changes in radiative forcing over the last millennium, CO<sub>2</sub> fluctuations of at least 20 ppmv were



**Figure 4. Comparison between reconstructed CO<sub>2</sub> mixing ratios and Northern Hemisphere climate records. A:** CO<sub>2</sub> mixing ratios from stomatal counts on *Tsuga heterophylla* needles from Jay Bath. Thin dashed line connects means of 3–5 needles per sample depth, thick line three-point moving average to emphasize centennial scale trends. Dark gray bands—periods of high CO<sub>2</sub> mixing ratios; light gray bands—periods of low CO<sub>2</sub> mixing ratios (based on three-point average trends). **B:** Sea-surface temperatures of Chesapeake Bay (Mid-Atlantic United States) reconstructed from Mg/Ca ratios of ostracods (Cronin et al., 2003). **C:** Sea-surface temperature anomalies offshore West Africa as reconstructed from foraminiferal assemblages (DeMenocal et al., 2000). Shown are cold-season anomalies; warm-season anomalies have similar pattern (not depicted). **D:** Global mean temperatures (45 yr running average) from multiproxy records based on 1961–1990 reference period (Mann and Jones, 2003). **E:** Summer temperature anomalies in tree-ring records from Northern Hemisphere. Dashed line is from Briffa (2000); black line is from Esper et al. (2002).

connected with Northern Hemisphere temperature changes on the scale of the Esper tree-ring record (under very high solar forcing conditions) (Gerber et al., 2003). Such changes are larger than those observed in ice cores, but are compatible with the stomata-based record.

The CO<sub>2</sub> minimum at A.D. 1150 and the maximum ca. A.D. 1400 seem to correspond to terrestrial temperature changes that occurred 50–100 yr later, suggesting a forcing role for CO<sub>2</sub> fluctuations. However, attempts to determine leads and lags by simply comparing the CO<sub>2</sub> and air temperature records have some important drawbacks. The chronology of the CO<sub>2</sub> record contains uncertainties on the scale of several decades. Furthermore, climate changes over the last millennium are the result of the interplay between a number of forcing factors; variations in solar irradiance output and volcanic eruptions are at least as important as CO<sub>2</sub> in terms of influence on radiative forcing and thus on global temperature records. In order to truly assess the impact of rapid

CO<sub>2</sub> changes on Holocene climate, radiative forcing models must be run, including other known forcing factors as well as a more dynamic CO<sub>2</sub> regime for which this study provides support.

## CONCLUSIONS

The centennial-scale variability in atmospheric CO<sub>2</sub> concentration linked to documented global and regional temperature change since A.D. 800, recognized in this study, corroborates continuous coupling of CO<sub>2</sub> and climate during the Holocene. For the first time, CO<sub>2</sub> changes inferred from stomatal frequency analysis have been related to coeval variation in Atlantic SSTs providing evidence that CO<sub>2</sub> fluctuations over the last millennium at least partly could have originated from temperature-driven changes in CO<sub>2</sub> flux between ocean surface waters and atmosphere. Because the CO<sub>2</sub> variation also shows similarities with terrestrial air temperature trends in the extratropical Northern Hemisphere regions—which are the areas most sensitive to global warming—it may be hypothesized that throughout the last millennium, CO<sub>2</sub> could have served as a forcing factor for terrestrial air temperature.

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