

# A role for atmospheric CO<sub>2</sub> in preindustrial climate forcing

Thomas B. van Hoof<sup>\*†</sup>, Friederike Wagner-Cremer<sup>†</sup>, Wolfram M. Kürschner<sup>†</sup>, and Henk Visscher<sup>†</sup>

<sup>\*</sup>TNO Geological Survey of the Netherlands, Princetonlaan 6, 3584 CB Utrecht, The Netherlands; and <sup>†</sup>Palaeoecology, Institute of Environmental Biology, and Laboratory of Palaeobotany and Palynology, Utrecht University, Budapestlaan 4, 3584 CD Utrecht, The Netherlands

Communicated by David L. Dilcher, University of Florida, Gainesville, FL, August 21, 2008 (received for review March 3, 2007)

**Complementary to measurements in Antarctic ice cores, stomatal frequency analysis of leaves of land plants preserved in peat and lake deposits can provide a proxy record of preindustrial atmospheric CO<sub>2</sub> concentration. CO<sub>2</sub> trends based on leaf remains of *Quercus robur* (English oak) from the Netherlands support the presence of significant CO<sub>2</sub> variability during the first half of the last millennium. The amplitude of the reconstructed multidecadal fluctuations, up to 34 parts per million by volume, considerably exceeds maximum shifts measured in Antarctic ice. Inferred changes in CO<sub>2</sub> radiative forcing are of a magnitude similar to variations ascribed to other mechanisms, particularly solar irradiance and volcanic activity, and may therefore call into question the concept of the Intergovernmental Panel on Climate Change, which assumes an insignificant role of CO<sub>2</sub> as a preindustrial climate-forcing factor. The stomata-based CO<sub>2</sub> trends correlate with coeval sea-surface temperature trends in the North Atlantic Ocean, suggesting the possibility of an oceanic source/sink mechanism for the recorded CO<sub>2</sub> changes.**

carbon cycle | global warming | past millennium | stomata

It is increasingly realized that temperature-sensitive proxy records inferred from tree rings, lake deposits, and historical documents corroborate occurrences of significant preindustrial air-temperature fluctuations during the last millennium (1–5). Also, the *Fourth Assessment Report* of the Intergovernmental Panel on Climate Change (IPCC) (6) now cautiously presents a whole range of historical temperature reconstructions instead of favoring the earlier “hockey-stick” graph of the *Third Assessment Report* of the IPCC (7). The reconstructed fluctuations show largely differing amplitudes and timing. It is obvious that individual proxy temperature curves are not parallel to the generally accepted atmospheric CO<sub>2</sub> curve for the last 1,000 years, which is characterized by a very low degree of preindustrial variability. This curve is based on CO<sub>2</sub> records from Antarctic ice cores, which suggest that until the onset of industrialization in the 19th century, atmospheric CO<sub>2</sub> concentration (expressed as mixing ratio) varied by not more than 12 parts per million by volume (ppmv) (8–12). Although modest negative CO<sub>2</sub> anomalies have been associated with the Little Ice Age (10, 11, 13, 14), the *Fourth Assessment Report* treats such variation as an insignificant forcing mechanism for generating preindustrial air-temperature changes (6), especially when compared with effects of changes in solar irradiance and explosive volcanic activity (15–18).

Estimates of preindustrial CO<sub>2</sub> levels are available not only from Antarctic ice but also from leaves of land plants preserved in peat and lake deposits. Particularly in a wide variety of woody plants, the genetically controlled inverse relationship between numbers of leaf-stomata (gas exchange pores) and ambient CO<sub>2</sub> concentration during the growth period (19) permits detection and quantification of past CO<sub>2</sub> changes by analyzing time-series data on stomatal frequency. The *Fourth Assessment Report* recognizes that stomatal frequency may provide reasonable constraints on past CO<sub>2</sub> variations on long geological time scales (10<sup>5</sup> to 10<sup>8</sup> years), but does not appreciate the applicability of this proxy for identifying decadal to millennial scale CO<sub>2</sub> changes

during the Holocene Epoch (6). Yet, the integrity of short-term leaf-based CO<sub>2</sub> changes has been verified by fine-resolution analysis of the lifetime CO<sub>2</sub> responsiveness of individual trees (20) and by numerous other response curves based on well dated herbarium material and subfossil leaves, which consistently mimic the ongoing CO<sub>2</sub> increase apparent from Mauna Loa instrumental monitoring (21–24). Reproducibility of leaf-based CO<sub>2</sub> reconstructions is further demonstrated by coeval stomatal frequency records of taxonomically, geographically, and ecologically contrasting tree species, which confirm a coupling between CO<sub>2</sub> anomalies and early Holocene cooling events (25–28).

For the last millennium, pronounced preindustrial CO<sub>2</sub> variability has been reconstructed on the basis of needles of *Tsuga heterophylla* (western hemlock) from Mount Rainier, Washington, USA (29), and leaf remains of *Quercus robur* (English oak) from the southeastern part of the Netherlands (27, 30). The timing of the detected CO<sub>2</sub> changes is in good agreement with perturbations observed in Antarctic ice core records. Remarkably, however, reconstructed amplitudes >30 ppmv significantly exceed the maximum shifts of 12 ppmv CO<sub>2</sub> found in Antarctic ice. These discrepancies can be explained as an effect of smoothing resulting from diffusion processes in the firn layer at the site of the ice cores. Such processes lead to a reduced signal of the original atmospheric variability and may obscure high-frequency CO<sub>2</sub> variations (31). A modeling exercise, in which raw stomatal frequency data from *Q. robur* leaves were smoothed analogously to natural CO<sub>2</sub> smoothing in the firn, demonstrates that measured CO<sub>2</sub> mixing ratios in the Antarctic D47 core (9) considerably underestimate the actual atmospheric CO<sub>2</sub> variability during the 13th century (32). Apart from smoothing, diffusion is also responsible for a gas-ice age difference in ice cores, resulting in inadequate dating control with age uncertainties of up to 100 years for CO<sub>2</sub> data for the last millennium (11). Unlike ice-based CO<sub>2</sub> records, leaf-based records have the advantage of providing real-time data because the leaf-morphological CO<sub>2</sub> signature becomes permanently fixed at the moment of leaf development and is unaffected by burial processes.

The presence of high-amplitude CO<sub>2</sub> fluctuations as documented by stomatal frequency studies may falsify the IPCC concept that preindustrial temperature variability is constrained by relatively stable atmospheric CO<sub>2</sub> levels (6, 14, 33, 34). A higher degree of CO<sub>2</sub> variability during the last millennium must have resulted in a more prominent role for CO<sub>2</sub> as a forcing factor of air-temperature changes. In this study, the impact of CO<sub>2</sub> changes on preindustrial temperature is reassessed by quantifying the radiative forcing of the alternative CO<sub>2</sub> record

Author contributions: T.B.v.H., F.W.-C., W.M.K., and H.V. designed research; T.B.v.H., F.W.-C., and W.M.K. performed research; T.B.v.H., F.W.-C., W.M.K., and H.V. analyzed data; and T.B.v.H. and H.V. wrote the paper.

The authors declare no conflict of interest.

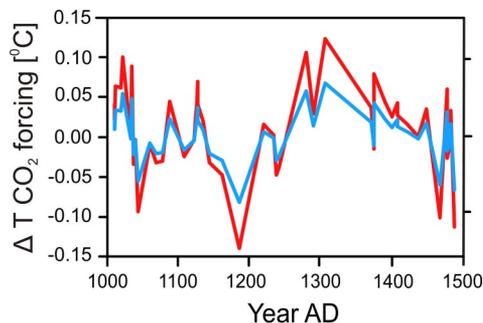
Freely available online through the PNAS open access option.

<sup>†</sup>To whom correspondence should be addressed. E-mail: tom.vanhoof@tno.nl.

This article contains supporting information online at [www.pnas.org/cgi/content/full/0807624105/DCSupplemental](http://www.pnas.org/cgi/content/full/0807624105/DCSupplemental).

© 2008 by The National Academy of Sciences of the USA





**Fig. 2.** Estimated global temperature effects on the SI-based CO<sub>2</sub> forcing calculated with a low (blue line) and high (red line) sensitivity mode of the ECBILT-CLIO coupled atmospheric-ocean-sea ice model (43, 45).

sinks in the terrestrial biosphere. It is likely that, analogous to early Holocene CO<sub>2</sub> changes (25–28), depletion and restoration of atmospheric CO<sub>2</sub> between A.D. 1000 and 1500 was driven mainly by short-term perturbations of sea-surface temperature and/or salinity. Similar to the CO<sub>2</sub> trend based on *Tsuga heterophylla* needles (29), within the dating uncertainties, the present stomata-based CO<sub>2</sub> reconstruction correlates to a large extent with proxy sea-surface temperature records from various parts of the North Atlantic Ocean (36–38).

### Concluding Remarks

A coherent scenario explaining preindustrial atmospheric CO<sub>2</sub> changes of the last millennium and their possible temporal link with changes in terrestrial and marine carbon uptake or release still needs to be established. Reconstructed multidecadal changes are not as prominent as man-made CO<sub>2</sub> increases since the onset of industrialization. Yet it seems obvious that a dynamic CO<sub>2</sub> regime with fluctuations of up to 34 ppmv implies that CO<sub>2</sub> can no longer be discarded as a forcing factor of preindustrial air-temperature changes. The results of our study therefore underscore the need to understand anthropogenic global warming within the context of rates and amplitudes of natural CO<sub>2</sub> variability of the last millennium. A stomata-based CO<sub>2</sub> record may provide an important observational constraint on the sensitivity of climate models.

### Materials and Methods

We based our study on a series of CO<sub>2</sub> estimates derived from well preserved *Q. robur* leaf remains, which occur continually in the organic-rich infill of an oxbow lake of the river Roer near the village of Sint Odiliënberg, Province of

Limburg, southeastern part of the Netherlands (51.088 N 6.008 E; for details see ref. 30). The studied leaf record was derived from 60 successive horizons, which were accurately dated by accelerator mass spectrometry <sup>14</sup>C wiggle-match dating (for details see ref. 30).

Because of significant differences between the stomatal frequency in sun and shade leaves of *Quercus*, we restricted the analysis to sun morphotypes. Standardized stomatal frequency counts were made by using the image-analysis program analySIS 3.0 (Soft Imaging System) on the digitized images. Parameters measured were (mean) epidermal cell density (ED; number per mm<sup>2</sup>) and (mean) stomatal density (SD; number per mm<sup>2</sup>). To evade influences of lateral epidermal cell expansion resulting from contrasting light regimes, leaf age, or water availability (39, 40) from SD and ED, the area-independent (mean) SI (41) was calculated as

$$SI[\%] = [SD/(SD + ED)] \cdot 100. \quad [1]$$

Calculated SI values (Fig. 1A) are mean values for five leaves per sampling point. Seven images per leaf with a field area of 0.03 mm<sup>2</sup> were analyzed (standard deviations are constant after seven counts). SI values were transferred into CO<sub>2</sub> mixing ratios (Fig. 1C) by means of an inference model based on the species-specific stomatal frequency adjustment to the historical atmospheric CO<sub>2</sub> increase of the last ≈150 years. For this model (Fig. 1B), SI values of accurately dated *Q. robur* leaves from Dutch herbaria and young peat deposits were compared with the global atmospheric CO<sub>2</sub> trends recognized at Mauna Loa and in shallow Antarctic ice cores (for details see ref. (23), resulting in the following inference model:

$$CO_2[\text{ppmv}] = -63.902 \ln(SI) + 484.33. \quad [2]$$

To calculate the strength of radiative forcing induced by the CO<sub>2</sub> changes observed in the Dutch stomatal frequency study, we followed the approach of Myhre *et al.* (42), who expressed the radiative forcing as:

$$dF[\text{W/m}^2] = \alpha \cdot \ln(C/CO) + \beta \cdot (\sqrt{C} + \sqrt{CO}), \quad [3]$$

where  $dF$  represents the radiative forcing,  $C$  represents the CO<sub>2</sub> mixing ratio,  $CO$  represents the unperturbed mixing ratio,  $\alpha = 5.35$ , and  $\beta = 0.0906$ .

IPCC arbitrarily takes A.D. 1750 as the preindustrial baseline (43). Therefore, to identify changes in radiative forcing induced by the reconstructed CO<sub>2</sub> changes, normalized stomata-derived CO<sub>2</sub> data were superimposed on the corresponding CO<sub>2</sub> reference level of 278 ppmv. It should be noted that, in general, CO<sub>2</sub> data derived from stomatal frequency analysis have higher average values (≈300 ppmv) compared with the IPCC baseline (21, 25–28). Effects of the changes on global air temperatures were estimated with the ECBILT-CLIO coupled atmosphere–ocean–sea ice model (44, 45).

**ACKNOWLEDGMENTS.** We thank David Dilcher for his continuous and stimulating interest in this work, and Hans Renssen and Hughes Gooose for their invaluable advice with calculating radiative forcing and temperature. We also appreciate the insightful comments of two anonymous reviewers. This study was supported by the Council for Earth and Life Sciences of the Netherlands Organization for Scientific Research. This work is Netherlands Research School of Sedimentary Geology publication no. 2008.09.02.

- Esper J, Cook ER, Schweingruber FH (2002) Low-frequency signals in long tree-ring chronologies for reconstructing past temperature variability. *Science* 295:2250–2253.
- Cook ER, Esper J, D'Arrigo RD (2004) Extra-tropical Northern Hemisphere land temperature variability over the past 1000 years. *Quat Sci Rev* 23:2063–2074.
- Moberg A, Sonechkin DM, Holmgren K, Datsenko NM, Karlén W (2005) Highly variable Northern Hemisphere temperatures reconstructed from low- and high-resolution proxy data. *Nature* 433:613–617.
- Osborn TJ, Briffa KR (2006) The spatial extent of 20th-century warmth in the context of the past 1200 years. *Science* 311:831–834.
- Committee on Surface Temperature Reconstructions for the Past 2,000 Years, National Research Council (2006) *Surface Temperature Reconstructions for the Past 2,000 Years* (National Academies Press, Washington, DC).
- Jansen E, *et al.* (2007) *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, eds Solomon S, *et al.* (Cambridge Univ Press, Cambridge, UK), pp 433–497.
- Intergovernmental Panel on Climate Change (2001) *Climate Change 2001: Synthesis Report. A Contribution of Working Groups I, II, and III to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, eds Watson RT, *et al.* (Cambridge Univ Press, Cambridge, UK).
- Siegenthaler U, *et al.* (1988) Stable-isotope ratios and concentration of CO<sub>2</sub> in air from polar ice cores. *Ann Glaciol* 10:151–156.
- Barnola JM, *et al.* (1995) CO<sub>2</sub> evolution during the last millennium as recorded by Antarctic and Greenland ice. *Tellus* 47:264–272.
- Etheridge DM, *et al.* (1996) Natural and anthropogenic changes in atmospheric CO<sub>2</sub> over the last 1000 years from air in Antarctic ice and firn. *J Geophys Res* 101:4115–4128.
- Indermühle A, *et al.* (1999) Holocene carbon-cycle dynamics based on CO<sub>2</sub> trapped in ice at Taylor Dome, Antarctica. *Nature* 398:121–126.
- Siegenthaler U, *et al.* (2005) Supporting evidence from the EPICA Dronning Maud Land ice core for atmospheric CO<sub>2</sub> changes during the past millennium. *Tellus* 57B:51–57.
- Ruddimann WF (2003) The anthropogenic greenhouse era began thousands of years ago. *Clim Change* 61:261–293.
- Ruddimann WF (2007) The early anthropogenic hypothesis: Challenges and responses. *Rev Geophys* 45:RG4001.
- Bard E, Raisbeck G, Yiou F, Jouzel J (2000) Solar irradiance during the last 1200 years based on cosmogenic nuclides. *Tellus* 52B:985–992.
- Crowley TJ (2000) Causes of climate change over the past 1000 years. *Science* 289:270–277.
- Bauer E, Claussen M, Brovkin V (2003) Assessing climate forcings of the Earth system for the past millennium. *Geophys Res Lett* 30:1–4.
- Bradley RS, Briffa KR, Cole JE, Hughes MK, Osborn TJ (2003) *Paleoclimate, Global Change and the Future*, eds Alvenson K, Bradley RS, Pedersen TF (Springer, Berlin), pp 105–141.
- Gray JE, *et al.* (2000) The HIC signalling pathway links CO<sub>2</sub> perception to stomatal development. *Nature* 408:713–715.
- Wagner F, *et al.* (1996) A natural experiment on plant acclimation: Lifetime stomatal frequency response of an individual tree to annual atmospheric CO<sub>2</sub> increase. *Proc Natl Acad Sci USA* 93:11705–11708.

21. Kouwenberg LLR, et al. (2003) Stomatal frequency adjustment of four conifer species to historical changes in atmospheric CO<sub>2</sub>. *Am J Bot* 90:610–619.
22. Wagner F, Dilcher DL, Visscher H (2005) Stomatal frequency responses in hardwood-swamp vegetation from Florida during a 60-year continuous CO<sub>2</sub> increase. *Am J Bot* 92:690–695.
23. van Hoof TB, Kürschner WM, Wagner F, Visscher H (2006) Stomatal index response of *Quercus robur* and *Quercus petraea* to the anthropogenic atmospheric CO<sub>2</sub> increase. *Plant Ecol* 183:237–243.
24. Garcia-Amorena I, Wagner F, van Hoof TB, Gomez Manzoneque F (2006) Monitoring the stomatal response to the anthropogenic CO<sub>2</sub> increase in the southern European realm. *Rev Palaeobot Palynol* 141:303–312.
25. Wagner F, et al. (1999) Century-scale shifts in Early Holocene CO<sub>2</sub> concentration. *Science* 284:1971–1973.
26. Wagner F, Aaby B, Visscher H (2002) Rapid atmospheric CO<sub>2</sub> changes associated with the 8200-years-B.P. cooling event. *Proc Natl Acad Sci USA* 99:12011–12014.
27. Wagner F, Kouwenberg LLR, van Hoof TB, Visscher H (2004) Reproducibility of Holocene atmospheric CO<sub>2</sub> records based on stomatal frequency. *Quat Sci Rev* 23:1947–1954.
28. Jessen CA, Rundgren M, Björck S, Muscheler R (2007) Climate forced CO<sub>2</sub> variability in the early Holocene: A stomatal frequency reconstruction. *Glob Planet Change* 57:247–260.
29. Kouwenberg LLR, Wagner F, Kürschner WM, Visscher H (2005) Atmospheric CO<sub>2</sub> fluctuations during the last millennium reconstructed by stomatal frequency analysis of *Tsuga heterophylla* needles. *Geology* 33:33–36.
30. van Hoof TB, Bunnik FPM, Waucomont JGM, Kürschner WM, Visscher H (2006) Forest re-growth on medieval farmland after the Black Death pandemic: Implications for atmospheric CO<sub>2</sub> levels. *Palaeogeogr Palaeoclim Palaeoecol* 237:396–411.
31. Trudinger CM, Rayner PJ, Enting IG, Heimann M, Scholze M (2003) Implications of ice core smoothing for inferring CO<sub>2</sub> flux variability. *J Geophys Res Atmos* 108(D16):4492.
32. van Hoof TB, et al. (2005) Atmospheric CO<sub>2</sub> during the 13th century AD: Reconciliation of data from ice core measurements and stomatal frequency analysis. *Tellus* 57B:351–355.
33. Gerber S, et al. (2003) Constraining temperature variations over the last millennium by comparing simulated and observed atmospheric CO<sub>2</sub>. *Clim Dynam* 20:281–299.
34. Joos F, Gerber S, Prentice IC, Otto-Bliesner BL, Valdes PJ (2004) Transient simulations of Holocene atmospheric carbon dioxide and terrestrial carbon since the Last Glacial Maximum. *Glob Biogeochem Cycles* 18:doi:10.1029/2003GB002156.
35. Oppenheimer C (2003) Ice core and palaeoclimatic evidence for the timing and nature of the great mid-13th century volcanic eruption. *Int J Clim* 23:417–426.
36. deMenocal P, Ortiz J, Guilderson T, Sarntheim M (2000) Coherent high- and low-latitude climate variability during the Holocene Warm Period. *Science* 288:2198–2202.
37. Cronin TM, Dwyer GS, Kamiya T, Schwede S, Willard DA (2003) Medieval Warm Period, Little Ice Age and 20th century temperature variability from Chesapeake Bay. *Glob Planet Change* 36:17–29.
38. Eiriksson J, et al. (2006) Variability of the North Atlantic Current during the last 2000 years based on shelf bottom water and sea surface temperatures along an open ocean shallow marine transect in western Europe. *Holocene* 16:1017–1029.
39. Poole I, Kürschner WM (1999) *Fossil Plants and Spores: Modern Techniques*, eds Jones TP, Rowe NP (Geol Soc, London), pp 257–260.
40. Royer DL (2001) Stomatal density and stomatal index as indicators of paleoatmospheric CO<sub>2</sub> concentration. *Rev Palaeobot Palynol* 114:1–28.
41. Salisbury EJ (1927) On the causes and ecological significance of stomatal frequency, with special reference to the woodland flora. *Phil Trans R Soc London B* 216:1–65.
42. Myhre G, Highwood EK, Shine KP, Stordal F (1998) New estimates of radiative forcing due to well mixed greenhouse gases. *Geophys Res Lett* 25:2715–2718.
43. Forster P, et al. (2007) *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, eds Solomon S, et al. (Cambridge Univ Press, Cambridge, UK), pp 129–234.
44. Opsteegh JD, Haarsma RJ, Selten FM, Kattenberg A (1998) ECBILT: A dynamic alternative to mixed boundary conditions in ocean models. *Tellus* 50A:348–367.
45. Goosse H, Fichefet T (1999) Importance of ice-ocean interactions for the global ocean circulation: A model study. *J Geophys Res* 104:23337–23355.