water depressions where accumulation of enriched water may provide a permanent threat to the environment. Furthermore, if injected CO_2 is carried towards deep-water sediments rich in $CaCO_3$, dissolution of these sediments may provide efficient long-term sequestration of CO_2 as well as buffering of pH[19].

One possible application area could be along the west coast of Norway around 60-62° N, where water depths of 300-400 m are accessible close to the coast, and where bottom topography is sloping towards deep water in the Norwegian Sea. Furthermore, mean currents in this area are directed towards the deep ocean and the stratification is very weak below about 100 m (ref. 20) allowing easy vertical sinking of enriched water.

In other locations, strong stratification and unfavourable large-scale currents may inhibit vertical sinking and eventually lead to outgassing on a timescale of years to decades. Strong eddies or coastal upwelling may also contribute to dilute and lift CO_2 to the ocean surface in a matter of days or weeks after injection. But in many regions a starting depth of 200-400 m will be sufficient to avoid strong near-surface disturbances and ensure that the remaining ambient stratification can be penetrated by the dense CO_2-enriched sea water.

In general, pH levels of 5 or less occur when C_T is high enough to give an appreciable density effect (Fig. 1 and equation (1)). Marine life within the area swept by the sinking bottom gravity current might therefore be severely affected. The associated temperature increase is believed to be of secondary importance. Formation of hydrates, which might create operational problems in the case of deep injection and which would sink and collect at the ocean bottom as a marine waste dump, is expected to be reduced or eliminated in the case of shallow injection (Fig. 2).

Certainly one should not recommend injection of CO_2 in the ocean to slow down the increase in greenhouse warming unless the above theoretical predictions are confirmed by experimental investigations and further analyses, and the environmental impact downstream of the injection point is deemed to be acceptable. But the chemical carrying capacity of the ocean is formidable compared with anthropogenic emissions. If shallow injection is working as indicated by these preliminary investigations, at least in some locations, it may contribute to obtaining the required reduction in net emissions to the atmosphere in the future. ☐

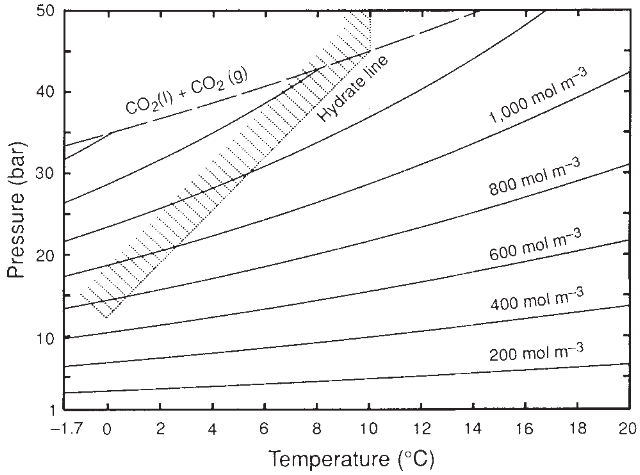

FIG. 2 *Solubility of CO_2 in sea water.* The number of moles of CO_2 that may be dissolved in one m^3 of sea water with salinity 35‰ (contoured), phase line between stable gaseous and liquid phase of pure CO_2 (dashed), and rough limit of conditions susceptible to hydrate formation (hatched). The pressure scale is in bar$=10^5$ Pa$=0.9869233$ atm; hydrostatic pressure incrase in water is about 1 bar for each 10 m water depth. Maximum solubility has been calculated from a modified Henry's law, with temperature- and salinity-dependent Henry's law constant from ref. 24 and fugacity from refs 8, 25. Phase line from ref. 26. Rough limit of hydrate formation from data for pure water is discussed in ref. 8 and agrees with ref. 27.

Received 13 January; accepted 6 April 1992.

1. Marchetti, C. *Clim. Change* **1,** 59-68 (1977).
2. Baes, C. F. Jr *et al.* in *Interactions of Energy and Climate* (eds Bach, W., Pankrath, J. & Williams, J.) 495-519 (Reidel, Dordrecht, 1980).
3. Herzog, H., Golomb, D. & Zemba, S. *Env. Prog.* **10,** 64-74 (1991).
4. Bacastow, R. & Stegen, G. R. in *Proc. Oceans '91* Vol. 3, 1654-1657 (IEEE, Honolulu, Hawaii, 1991).
5. Stegen, G. R., Cole, K. H. & Bacastow, R. in *Proc. IEA Int. Conf. Technology Responses to Global Environmental Challenges* (IEA, Kyoto, Japan, 1991).
6. Maier-Reimer, E. in *Strategies for Future Climate Research* (ed. M. Latif) 319-339 (Max-Planck-Institut für Meteorologie, Hamburg, 1991).
7. Watanabe, H. & Iizuka, K. *Metrologia* **21,** 19-26 (1985).
8. King, M. B. *Phase Equilibrium in Mixtures* (Pergamon, Oxford, 1969).
9. UNESCO *Technical Papers in Marine Science* **36** (UNESCO, Paris, 1981).
10. Smith, P. C. *Deep Sea Res.* **22,** 853-873 (1975).
11. Chu, P. C. & Gascard, J.-C. (ed.) *Deep Convection and Deep Water Formation in the Oceans* (Elsevier, Amsterdam, 1991).
12. Clift, R., Grace, J. R. & Weber, M. E. *Bubbles, Drops and Particles* (Academic, New York, 1978).
13. Tse, F. C. & Sandall, O. C. *Chem. Engng. Commun.* **3,** 147-153 (1979).
14. Calderbank, P. H., Johnson, D. S. L. & Loudon, J. *Chem. Engng. Sci.* **25,** 235-256 (1970).
15. Grace, J. R., Wairegi, T. & Brophy, J. *Can. J. Chem. Engng.* **56,** 3-8 (1978).
16. Killworth, P. D. *Deep Sea Res.* **24,** 427-448 (1977).
17. Turner, J. S. *Buoyancy Effects in Fluids* (Cambridge Univ. Press, 1973).
18. Turner, J. S. in *The Evolution of Physical Oceanography* (ed. Warren, B. A. & Wunsch, C.) 236-262 (MIT Press, Cambridge, Massachusetts, 1981).
19. Stumm, W. & Morgan, J. J. *Aquatic Chemistry* 2nd edn (Wiley, New York, 1981).
20. Otto, L. *et al. Neth. J. Sea Res.* **26,** 161-238 (1990).
21. Bishnoi, P. R. & Robinson, D. B. *Can. J. Chem. Engng.* **49,** 642-650 (1971).
22. Cheney, W. & Kincaid, D. *Numerical Mathematics and Computing* (Brooks/Cole, Monterey, California, 1985).
23. UNESCO *Technical Papers in Marine Science* **51** (UNESCO, Paris, 1987).
24. Weiss, R. F. *Mar. Chem.* **2,** 203-215 (1974).
25. Levine, I. N. *Physical Chemistry* (McGraw-Hill, New York, 1988).
26. CRC *Handbook of Chemistry and Physics* 68*th* Edn (ed. Weast, R. C.) (CRC Press, Boca Raton, Florida, 1987).
27. Sakai, H. *et al. Science* **248,** 1093-1096 (1990).

ACKNOWLEDGEMENTS. We acknowledge financial support from Den norske stats oljeselskap (Statoil).

Possible methane-induced polar warming in the early Eocene

L. Cirbus Sloan, James C. G. Walker, T. C. Moore Jr, David K. Rea & James C. Zachos

Department of Geological Sciences, University of Michigan, Ann Arbor, Michigan 48109-1063, USA

RECONSTRUCTIONS of early Eocene climate depict a world in which the polar environments support mammals and reptiles, deciduous forests, warm oceans and rare frost conditions[1–5]. At the same time, tropical sea surface temperatures are interpreted to have been the same as or slightly cooler than present values[6]. The question of how to warm polar regions of Earth without noticeably warming the tropics remains unresolved; increased amounts of greenhouse gases would be expected to warm all latitudes equally[7]. Oceanic heat transport has been postulated as a mechanism for heating high latitudes[8–10], but it is difficult to explain the dynamics that would achieve this[7,11]. Here we consider estimates of Eocene wetland areas and suggest that the flux of methane, an important greenhouse gas, may have been substantially greater during the Eocene than at present. Elevated methane concentrations would have enhanced early Eocene global warming, and also might specifically have prevented severe winter cooling of polar regions because of the potential of atmospheric methane to promote the formation of optically thick, polar stratospheric ice clouds[12–14].

Methane is produced primarily by anaerobic bacteria. Major sources of methane are swamps and wetlands, rice paddies, termites and enteric fermentation by ruminants[15-17]. For the early Eocene, anthropogenic and ruminant contributions of methane can be discounted, increasing relative contributions from wetland and forest ecosystems. There were large areas of swamps and wetlands in the late Palaeocene and early to middle Eocene, at least in part related to the long-term rise in sea level

(Fig. 1). Existing 'coaly continental clastics' of Palaeocene-Eocene age are estimated to cover an area of 2.8×10^6 km^2 (ref. 18). These deposits are located in continental interior and coastal environments at various latitudes[18]. If one assumes, as a conservative estimate, that coaly deposits of a given age represent a minimum of half the total wetland environments existing at that time, the area of wetlands possible in the Palaeocene-Eocene would have been at least 5.6×10^6 km^2, compared with 2×10^6 km^2 for the present[16]. Thus during the Palaeocene-Eocene there would be the potential to triple modern methane production from wetland ecosystems alone. Additionally, there were probably more extensive areas of forest environments during the Eocene[3,4,19] relative to present distributions, which also could have contributed to increased methane production[16].

Quantitative estimates of atmospheric methane concentrations in the geological past are available only for the time recorded in ice. Thus, our hypothesis of greater methane concentrations is speculative, but is supported indirectly by a variety of evidence. We suggest that the long-term sea level rise in the late Palaeocene and early Eocene (Fig. 1) and the associated development of large areas of swamps and wetlands, along with greater areas of humid environments at this time[20], provided the environments for anaerobic decay of organic matter, producing increased methane flux to the atmosphere.

Sequestering of large quantities of organic, isotopically light carbon into sedimentary reservoirs should produce significant changes in global ocean ^{13}C composition (Fig. 1). The organic carbon could have been the substrate used by methanogenic bacteria to produce elevated methane fluxes. Although the relationship between late Palaeocene/early Eocene marine ^{13}C change, the carbon cycle and climate is unclear at this time, it seems that approximately concurrent with maximum ^{13}C values and the ensuing shift back towards more negative values, oxygen isotope ratios measured in benthic foraminifera shift to more negative values, indicating warmer global conditions (Fig. 2).

Methane is a powerful greenhouse gas[15,17]. Together with enhanced carbon dioxide concentrations it could have contributed to a generally warmer troposphere and cooler lower stratosphere[21]. In addition, methane oxidation is a significant source of stratospheric water vapour[15,17,22]. A wetter and colder lower stratosphere could have yielded polar stratospheric clouds which were optically thicker, covered a greater area and were more frequently occurring than those of today[12-14,23].

Optically thick polar stratospheric clouds warm the troposphere by trapping long-wave radiation from the Earth, thus preventing cooling of Earth's surface[12-14,24,25]. Their albedo effect on short-wave radiation is obviously small during polar winter. The net effect on the radiation balance is high-latitude warming (at present up to 0.5 K per day[13]), precisely what is needed to explain Eocene climate observations. We suggest that with a substantial methane increase and a large enough radiative effect, optically thicker, and more frequent polar stratospheric clouds with a more extensive area could potentially have kept winter polar surface temperatures above freezing, and dampened the intra-annual temperature variation at high latitudes by preventing strong winter cooling. This condition could potentially have affected Eocene deepwater structure and circulation through an influence on high-latitude surface water minimum temperatures and therefore bottom water formation[3].

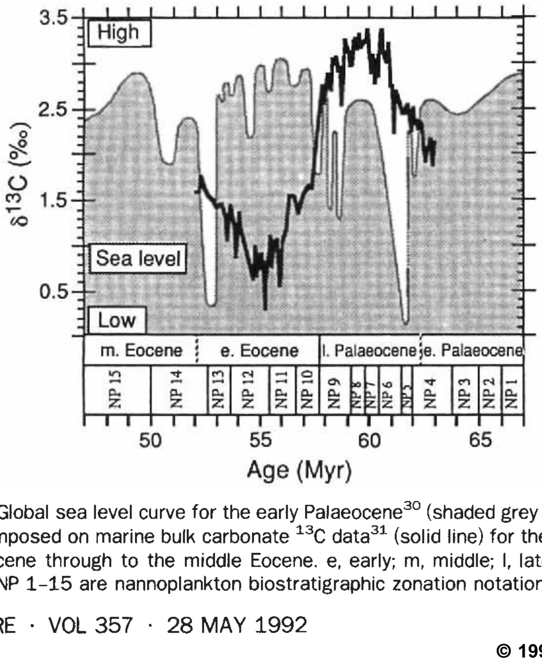

FIG. 1 Global sea level curve for the early Palaeocene[30] (shaded grey curve) superimposed on marine bulk carbonate ^{13}C data[31] (solid line) for the early Palaeocene through to the middle Eocene. e, early; m, middle; l, late. The labels NP 1–15 are nannoplankton biostratigraphic zonation notation.

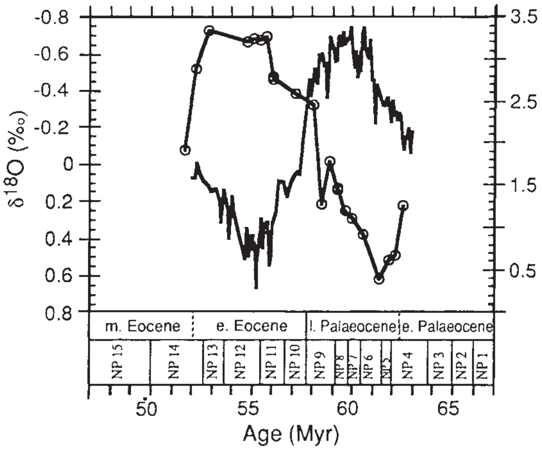

FIG. 2 Marine bulk carbonate ^{13}C data[31] (solid line) and global oxygen curve from benthic foraminifera[32] (line with circles) over the same time interval as shown in Fig. 1. e, early; m, middle; l, late.

A methane polar stratospheric cloud system during the Eocene could have worked in conjunction with high atmospheric carbon dioxide concentration, for which there is some evidence (refs 26, 27, and K. Freeman and P. Hayes, manuscript in preparation). The response of biogenic methane sources to surface temperature changes are not well known[24], but warm temperatures could have enhanced methane production rates, providing a positive feedback to the system.

The photochemistry of ancient atmospheres may have been significantly different from now. The unusual climate of the Eocene may have been caused not just by enhanced greenhouse effects and modified oceanic circulation, but by enhanced methane concentrations as well. But if interpretations of early Eocene tropical sea surface temperatures[6] are correct, then the enhanced greenhouse effect, at least in the tropics, would have had to be minor. Clearer interpretations of latitudinal surface temperature values may lead to understanding the heirarchy of the causal mechanisms for climate at this time. To describe more fully the mechanism outlined here we need more comprehensive Palaeocene-Eocene reconstructions of environments and more quantitative estimates of methane fluxes. We must also consider the interaction of atmospheric methane with other trace gases and other parts of the Palaeocene-Eocene climate system. The methane polar stratospheric cloud mechanism may also be applicable to other times when polar environments were warm, such as the Cretaceous[28,29] and the Pliocene[7]. ☐

Received 3 January; accepted 6 April 1992.

1. Estes, R. & Hutchison, J. H. *Palaeogeogr. Palaeoclimatol Palaeoecol.* **30**, 325-347 (1980).
2. Schweitzer, H.-J. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **30**, 297-311 (1980).
3. Kennett, J. P. & Stott, L. D. *Nature* **353**, 225-229 (1991).
4. Kemp, E. M. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **24**, 169-208 (1978).
5. Case, J. A., in *Geology and Palaeontology of Seymour Island, Antarctic Peninsula* (eds Feldmann, R. M. & Woodburne, M. O.) 523-530 (Geol. Soc. Am., Boulder, Colorado, 1988).
6. Shackleton, N. J. & Boersma, A. *Geol. Soc. Lond. J.* **138**, 153-157 (1981).
7. Crowley, T. J. *Quat. Sci. Rev.* **10**, 275-282. (1991).

8. Barron, E. J. *Paleoceanography* **2,** 729–739 (1987).
9. Rind, D. & Chandler, M. *J. geophys. Res.* **96,** 7437–7461 (1991).
10. Crowley, T. J. *Paleoceanography* **6,** 387–394 (1991).
11. Sloan, L. C. & Walker, J. C. G. *Geophys. Res. Lett.* (submitted).
12. Pollack, J. B. & McKay, C. P. *J. atmos. Sci.* **42,** 245–262 (1985).
13. Kinne, S. & Toon, O. B. *Geophys. Res. Lett.* **17,** 373–376 (1990).
14. McCormick, M. P., Trepte, C. R. & Pitts, M. C. *J. geophys. Res.* **94,** 11241–11251 (1989).
15. World Meteorological Organization/United Nations Environment Programme *Climate Change, The IPCC Scientific Assessment* (eds Houghton, J. T., Jenkins, G. J. & Ephraums, J. J.) (Cambridge Univ. Press, 1990).
16. Sheppard, J. C., Westberg, H., Hopper, J. F., Ganesan, K. & Zimmerman, P. *J. geophys. Res.* **87,** 1305–1312 (1982).
17. Cicerone, R. J. & Oremland, R. S. *Glob. Biogeochem. Cycles* **2,** 299–327 (1988).
18. Ronov, A. B., Khain, V. Y. & Balukhovskiy, A. N. *Int. Geol. Rev.* **21,** 415–446 (1979).
19. Askin, R. A. in *Geology and Paleontology of Seymour Island, Antarctic Peninsula* (eds Feldmann, R. M. & Woodburne, M. O.) 131–135 (Geol. Soc. Am., Boulder, Colorado, 1988).
20. Sloan, L. C. thesis, Pennsylvania State Univ. (1990).
21. Rasmussen, R. A. & Kahlil, M. A. K. *J. geophys. Res.* **86,** 9826–9832 (1981).
22. Rinsland, C. P. *et al. J. geophys. Res.* **89,** 7259–7256 (1984).
23. McCormick, M. P. & Trepte, C. R. *J. geophys. Res.* **92,** 4297–4306 (1987).
24. Ramanathan, V. *et al. Rev. Geophys.* **25,** 1441–1482 (1987).
25. Ramanathan, V. *et al. Science* **243,** 57–59 (1989).
26. Arthur, M. A., Allard, D. & Hinga, K. R. *Geol. Soc. Am. Prog.* Vol. 23, 178 (1992).
27. Berner, R. *Am. J. Sci.* **291,** 339–376 (1991).
28. Barron, E. J. & Washington, W. M. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **40,** 103–133 (1982).
29. Wolfe, J. A. & Upchurch, G. R. Jr *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **61,** 33–77 (1987).
30. Haq, B. U., Hardenbol., J. & Vail, P. R. *Science* **235,** 1156–1167 (1987).
31. Shackleton, N. J., Hall, M. A. & Boersma, A. *Init. Rep. Deep Sea Drilling Proj.* **74,** 599–612 (US Govt Printing Office, Washington DC, 1984).
32. Miller, K. G., Janacek, T. R., Katz, M. E. & Keil, D. J. *Paleoceanography* **2,** 741–761 (1987).

Productivity and compensatory responses of yellow-poplar trees in elevated CO_2

Richard J. Norby, Carla A. Gunderson,
Stan D. Wullschleger, E. G. O'Neill
& Mary K. McCracken

Environmental Sciences Division, Oak Ridge National Laboratory,
PO Box 2008, Oak Ridge, Tennessee 37831-6034, USA

INCREASED forest growth in response to globally rising CO_2 concentrations could provide an additional sink for the excess carbon added to the atmosphere from fossil fuels[1,2]. The response of trees to increased CO_2, however, can be expected to be modified by the interactions of other environmental resources and stresses, higher-order ecological interactions and internal feedbacks inherent in the growth of large, perennial organisms[3,4]. To test whether short-term stimulation of tree growth by elevated CO_2 can be sustained without inputs from other environmental resources, we grew yellow-poplar (*Liriodendron tulipifera* L.) saplings for most of three growing seasons with continuous exposure to ambient or elevated concentrations of atmospheric CO_2. Despite a sustained increase in leaf-level photosynthesis and lower rates of foliar respiration in CO_2-enriched trees, whole-plant carbon storage did not increase. The absence of a significant growth response is explained by changes in carbon allocation patterns, specifically a relative decrease in leaf production and an increase in fine root production. Although these compensatory responses reduced the potential increase in carbon storage in increased CO_2 concentrations, they also favour the efficient use of resources over the longer term.

Yellow-poplar trees were grown in ambient or elevated CO_2 concentrations in open-top chambers[5]. Open-top chambers allowed the experimental plants, which before outplanting had been grown from seed under the experimental CO_2 concentrations, to be exposed in the field under ambient light, temperature, precipitation and soil conditions. The CO_2 concentration in the six chambers was maintained day and night during the growing seasons at ambient (354.5 μmol mol^{-1}), ambient + 150 (502.9 μmol mol^{-1}) or ambient + 300 (655.6 μmol mol^{-1}). No fertilizer or supplemental water was provided during the course of the experiment. Five yellow-poplar saplings were harvested from each chamber after 28 months (2.7 growing season).

TABLE 1 Characteristics of yellow-poplar leaves in 1991

CO_2 enrichment (μmol mol^{-1})	Specific leaf area (cm^2 g^{-1})	Nitrogen (g m^{-2})	Chlorophyll (mg m^{-2})	Photo-synthesis (μmol m^{-2} s^{-1})	Respiration (μmol m^{-2} s^{-1})
+0	151 ± 6	1.52 ± 0.09	488 ± 54	7.4 ± 0.6	3.0 ± 0.2
+150	136 ± 5	1.30 ± 0.13	401 ± 82	10.8 ± 0.9	2.3 ± 0.2
+300	132 ± 5	1.18 ± 0.05	387 ± 32	12.3 ± 0.8	2.2 ± 0.1
*P**	0.004	0.058	0.20	0.001	0.001

All data are means ±s.e. of five plants in each of two replicate chambers. Specific leaf area was determined at final harvest from total leaf area and total leaf mass. Nitrogen concentration was measured by near-infrared reflectance spectroscopy of oven-dried and ground leaves from leaf position 3-5 collected on 2 July. Chlorophyll concentrations were measured on 25 July in ethanol extracts of five 6-mm-diameter disks cut from one leaf per tree. Photosynthesis was measured with a closed gas-exchange system (LiCor 6200) on the fourth leaf from an upper branch apex. Measurements were made at midday under full sun on 1 July. Respiration was measured with the gas exchange system before sunrise on 5 July.

* Probability of a significant effect of CO_2 concentration, using sampling error in the *F*-test.

The accumulation of above-ground biomass was similar in all treatments throughout the course of the experiment, and although stem mass was usually lowest in ambient CO_2, there were no differences in stem mass that were statistically significant (Fig. 1*a*). Leaf area tended to decline with increasing CO_2 concentration in 1990 and 1991, and combining the stem growth and leaf area data provided a better (less variable, more robust) indicator of the response of above-ground productivity to CO_2 enrichment (Fig. 1*b*). Growth efficiency, the annual production of stem mass per unit leaf area, is akin to the term net assimilation rate used with herbaceous plants and is a good indicator of general tree vigour[6]. Growth efficiency in 1990 was 41% greater

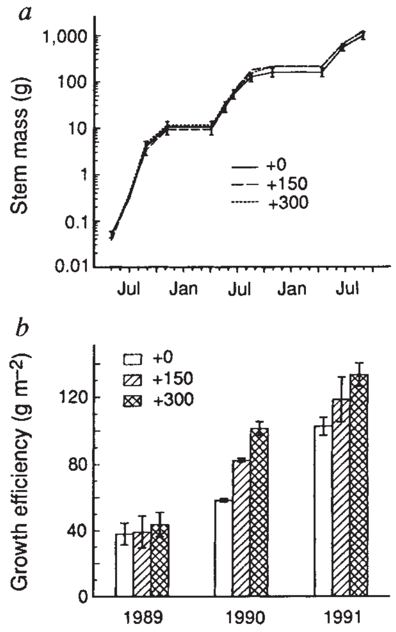

FIG. 1 Stem mass and growth efficiency of yellow-poplar saplings between May 1989 and August 1991 in ambient (+0), ambient +150, and ambient +300 μmol CO_2 mol^{-1}. All data are the means ±s.e. of five trees per chamber in two replicate chambers. *a*, Stem dry mass was estimated from periodic measurements of bole diameter (*D*) 10 cm from the ground and total height (*H*) of the bole, using a log–log regression between stem mass and D^2H. *b*, Growth efficiency was calculated as annual stem mass increment divided by leaf area at the end of the year. The effect of CO_2 on growth efficiency was significant at $P = 0.041$ in 1990, and $P = 0.002$ in 1991.