Terrestrial record of methane hydrate dissociation in the Early Cretaceous

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ABSTRACT

Reconstruction of changing C isotopic composition of Early Cretaceous atmospheric CO_2 from fossilized C3 vascular land-plant tissue revealed a brief and striking negative excursion ($\Delta \approx -5‰$) in atmospheric $\delta^{13}C$, followed by a rapid positive compensation ($\Delta \approx +5‰$) during the Aptian (ca. 117 Ma). Mass-balance calculations show that dissociation of a small amount of methane gas hydrate is the most tenable cause of the negative excursion; this would also result in an increased CO_2:O_2 mixing ratio as O_2 is consumed during CH_4 oxidation to CO_2, spurring the exponential phase of angiosperm biogeographic expansion.

Keywords: carbon-13, excursions, angiosperms, methane, Aptian.

INTRODUCTION

The total present-day mass of carbon stored as methane hydrate is thought to be large (11 000 Gt of C) (1 Gt C = 10^{15} g C) and very ^{13}C depleted ($\delta^{13}C \approx -60‰$) (Kvenvolden, 1993). Processes as diverse as gradual pressure increase due to sedimentary burial, submarine slope failure, sea-level regression, or increase in bottom water temperatures could induce rapid liberation of CH_4 (Kvenvolden, 1993). The carbon isotope signature of methane dissociation into local seawater and ultimately to the atmosphere would be reflected in the $\delta^{13}C$ of the actively cycling carbon pools.

A methane release from ocean hydrates at the late Paleocene thermal maximum (55.6 Ma) has been inferred from a negative excursion ($\Delta = -2.5‰$) in the marine carbonate record (Dickens et al., 1995, 1997). This anomaly was correlated to a negative excursion of similar magnitude in the terrestrial record; i.e., soil carbonate $\delta^{13}C$ (Koch et al., 1995) and terrestrial organic matter $\delta^{13}C$ (Stott et al., 1996). Recent work has confirmed that the global biogeochemical response to such an event is extremely rapid (Norris and Rohl, 1999), and changes in the $\delta^{13}C$ of atmospheric CO_2 have been shown to reflect source-specific changes in global carbon cycling (Peng et al., 1983). Plant compression fossils are useful isotopic substrates because land plants sample the atmosphere directly and in a predictable way during photosynthetic carbon fixation. Our work applies a recently developed relationship between the $\delta^{13}C$ of C3 vascular land plant tissue and the $\delta^{13}C$ of atmospheric CO_2 (Arens et al., 2000) to estimate changes in the $\delta^{13}C$ of atmospheric CO_2 through the Early Cretaceous and discusses implications for biogeochemical cycling in the early Aptian (ca. 117 Ma).

SAMPLES AND METHODS

Organic material from estuarine and nearshore terrestrial sediments of Early Cretaceous age at three localities in the Cordillera Oriental of the Colombian Andes, South America, was isolated and analyzed for C isotope ratios. The three localities are within 100 km of one another and constitute ~2250 m of section within the same biostratigraphic province. Units sampled include the Une, Fómeque, and Las Juntas Formations, which encompass an 18 m.y. interval during the Early Cretaceous with no evidence of depositional disconformity. Age dates were based on biostratigraphic correlation with well-established dinoflagellate and ammonite zones (Guerrero and Sarmiento, 1996). All samples were analyzed for $\delta^{13}C$ of bulk organic material, and only strata bearing abundant land-plant macrofossils were analyzed. For bulk organic material determinations, a 1–5 g sample of the rock was subsampled and immersed in 1*M* HCl overnight to remove carbonate. The remaining bulk organic residue was analyzed for $\delta^{13}C$ in triplicate. To address the possible incorporation of any carbon derived from other than terrestrial productivity, we isolated two components specific to vascular land plants: vitrinite and cuticle. Vitrinite is the coalified remains of highly lignified plant tissues, primarily xylem; cuticle is a waxy compound that coats plant epidermal surfaces. Both materials are restricted to vascular land plants and represent uniquely terrestrial carbon compounds that can be recognized at low magnification. Vitrinite and cuticle isolates were collected and prepared directly from abundant fern and cycad fossils when possible; when no suitable fossils were available, rock was acid macerated (HF and HCl), rinsed, and handpicked. In this case, isolated tracheids and cuticle could not be identified to taxon. Vitrinite was isolated and manually separated via its color, texture, and fracture in rock hand specimens; it was then soaked in 60% HF overnight to remove any inorganic components. In some cases, clumps of tracheids were isolated from rock macerate after HF treatment. Cuticle was isolated after exposure to 60% HF via manual separation at 5× magnification; cuticle morphology was verified under 40× magnification. Vitrinite and cuticle C isotope determinations were made in duplicate and triplicate when sufficient material could be isolated.

All organic carbon samples were prepared for stable isotope analysis via combustion in sealed tubes containing Cu, CuO, and Ag. Released CO_2 was purified and collected for $^{13}C/^{12}C$ measurement on the mass spectrometer. All isotope values are reported in delta notation relative to the Vienna Peedee belemnite standard (VPDB).

RESULTS

The C isotope data from the sections are presented in Figure 1A. Bulk $\delta^{13}C$ values range from −22‰ to −24‰ during the late Valanginian, through the Hauterivian and early Barremian. Values increase in the late Barremian ($\delta^{13}C \approx -21‰$), followed by a dramatic decrease to ≈−28‰ in the early Aptian, which is followed by a sharp increase to ≈−21‰ in the mid-Aptian. There is no evidence for depositional discontinuities in this part of the section, and the samples representative of the dramatic excursion compose ~100 m of sediment (application of average sedimentation rates for the Fómeque Formation suggest that this event was <1 m.y. in duration). Multiple studies have shown that decomposition does not usually alter the $\delta^{13}C$ of plant tissues by more than 2‰; according-

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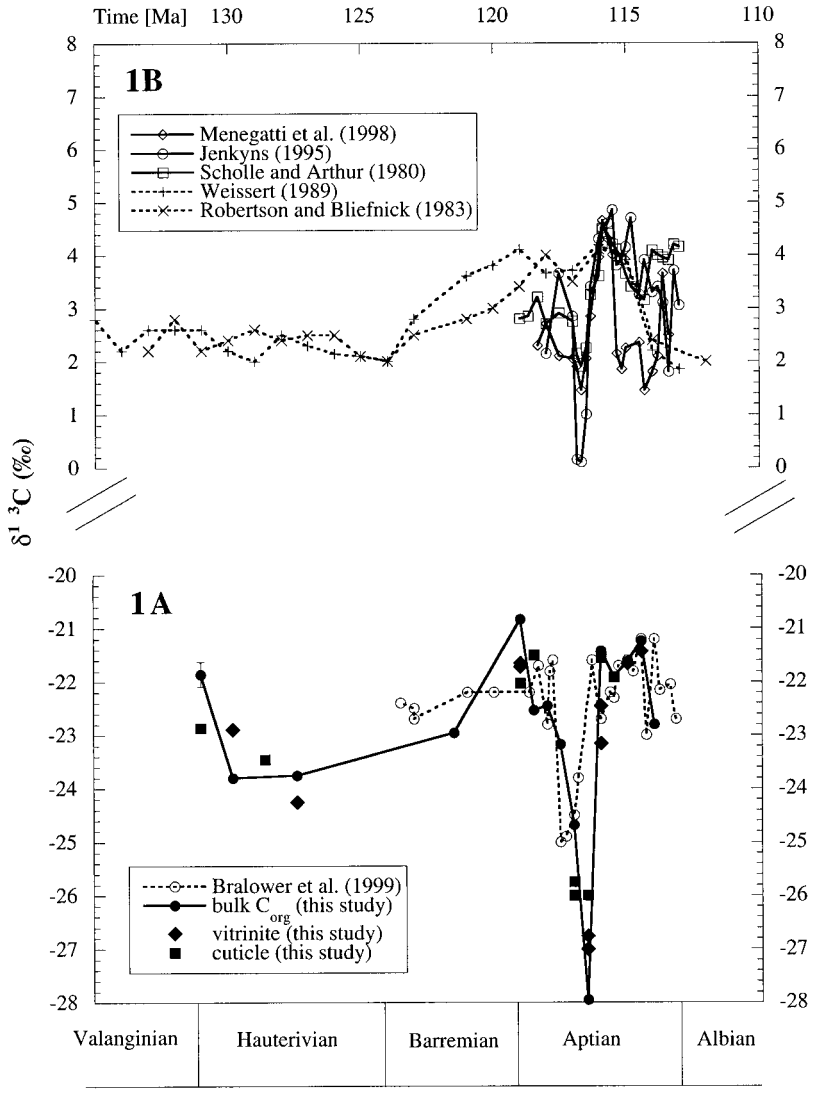

Figure 1. $\delta^{13}C$ data from Cretaceous: terrestrial and marine organic carbon (1A) and marine carbonate (1B) analyses. Overall time scale presented here is based on Harland et al. (1989); marine and terrestrial records cross-correlate to ≤1 m.y.

ly, the maximum difference seen between bulk organic matter $\delta^{13}C$ and lignin and cuticle isolate $\delta^{13}C$ approaches 2‰. The congruence of bulk organic, cuticle, and vitrinite $\delta^{13}C$ affirms that the bulk organic carbon signal is primarily derived from land-plant organic input. Alteration of organic constituents was evaluated using the thermal alteration scale (Batten, 1996); all constituents were within low-maturation categories 4 and 5, suggesting maximum burial of 3 km and maximum temperatures of 70 °C. Regression analysis of $\delta^{13}C$ against the maturation estimates showed no significant relationship, suggesting that patterns observed in $\delta^{13}C$ through time are not perturbed by differing thermal histories of the rocks. In addition, the similarity in degree of thermal maturity throughout the section supports the conclusion that patterns observed in $\delta^{13}C$ through time reflect genetic, rather than diagenetic, phenomena.

We reconstruct the $\delta^{13}C$ record of the atmosphere (Fig. 2) using the average $\delta^{13}C$ of strata within these terrestrial sediments, including isolated cuticle and vitrinite, components unique to land plants. Cretaceous ecosystems were dominated by C3 plants (Bocherens et al., 1993); isotopic fractionation during C3 photosynthesis is influenced by differential diffusion of $^{12}CO_2$ versus $^{13}CO_2$ in air, discrimination imparted by the primary carbon fixation enzyme, and ecophysiological factors that balance leaf carbon gain with water loss (Farquhar et al., 1989, reviewed *in* Arens et al., 2000). With notable exceptions (Gröcke et al., 1999), land plants have been overlooked as a source of information about the isotopic composition of atmospheric CO_2 because these ecophysiological vital effects were believed to swamp the atmospheric signal (reviewed in Farquhar et al., 1989). However, in an analysis of a large data set (519 published $\delta^{13}C_{plant}$ measurements on 176 C3 species) Arens et al. (2000) showed that the relationship between the $\delta^{13}C$ of plant tissue and atmospheric CO_2 is linear and significant ($r^2 = 0.91$ for the full data set). Using a subset of these data, Arens et al. (2000) proposed an empirically derived relationship ($\delta^{13}C_{atmosphere} = [\delta^{13}C_{plant} + 18.67]/1.10$) that could be used to estimate the $\delta^{13}C$ of atmospheric CO_2 from preserved terrestrial plant material. Arens et al. (2000) quantified the error due to vital effects for the large set of species and ecological conditions studied (calculated here as a 99% confidence interval). In actualistic tests to verify the predictive ability of this method, Arens et al. (2000) showed that for most mesic environments, the relationship predicted both ancient and modern atmospheric $\delta^{13}C$ within the defined confidence interval.

Our analyses of bulk organic carbon reflect an ecosystem average signal of rapidly buried land-plant organic material; palynology of these sediments indicates moderate palynospecies richness and does not show major changes in the taxonomic composition or relative abundance of plant groups during the interval studied (Arens, 1996). Therefore, the significant $\delta^{13}C$ excursions observed in this sequence cannot be explained by changing floristics that might add new physiologies or climate-selected ecologies into the material preserved.

DISCUSSION

The striking negative excursion ($\Delta \approx -5‰$) inferred in atmospheric CO_2 $\delta^{13}C$ in the early Aptian, followed by a rapid positive compensation ($\Delta \approx +5‰$) during the mid-Aptian (Fig. 2), suggests substantial disruption in global carbon cycling. Several sources of isotopically light carbon must be considered to identify the process underlying this atmospheric excursion: magmatically derived CO_2, volatilized organic carbon from standing or buried biomass, and methane. Cretaceous pCO_2 was modeled to be about four times present-day partial pressure (p) (Berner, 1990), suggesting an atmosphere containing ~2400 Gt of carbon. Using this assumption, the following source-mixing analysis was used to estimate the amount of carbon (n) required from each of the potential sources to drive the negative atmospheric excursion: $(2400 + n \text{ Gt C}) \cdot (\delta^{13}C_{atmosphere}$ early Aptian$) = 2400 \text{ Gt C} \cdot (\delta^{13}C_{atmosphere}$ late Barremian$) + n \text{ Gt C} \cdot (\delta^{13}C_{emission})$. These calculations (Table 1) indicate that the Aptian atmospheric excursion described here would require more than a 10-fold increase in atmospheric CO_2 with magmatically derived CO_2 ($\delta^{13}C \approx -8$; Taylor, 1986), making volcanic CO_2 an untenable carbon source, despite abundant evidence for enhanced volcanism during the Aptian. If volatilized organic carbon were the source of ^{13}C-depleted CO_2 ($\delta^{13}C \approx -25$; Peng et al.,

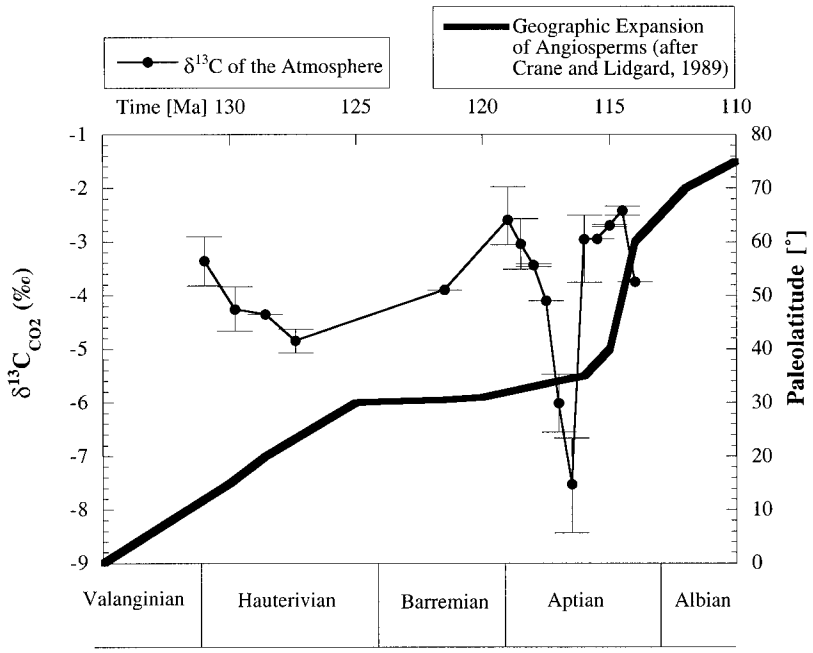

Figure 2. Calculated $\delta^{13}C$ of atmospheric CO_2 using $\delta^{13}C_{atmosphere} = (\delta^{13}C_{plant} + 18.67)/1.10$ ($r = 0.95$, $r^2 = 0.91$) for average bulk C3 vascular land-plant tissue (Arens et al., 2000). Calculations were made using average $\delta^{13}C$ for each sample (including cuticle and vitrinite isolates) presented in Figure 1A. Error bars encompass maximum and minimum predictions for sample given total variability seen in $\delta^{13}C$ of each sample. Overall time scale correlation is based on Harland et al. (1989).

1983), ~677 Gt of carbon would be required to produce the observed excursion, more than all the carbon stored in today's terrestrial vegetation (Sundquist, 1993). A rapid (<1 m.y.) volatilization of a significant portion of Aptian terrestrial biomass would result in major floristic and terrestrial facies change. Early Cretaceous flora was changing in response to the biogeographic expansion of angiosperms (discussed in the following) and the onset of the Cretaceous greenhouse (Crane and Lidgard, 1989), but there is no evidence of rapid or catastrophic tempo to these transitions, nor does the Aptian sedimentary record contain unusual amounts of charcoal (Chaloner and McElwain, 1997).

In contrast, a relatively small contribution of methane from methane hydrates could have produced the Aptian atmospheric excursion. A contribution to the atmosphere of only ~225 Gt of carbon from methane hydrate, equal to 2% of the present-day oceanic reservoir, could account for the excursion. Methane hydrate stability in sediment pore spaces partially depends on temperature, and warming could induce catastrophic liberation (Dickens et al., 1995). The first pulses of Cretaceous greenhouse warming began in the late Valanginian (~13 m.y. prior to the negative excursion shown in Fig. 2) (Lini et al., 1992), and may have contributed to the destabilization and subsequent liberation of accumulated methane hydrate.

TABLE 1. AMOUNT OF CARBON REQUIRED TO DRIVE THE ATMOSPHERIC EXCURSION

Magnitude of excursion (‰)	Emission required from volcanic sources (Gt)	Emission required from volatilized organic carbon (Gt)	Emission required from methane gas hydrate (Gt)
Average*			
−4.9	24 758#	677**	225††
Minimum†			
−3.6	6476§§	472##	162***
Maximum§			
−6.5	N.P.†††	937§§§	301###

* Calculated excursion is based on average $\delta^{13}C$ for each sample, including cuticle and vitrinite isolates.
† Minimum excursion based on variability seen in $\delta^{13}C$ for each sample, including cuticle and vitrinite isolates.
§ Maximum excursion based on variability seen in $\delta^{13}C$ for each sample, including cuticle and vitrinite isolates.
# More than a 10-fold increase in atmospheric CO_2 content.
** More than the total amount of carbon stored in living terrestrial plants.
†† 2.0% of total present-day mass of carbon stored as methane hydrate.
§§ More than a 3-fold increase in atmospheric CO_2 content.
77% of the total amount of carbon stored in living terrestrial plants.
*** 1.5% of total present-day mass of carbon stored as methane hydrate.
††† Not possible, given the assumed $\delta^{13}C$ of volcanic CO_2.
§§§ More than the total amount of carbon stored in living terrestrial plants.
1.5% of total present-day mass of carbon stored as methane hydrate.

Although the amount of carbon released from methane hydrates need only have been small (~9%) relative to the total mass of carbon in the preexisting Aptian atmosphere, the consequences for global biogeochemical cycling would have been extensive. To account for the negative isotopic excursion, ~300 Gt of CH_4 would have to be liberated to the Aptian atmosphere. Atmospheric methane inhibits the reradiation of terrestrial infrared radiation, and so an increase in CH_4 would have temporarily exacerbated greenhouse conditions. Intense continental-scale warming in the early Aptian has been inferred from terrestrial secondary minerals (Ruffel and Batten, 1990).

Hydrate-released methane must traverse a long water column after escaping from sediment, prompting dissolution and oxidation of methane in the ocean (Kvenvolden, 1993). Widespread deposition of black shales has been observed in the Early Cretaceous marine record (e.g., Bralower et al., 1994; Menegatti et al., 1998); these shales are sometimes interpreted as the result of marine anoxic events (Sliter, 1989). We expect that methane hydrate dissociation during the Early Cretaceous first saturated the deep ocean with methane, perhaps resulting in anoxia; excess methane then migrated up the water column and was released to the atmosphere, oxidized to CO_2, and photosynthetically fixed by the standing terrestrial plant ecosystem. Figure 1B presents $\delta^{13}C_{carbonate}$ from Deep Sea Drilling Project Hole 534 (Robertson and Bliefnick, 1983) and from outcrop data that have been correlated to standard microfossil zones (Weissert, 1989); in addition, three high-resolution records of marine $\delta^{13}C_{carbonate}$ are presented for the Aptian (Jenkyns, 1995; Menegatti et al., 1998; Scholle and Arthur, 1980). Comparison of the terrestrial versus marine records has been established as a means to examine carbon reservoir shifts (Koch et al., 1992); inspection of these curves in the Early Cretaceous (Fig. 1) reveals similar trends in both records, but lower response magnitude in the marine record. Most notable is the apparent 3.0‰ decrease (Bralower et al., 1999) indicated in the marine organic carbon record during the Aptian (Fig. 1A).

Methane oxidation in the atmosphere results in a net consumption of 4 moles of O_2 for every mole of CH_4 oxidized: complete oxidation of the Aptian ~300 Gt CH_4 atmospheric emission would consume ~2400 Gt of O_2. Atmospheric O_2 levels during the Early Cretaceous are thought to be very similar (i.e., 28% ± 4% of the atmosphere) to present-day

levels (Berner and Canfield, 1989), and the Aptian CH_4 emission would consume ~0.2% of Cretaceous atmospheric O_2. Atmospheric oxygen reduction would functionally increase the CO_2:O_2 mixing ratio for terrestrial plants as O_2 is consumed during CH_4 oxidation to CO_2. When CO_2:O_2 mixing ratios increase, flowering plants gain a competitive edge over other seed plants (Bolker et al., 1995). During the Aptian, angiosperms began the exponential phase of their taxonomic diversification and expanded from their biogeographic centers of origin (Lidgard and Crane, 1988). Angiosperms first appeared in the conifer-dominated forests of Victoria, Australia (Taylor and Hickey, 1990), and southern Argentina (Romero and Archangelsky, 1986) during the middle Aptian, in the mid-latitude disturbed riparian communities of eastern North America during the latest Aptian (Hickey and Doyle, 1990), and in the Arctic by latest Albian (Spicer, 1990). Comparison of our data with the trend revealed in 1125 Cretaceous palynofloral assemblages by Crane and Lidgard (1989) shows that the Aptian atmospheric excursion closely preceded the global biogeographic expansion of angiosperms (Fig. 2). This expansion may have been facilitated by changes in the CO_2:O_2 mixing ratio produced by methane oxidation that competitively favored early angiosperms, offering a mechanistic link between Earth system atmospheric events and significant changes in terrestrial communities.

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REFERENCES CITED

Arens, N.C., 1996, Evolution of the Cretaceous tropical flora: Preliminary overview from the Andes of Colombia: American Journal of Botany, v. 83, p. 109.

Arens, N.C., Jahren, A.H., and Amundson, R., 2000, Can C3 plants faithfully record the carbon isotopic composition of atmospheric carbon dioxide?: Paleobiology, v. 26, p. 137–164.

Batten, D.J., 1996, Palynofacies and petroleum potential, *in* Jansonius, J., and McGregor, D.C., eds., Palynology: Principles and applications: Dallas, American Association of Stratigraphic Palynologists Foundation, p. 1065–1084.

Berner, R.A., 1990, Atmospheric carbon dioxide levels over Phanerozoic time: Science, v. 249, p. 1382–1386.

Berner, R.A., and Canfield, D.E., 1989, A new model for atmospheric oxygen over Phanerozoic time: American Journal of Science, v. 289, p. 333–361.

Bocherens, H., Friis, E.M., Mariotti, A., and Pedersen, K.R., 1993, Carbon isotopic abundances in Mesozoic and Cenozoic fossil plants: Palaeoecological implications: Lethaia, v. 26, p. 347–358.

Bolker, B.M., Pacala, S.W., Bazzaz, F.A., Canhams, C.D., and Levin, S.A., 1995, Species diversity and ecosystem response to carbon dioxide fertilization: Conclusions from a temperate forest model: Global Change Biology, v. 1, p. 373–381.

Bralower, T.J., Arthur, M.A., Leckie, R.M., Sliter, W.V., Allard, D.J., and Shlanger, S.O., 1994, Timing and paleoceanography of oceanic dysoxia/anoxia in the late Barremian to early Aptian (Early Cretaceous): Palaios, v. 9, p. 335–369.

Bralower, T.J., CoBabe, E., Clement, B., Sliter, W.V., Osburn, C.L., and Longoria, J., 1999, The record of global change in mid-Cretaceous (Barremian-Albian) sections from the Sierra Madre, northeastern Mexico: Journal of Foraminiferal Research, v. 29, p. 418–437.

Chaloner, W.G., and McElwain, J., 1997, The fossil plant record and global climate change: Review of Palaeobotany and Palynology, v. 95, p. 73–82.

Crane, P.R., and Lidgard, S.L., 1989, Angiosperm diversification and paleolatitudinal gradients in Cretaceous floristic diversity: Science, v. 246, p. 675–678.

Dickens, G.R., O'Neil, J.R., Rea, D.K., and Owen, R.M., 1995, Dissociation of oceanic methane hydrate as a cause of the carbon isotope excursion at the end of the Palaeocene: Palaeoceanography, v. 10, p. 965–971.

Dickens, G.R., Castillo, M.M., and Walker, J.C.G., 1997, A blast of gas in the latest Paleocene: Simulating first-order effects of massive dissociation of oceanic methane hydrate: Geology, v. 25, p. 259–262.

Farquhar, G.D., Ehleringer, J.R., and Hubick, K.T., 1989, Carbon isotope discrimination and photosynthesis: Annual Review of Plant Physiology and Plant Molecular Biology, v. 40, p. 503–538.

Gröcke, D.R., Hesselbo, S.P., and Jenkyns, H.C., 1999, Carbon-isotope composition of Lower Cretaceous fossil wood: Ocean-atmosphere chemistry and relation to sea-level change: Geology, v. 27, p. 155–158.

Guerrero, J., and Sarmiento, G., 1996, Estratigrafía física, palinológica, sedimentológica y secuencial del Cretácico Superior y Paleoceno del Piedemonte Llanero: Implicaciones en exploración petrolera: Geología Colombiana, v. 20, p. 3–66.

Harland, W.B., Armstrong, R.L., Cox, A.V., Craig, L.E., Smith, A.L., and Smith, D.G., 1989, A geologic time scale 1989: Cambridge, UK, Cambridge University Press.

Hickey, L.J., and Doyle, J.A., 1990, Early Cretaceous fossil evidence for angiosperm evolution: Botanical Review, v. 43, p. 3–104.

Jenkyns, J.C., 1995, Carbon-isotope stratigraphy and paleoceanographic significance of the Lower Cretaceous shallow-water carbonates of Resolution Guyot, Mid-Pacific Mountains, *in* Scientific results of the Ocean Drilling Program, Volume 143: College Station, Texas, Ocean Drilling Program, p. 99–104.

Koch, P.L., Zachos, J.C., and Gingerich, P.D., 1992, Correlation between isotope records in marine and continental carbon reservoirs near the Palaeocene/Eocene boundary: Nature, v. 358, p. 319–322.

Koch, P.L., Zachos, J.C., and Dettman, D.L., 1995, Stable isotope stratigraphy and paleoclimatology of the Paleogene Bighorn Basin: Palaeogeography, Palaeoclimatology, Palaeoecology, v. 115, p. 61–89.

Kvenvolden, K.A., 1993, Gas hydrates: Geological perspectives and global change: Reviews of Geophysics, v. 31, p. 173–187.

Lidgard, S., and Crane, P.R., 1988, Quantitative analyses of the early angiosperm radiation: Nature, v. 331, p. 344–346.

Lini, A., Weissert, H., and Erba, E., 1992, The Valanginian carbon isotope event: A first episode of greenhouse climate conditions during the Cretaceous: Terra Nova, v. 4, p. 374–384.

Menegatti, A., Weissert, H., Brown, R.S., Tyson, R.V., Farrimond, P., Strasser, A., and Caron, M., 1998, High-resolution $\delta^{13}C$ stratigraphy through the early Aptian "Livello Selli" of the Alpine Tethys: Paleoceanography, v. 13, p. 530–545.

Norris, R.D., and Rohl, U., 1999, Carbon cycling and chronology of climate warming during the Palaeocene-Eocene transition: Nature, v. 401, p. 775–778.

Peng, T.H., Broecker, W.S., Greyer, H.D., and Trumbore, S., 1983, A deconvolution of the tree-ring based $\delta^{13}C$ record: Journal of Geophysical Research, v. 88, p. 3609–3620.

Robertson, A.H.F., and Bliefnick, D.M., 1983, Sedimentology and origin of Lower Cretaceous pelagic carbonates and redeposited clastics, Blake-Bahama Formation, Deep Sea Drilling Project site 534, western equatorial Atlantic, *in* Initial reports of the Deep Sea Drilling Project, Volume 76: Washington, D.C., U.S. Government Printing Office, p. 795–820.

Romero, E.J., and Archangelsky, S., 1986, Early Cretaceous angiosperm leaves from southern South America: Science, v. 234, p. 1580–1582.

Ruffel, A.H., and Batten, D.J., 1990, The Barremian-Aptian arid phase in western Europe: Palaeogeography, Palaeoclimatology, Palaeoecology, v. 80, p. 197–212.

Scholle, P.A., and Arthur, M.A., 1980, Carbon isotope fluctuations in Cretaceous pelagic limestones: Potential stratigraphic and petroleum exploration tool: American Association of Petroleum Geologists Bulletin, v. 64, p. 67–87.

Sliter, W.V., 1989, Aptian anoxia in the Pacific Basin: Geology, v. 17, p. 909–912.

Spicer, R.A., 1990, Latest Cretaceous woods of the central north slope, Alaska: Palaeontology, v. 33, p. 225–242.

Stott, L.D., Sinha, A., Thiry, M., Aubry, M.-P., and Berggren, W.A., 1996, The transfer of ^{12}C changes from the ocean to the terrestrial biosphere across the Paleocene/Eocene boundary: Criteria for terrestrial-marine correlations, *in* Knox, R.O., et al., eds., Correlations of the early Paleogene in Northwest Europe: Geological Society [London] Special Publication 101, p. 381–399.

Sundquist, E.T., 1993, The global carbon budget: Geology, v. 259, p. 934–941.

Taylor, B.E., 1986, Magmatic volatiles: Isotopic variation of C, H, and S, *in* Valley, J.W., et al., eds., Stable isotopes in high temperature geological processes: American Mineralogical Society Reviews in Mineralogy, v. 16, p. 561–570.

Taylor, D.W., and Hickey, L.J., 1990, An Aptian plant with attached leaves and flowers: Implications for angiosperm origin: Science, v. 247, p. 702–704.

Weissert, H., 1989, C-isotope stratigraphy, a monitor of paleoenvironmental change: A case study from the Early Cretaceous: Surveys in Geophysics, v. 10, p. 1–61.

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