

FIG. 2 *a*, A 12/25- μm – 25/60- μm two-colour diagram for 48 IRAS point sources selected in L1641 (ref. 14). A rectangular area corresponds to the T Tauri box¹⁵. Filled circles denote IRAS point sources associated with molecular outflow. *b*, Bolometric luminosity plotted against the 12/25- μm colour diagram for the 48 IRAS point sources. Upper horizontal scale is the dust colour temperature derived from 12- μm and 25- μm flux densities on the assumption of a λ^{-1} emissivity law. Filled circles denote IRAS point sources associated with molecular outflow.

sponds to this epoch, and their ages on the birthline, 1×10^5 yr (ref. 21), are consistent with the timescales of outflow and the main accretion phase.

A consequence of this interpretation is that molecular outflow represents the main accretion phase of a solar-type low-mass protostar. It is then likely that mass accretion takes place inside a flattened circumstellar disk around a protostellar core, with outflow occurring along the poles³. If outflow is ultimately powered by the energy released by the accretion²³, the mechanical luminosity of outflow should be smaller than the bolometric luminosity. In Table 1, we find this to be the case. On the other hand, mass-ejection rate in the outflows in Table 1 is typically $10^{-5} M_{\odot} \text{ yr}^{-1}$, comparable to the mass accretion rate above. This is, however, not a problem in star formation as most of the outflowing mass is actually of interstellar origin ($r \geq 10^{16}$ cm) (ref. 1), and mass flux from the inner circumstellar region ($r \leq 10^{15}$ cm) must be significantly smaller than a mass accretion rate of $10^{-5} M_{\odot} \text{ yr}^{-1}$. □

Received 11 July; accepted 28 September 1989.

- Lada, C. J. *A. Rev. Astr. Astrophys.* **23**, 267–317 (1985).
- Fukui, Y. in *Proc. ESO Workshop on Low Mass Star Formation and Pre-main Sequence Objects* (ed. Reipurth, B.) (European Southern Observatory, Garching, in the press).
- Uchida, Y. & Shibata, K. *Publs astr. Soc. Japan* **37**, 515–535 (1985).
- Stahler, S. W., Shu, F. H. & Taam, R. E. *Astrophys. J.* **241**, 637–654 (1980).
- Takaba, H. et al. *Astron. Astrophys.* **166**, 276–282 (1986).
- Reipurth, B. & Bally, J. *Nature* **320**, 336–338 (1986).
- Bally, J., Langer, W. D., Stark, A. A. & Wilson, R. W. *Astrophys. J.* **312**, L45–L49 (1987).
- Fukui, Y. et al. *Astrophys. J.* **311**, L85–L88 (1986).
- Fukui, Y. *Vistas Astr.* **31**, 217–226 (1988).
- Fukui, Y., Takaba, H., Iwata, T. & Mizuno, A. *Astrophys. J.* **325**, L13–L15 (1988).
- Levreault, R. M. *Astrophys. J. Suppl.* **67**, 283–371 (1988).
- IRAS Catalogs & Atlases*, Explanatory Suppl. (eds Beichman, C. A., Neugebauer, G., Habing, H. J., Clegg, P. E. & Chester, T. J.) (U.S. Government Printing Office, Washington DC, 1984).
- Snell, R. L., Loren, R. B. & Plambeck, R. L. *Astrophys. J.* **239**, L17–L22 (1980).
- Strom, K. M. et al. *Astrophys. J. Suppl.* **71**, 183–217 (1989).
- Harris, S. in *Proc. ESO-IRAM-ONSALA Workshop on (Sub)millimeter Astronomy* (eds Shaver, P. A. & Kjör, K.) 527–532 (European Southern Observatory, Garching, 1985).
- Beichman, C. A. et al. *Astrophys. J.* **307**, 337–349 (1986).
- Hayashi, C. *Publs astr. Soc. Japan* **13**, 450–452 (1961).
- Herbig, G. H. & Bell, K. R. *University of California, Lick Observatory Bulletin No. 1111*. (1988).
- Cohen, M. & Kuhl, L. V. *Astrophys. J. Suppl.* **41**, 743–843 (1979).
- Rucinski, S. M. *Astr. J.* **90**, 2321–2330 (1985).
- Stahler, S. W. *Astrophys. J.* **274**, 822–829 (1983).
- Shu, F. H., Adams, F. C. & Lizano, S. A. *Rev. Astr. Astrophys.* **25**, 23–81 (1987).
- Strom, S. E., Strom, K. M. & Edwards, S. in *Proc. NATO Advanced Study Institute on Galactic and Extragalactic Star Formation* (eds Pudritz, R. E. & Fich, M.) 53–88 (Kluwer, Dordrecht, 1988).
- Mergulis, M. & Lada, C. J. *Astrophys. J.* **299**, 925–938 (1985).
- Iwata, T., Fukui, Y. & Ogawa, H. *Astrophys. J.* **325**, 372–381 (1988).

Development of Asian monsoon revealed by marked ecological shift during the latest Miocene in northern Pakistan

Jay Quade, Thure E. Cerling & John R. Bowman

Department of Geology and Geophysics, University of Utah, Salt Lake City, Utah 84112, USA

CARBON isotopes from soil carbonate^{1–4} and soil organic matter^{5,6} yield palaeoecological information because the carbon in the soil carbonate forms in isotopic equilibrium with local soil CO₂ (refs 1, 7), the isotopic composition of which is in turn determined by local plant cover. Siwalik Group sediments in northern Pakistan contain a well exposed palaeosol record spanning the past 18 Myr. Here we report on stable-carbon-isotope results from associated pedogenic carbonate which indicate a dramatic ecological shift from C₃- to C₄-dominated floodplain biomass beginning ~7.4–7.0 Myr ago. The earlier C₃ floodplain biomasses were probably mainly composed of trees and shrubs, whereas C₄ grasslands dominated in the Plio-Pleistocene. Oxygen isotopes also exhibit a shift in the latest Miocene, probably corresponding to a major climate change which may have induced the forest-to-grassland transition. This dramatic ecological shift in the latest Miocene may mark the inception or a marked strengthening of the Asian monsoon system.

Plants display three distinct carbon isotopic groupings. C₃ plants include virtually all trees irrespective of climate, nearly all shrubs and herbs, and grasses favoured by a cool growing season. They average ~–27‰, but display a range from –35 to –20‰ depending on genus, plant longevity, moisture stress, light intensity and other factors^{8,9}. C₄ plants include a few shrubs in the families Euphorbiaceae and Chenopodiaceae, but especially grasses favoured by warm growing seasons. C₄ grasses average ~–13‰. CAM (crassulacean acid metabolism) plants, which include succulents like cactus and some yuccas, are not important components of ecosystems outside deserts, and will not be discussed here. The isotopic compositions of soil organic matter, where preserved, and of soil CO₂ where soil respiration rates are high, are then determined by the proportion of C₃:C₄ plants in the local biomass. Fractionation effects arising from gaseous diffusion and equilibrium exchange produce an enrichment of

$\delta^{13}\text{C}$ ($[(^{13}\text{C}/^{12}\text{C})_{\text{sample}}/(^{13}\text{C}/^{12}\text{C})_{\text{standard}}]-1$) in soil carbonate of $\sim 15\%$ with respect to soil-respired CO_2 (refs 1, 7, 10). Soil carbonate formed in the presence of a pure C_3 and pure C_4 biomass would then have $\delta^{13}\text{C}$ (PDB) values at 25°C of ~ -12 and $+2\%$, respectively. Oxygen isotopes in soil carbonate have been shown to have a strong positive correlation with the isotopic content of local rainfall¹.

Palaeosols from the well dated¹¹⁻¹⁵ (Fig. 1) Siwalik Group sediments in the Potwar Plateau region are abundant within the fine-grained floodplain facies of large river systems. Palaeosols typically contain pale red to yellow, clay-rich B horizons that have been thoroughly bioturbated and leached of carbonate. Organic A horizons, which often cap Plio-Pleistocene palaeosols, are rare in the Dhok Pathan formation (8.9–5.3 Myr), and are absent in older beds. We took great care in all our sections to sample carbonate nodules associated with this suite of pedologic features, preferably from the soil base (calic horizons). Most, but not all, palaeosols in the section contained pedogenic carbonate.

Our sampling density averaged ~ 1 palaeosol per 130,000 yr over 17 Myr. From the standpoint of both carbon and oxygen isotopes, the record contains three distinct phases.

From 18 to 7.4–7.0 Myr, almost all of the $\delta^{13}\text{C}$ results (using the PDB dating standard) for soil carbonate fall between -13 and -9% (Fig. 2). Organic matter from two A horizons in the Dhok Pathan formation is $\sim 15\%$ more negative than associated carbonate. This is the same isotope difference displayed between the two phases in modern soils¹⁰, indicating that neither com-

ponent has experienced diagenetic alteration. A single soil at the 8.5-Myr level from the Chinji village section yielded results in the range -7 to -5% . The $\delta^{18}\text{O}$ (PDB) of soil carbonate averaged ~ -10 to -9% , but showed a higher standard deviation than carbon (Fig. 3).

From 7.4–7.0 to 5 Myr, the stable carbon and oxygen isotopic compositions of soil carbonate shift towards more positive values (Figs 2 and 3). A carbon shift seems to begin at ~ 7.4 Myr at Kaulial, but values more positive than the -9% maximum typical of older levels do not occur consistently until 6.4 Myr. At Jalalpur the change had commenced by 7.3–7.2 Myr. We are uncertain whether the spread in these dates is real, or the result of small errors in palaeomagnetic age estimates. The shift at Rohtas, not included in Fig. 1 owing to dating uncertainties in the lower portion of the section, begins a few hundred metres below the base of the Gilbert Chron, a position consistent with a latest Miocene age. This shift was completed at Rohtas and at Jalalpur by ~ 5 Myr with the attainment of positive carbon values. The soil carbonate $\delta^{18}\text{O}$ (PDB) also changed by $\sim 3\%$ during this period, attaining values of ~ -7 to -5% (Fig. 3).

From 5 to 0.4 Myr, the carbon isotopes for soil carbonate fall between -2 and $+2\%$. Soil organic matter lies between -14 and -11% (ref. 10). The average isotope separation between organic matter and soil carbonate is $\sim 15\%$, again indicating that no diagenetic alteration of isotopic composition has occurred¹⁰. The soil carbonate $\delta^{18}\text{O}$ (PDB) falls largely between -7 and -5% for this period and for modern soils in the area.

The carbon isotope results from 52 different palaeosols indicate that a pure or nearly pure C_3 biomass dominated floodplains before 7.4–7.0 Myr ago. In modern ecosystems at low latitudes and altitudes, the narrow range of carbon values from this period can only be produced by closed canopy forests with or without an understorey of C_3 shrubs and herbs. C_3 grasses, normally restricted to cooler higher latitude/higher elevation climates, can also be present in the shaded understorey, but their overall contribution to the biomass is small. Where the canopy is broken, however, patches of C_4 -dominated grassland occur owing to the resulting higher irradiance, water stress, and temperatures^{16,17}. C_4 plants were present, as indicated by a single palaeosol from the level of ~ 8.5 Myr, but evidently they were not common. We therefore cannot interpret the floodplain ecosystem as having been a 'mosaic' of mixed grassland and woodland. Our reconstruction is at variance with previous interpretations based on palaeosols that identified a mosaic of habitats, including grasslands, in the lower Dhok Pathan formation at Khaur¹⁸.

The advent of isotopically more positive soil organic matter and carbonate values after 7.4–7.0 Myr marks the gradual

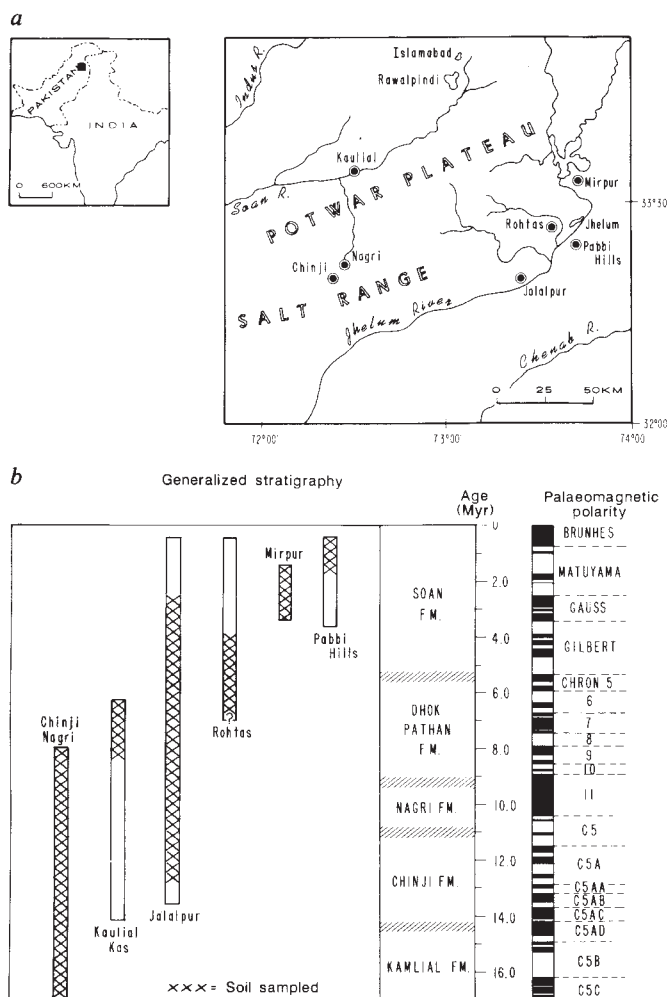


FIG. 1 a, Locations of study sections and b, approximate intervals over which palaeosols were sampled in the Potwar Plateau, northern Pakistan. (Fm. is formation.) Magnetic polarity timescale is from ref. 36.

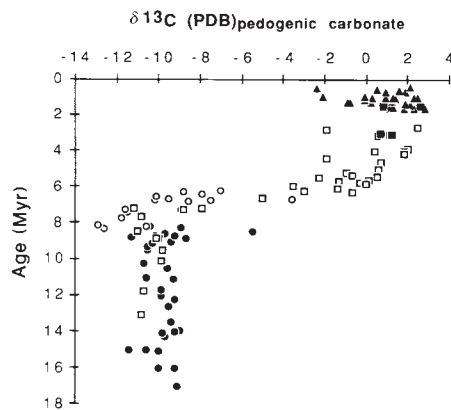


FIG. 2 The $\delta^{13}\text{C}$ (PDB) of palaeosol carbonate nodules plotted against age in the Siwalik Sequence, northern Pakistan. The negative values before 7.4 Myr indicate a C_3 biomass dominated the floodplain, whereas more positive values in the Plio-Pleistocene palaeosols indicate the dominance of C_4 grasslands. ●, Chinji-Nagri; ○, Kaulial Kas; ■, Mirpur; □, Jalalpur; ▲, Pabbi hills.

expansion of C_4 plants on to the floodplain. This must signify the appearance of grasslands because they are the only known C_4 biomasses of large extent. Therefore, a vegetation mosaic of C_4 grassland and C_3 forest was probably present between 7.4–7.0 and 5 Myr. By 5 Myr, C_4 grasses constituted >90% of the biomass, and this persisted through the top of the record (~0.4 Myr). The few negative values obtained perhaps signify limited riparian habitats in which some C_3 trees and shrubs grew. The isotope shift in the carbonates cannot be explained by changes in soil respiration rates over time because soil organic matter also reflects the shift. Moreover, leached zones in Siwalik palaeosols usually exceed 50 cm, consistent with high-respiration-rate soils in which the isotopic composition of carbonate is controlled by the proportion of C_3 : C_4 plants^{1,7,10}.

The reasons for the late Miocene ecological shift can be interpreted in two ways. The shift may have been in response to a climate change that allowed the invasion of C_4 grasses from outside the region. Alternatively, the ecological shift may mark the evolutionary first appearance of C_4 plants, and thus a floral invasion unrelated to a climate change. The oldest C_4 grasses yet documented appear in the late Miocene (~7.0 Myr)¹⁹ of North America, but the palaeobotanical record for grasses is very sparse.

The oxygen isotopes provide important evidence as they shift in every section towards more positive values slightly before the carbon isotope shift. Diagenesis does not seem to have modified the isotopic composition of the carbonates, and the oxygen event is widespread, affecting the entire Potwar Plateau. The isotopic composition of soil water is determined by temperature, the isotopic composition of regional rainfall, seasonality of precipitation, and other factors. Without further evidence we are reluctant to speculate on the exact reason for the shift except to say that a major change in some aspect of the regional climate is indicated.

Moderate rainfall, seasonal drought, and natural fires are all viewed as important to modern grasslands²⁰. One or more of these conditions may have been absent before 7.4 Myr. Higher rainfall in the Miocene, as indicated by thick leached zones in soils, is one explanation. Depth of leaching in modern soils in the area is <50 cm. Leaching depths of 1–2 m (after compaction) in the middle and late Miocene indicate ≥ 1 m of rain per year based on modern patterns²¹. The presence of soil carbonate in palaeosols throughout the record precludes the wet tropical climate called for by Krynine²².

Some soil features, as with isotopic composition, change sharply after ~7 Myr. For example, depth of soil leaching decreases, on average to 50 cm or less. Leached zones (B horizons) are notably yellow (2.5 Y 5/5 dry), in contrast to the strong reds (5 YR 4/4 dry) and oranges of older soils. Most notably, organic A horizons become common in younger sediments. Oxidation during diagenesis may have removed the older humic horizons and altered original soil colours. However, the humic zones and colour changes begin to appear at Kaulial Kas, Jalalpur and Rohtas at ~7.5 Myr, even though burial depths and therefore diagenetic histories differ between sections. Another possible explanation is that the appearance of humic soil zones marks the expansion of grasslands. Of the 17 humus horizons analysed so far, 15 indicate a pure or nearly pure C_4 biomass. In modern soils, organic accumulations under grasslands (mollic epipedons) are considerably greater than under forests²³.

To understand the origins of the vegetation shift in the latest Miocene, it is instructive to look at modern monsoonal climate and its impact on the modern vegetation and soils. Modern climate in northern Pakistan is monsoonal, with 70% of the 40-cm annual rainfall for the Potwar Plateau falling in the warm summer months. This moderate rainfall, the strongly seasonal rainfall distribution, and prevalence of natural fires from summer lightning strikes all favour grasslands over forest²⁰. Mean daily summer growing-season temperatures fall between 30 and

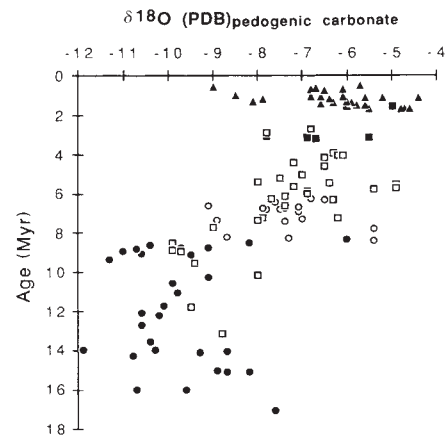


FIG. 3 The $\delta^{18}\text{O}$ (PDB) of palaeosol carbonate nodules in the Siwalik Sequence, northern Pakistan. Oxygen isotopes shift towards more positive values in the late Miocene, indicating a change in the average isotopic composition of soil water probably resulting from regional climatic change. Key, as in Fig. 2.

35 °C on the Potwar Plateau, the range favoured by C_4 grasses in particular^{24–27}. Plants listed for western India²⁸ and our own observations on the Jhelum River floodplain indicate that the extant native grasses of the Potwar Plateau region are C_4 s. It is therefore likely that the biomass on floodplains was a largely C_4 grassland before modification by man in the past few millennia.

Our evidence indicates that if the monsoon system already existed it experienced a strong intensification beginning at ~7.4–7.0 Myr, marked by the first appearance of C_4 grasslands favoured by the monsoonal climate of the Potwar Plateau today. The marine record from the northern Indian Ocean supports this, in that diatom assemblages specific to monsoon circulation patterns first appeared 11–10 Myr ago. Abundances of these diatoms increased markedly at 7.3 Myr, an event thought to indicate intensification of the reversing monsoon in the region²⁹.

Our vegetation reconstruction is also supported by mammalian fossil evidence. The larger animals before ~7.0 Myr (tragulids, suids, Okapia-like giraffes, low-crowned bovids and others) were browsers^{30–32}. At 7.4 Myr a major faunal turnover occurred, one which Barry *et al.*³³ speculated was related to climate change. After 7.4 Myr, a grazing adaptation is manifest in the high-crowned dentition of a newly appearing tragulid and large bovids. Overall, the browsers are much less evident in the younger assemblage. Rodents also show a major turnover at ~7.0 Myr, which has been interpreted to be caused by displacement of forests by grassland³⁴. Most significantly *Sivapithecus* sp., which is evidently allied to tree-dwelling orangutans, disappeared from the record at 7.4 Myr³⁵, at about the time grasslands appear. □

Received 4 July; accepted 2 October 1989.

1. Cerling, T. E. *Earth planet. Sci. Lett.* **71**, 229–240 (1984).
2. Cerling, T. E., Bowman, J. R. & O'Neill, J. R. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **63**, 335–356 (1988).
3. Cerling, T. E. & Hay, R. L. *Quat. Res.* **25**, 63–78 (1986).
4. Magaritz, M., Kaufman, A. & Yaalon, D. H. *Geoderma* **25**, 157–172 (1981).
5. Krishnamurthy, R. V. *Earth planet. Sci. Lett.* (submitted).
6. Guillet, B., Faivre, P., Mariotti, A. & Khobzi, J. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **65**, 51–58 (1988).
7. Quade, J., Cerling, T. E. & Bowman, J. R. *Bull. geol. Soc. Am.* **101**, 464–475 (1989).
8. Ehleringer, J. R. in *Applications of Stable Isotope Ratios to Ecological Research* (ed. Rundel, J. W., Ehleringer, J. R. & Nagy, K. A.) 41–54 (Springer, New York, 1989).
9. O'Leary, M. H. *Phytochemistry* **20**, 553–567 (1981).
10. Cerling, T. E., Quade, J., Wang, Y. & Bowman, J. R. *Nature* **341**, 138–139 (1989).
11. Johnson, N. M., Stix, J., Tauxe, L., Cervený, P. F. & Tahirkehi, R. A. K. *J. Geol.* **93**, 27–40 (1985).
12. Tauxe, L. & Opdyke, N. D. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **37**, 43–61 (1982).
13. Johnson, N. M., Opdyke, N. D., Johnson, G. D., Lindsay, E. H. & Tahirkehi, R. A. K. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **37**, 17–42 (1982).
14. Keller, H. M. *et al.* *Earth planet. Sci. Lett.* **36**, 187–201 (1982).
15. Barry, J. C., Lindsay, E. H. & Jacobs, L. L. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **37**, 95–130 (1982).

16. Ehleringer, J. R., Lin, Z. F., Field, C. B., Sun, G. C. & Kuo, C. Y. *Oecologia* **72**, 109–114 (1987).
17. Ehleringer, J. R., Field, C. B., Lin, Z. & Kuo, C. *Oecologia* **70**, 520–526 (1986).
18. Retallack, G. J. *Phil. Trans. R. Soc. Lond.* **B309**, 105–142 (1985).
19. Thomasson, J. R., Nelson, M. E. & Zakrzewski, R. J. *Science* **233**, 876–888 (1988).
20. Walter, H. *Ecology of Tropical and Subtropical Vegetation* (Oliver and Boyd, Edinburgh, 1971).
21. Jenny, H. *Factors in Soil Formation* (McGraw-Hill, New York, 1941).
22. Krynine, P. D. *Am. J. Sci.* **5**, 422–446 (1937).
23. Brady, N. C. *The Nature and Properties of Soils* (Macmillan, New York, 1984).
24. Hattersley, P. W. *Oecologia* **57**, 113–128 (1983).
25. Teeri, J. A. & Stowe, L. G. *Oecologia* **23**, 1–12 (1976).
26. Ehleringer, J. R. *Oecologia* **31**, 255–267 (1978).
27. Tiezen, L. L., Senyimba, M. M., Imbamba, S. K. & Troughton, J. H. *Oecologia* **37**, 337–350 (1979).
28. Sankhla, N., Ziegler, H., Vyas, O. P., Stickler, W. & Trimern, P. *Oecologia* **21**, 123–129 (1975).
29. Burkle, L. H. *J. Micropaleontology*, (in the press).
30. Tattersall, I. *Nature* **222**, 451–452 (1969).
31. Tattersall, I. *Nature* **224**, 821–822 (1969).
32. Prasad, K. N. *Nature* **232**, 413–414 (1971).
33. Barry, J. G., Johnson, N. M., Raza, S. M. & Jacobs, L. L. *Geology* **13**, 637–640 (1985).
34. Flynn, L. J. & Jacobs, L. L. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **38**, 129–138 (1982).
35. Barry, J. C. in *Primate Evolution* (eds Else, J. G. & Lee, P. C.) 93–106 (Cambridge University Press, 1986).
36. Berggren, W. A., Kent, D. V., Flynn, J. J. & van Couvering, J. A. *Bull. geol. Soc. Am.* **96**, 1407–1418 (1985).

ACKNOWLEDGMENTS. We thank J. C. Barry, A. K. Behrensmeyer, S. M. Raza, I. M. Shah and the Smithsonian Institution and Geological Survey of Pakistan for facilitating this work. We also thank M. Anwar, M. Asif Jah and R. Rennell for their help on critical sections. The NSF and the Smithsonian Foreign Currency Program funded this work.

Old carbon in living organisms and young CaCO₃ cements from abyssal brine seeps

C. K. Paull*, C. S. Martens*, J. P. Chanton†, A. C. Neumann*, J. Coston*, A. J. T. Jull‡ & L. J. Toolin‡

* University of North Carolina, Chapel Hill, North Carolina 27599, USA

† Florida State University, Tallahassee, Florida 32306, USA

‡ University of Arizona, Tucson, Arizona 85721, USA

ABYSSAL brine springs at the base of the Florida Escarpment in the Gulf of Mexico (~3,280 m, 26° 02' N 84° 55' W) are surrounded by communities of abundant heterotrophic organisms^{1–4} and carbonate-cemented crusts⁵. These organisms are apparently supported by local chemosynthetic primary production because the ¹³C content of their tissues is lower than that found in typical photosynthetically fixed organic carbon⁶, and the enzymes associated with chemosynthetic metabolism are present in their tissues^{7,8}. Here we report that fossil methane is the dominant source of carbon found in living tissues and recent authigenic carbonate minerals associated with abyssal brine seeps at the base of the Florida Escarpment. Most organic carbon and authigenic carbonates adjacent to the seeps contain progressively less modern carbon as their ¹³C contents approach that of methane carried in the brine. Incorporation of fossil methane (≤1.3% modern carbon) into living tissues and carbonate cements results in recently formed materials which are depleted in ¹⁴C. Therefore, ¹⁴C cannot be used to indicate the age of authigenic materials produced at the pore-water-seepage environments that speckle the continental margins of the world.

The energy source for deep-sea chemosynthetic communities is reduced compounds, such as HS⁻, CH₄, NH₄⁺ or petroleum^{9,10}, which may be carried in pore fluids from anoxic subsurface formations onto the oxygenated sea floor. Dissolved ΣH₂S (H₂S, HS⁻, S²⁻), NH₄⁺ and CH₄ are all present in brines that discharge onto the sea floor at the base of the Florida Escarpment^{11,12}. The extremely negative δ¹³C values [(¹³C/¹²C)_{sample} - (¹³C/¹²C)_{Std}] × 1,000 (< -70‰ relative to the PDB standard) found in tissues of animals living on patches of sediment enriched in isotopically light methane⁶ indicates the overriding importance of methylotrophy¹³. Biogenic methane is the only natural carbon source that has such depleted δ¹³C values⁶. The chemosynthetic use of ΣH₂S and NH₄⁺ would require uptake of dissolved inorganic carbon which is enriched

in ¹³C and therefore should produce isotopically 'heavy' organic tissue.

Earlier measurements of ¹⁴C in animal tissues from this same site produced a surprising result; tissues with very negative δ¹³C values contain significant amounts of modern carbon (~60%)⁶. Here we report new measurements of δ¹³C and ¹⁴C from the ambient methane, living animal tissues, bacterial mats, sedimentary organic carbon and authigenic carbonate carbon collected at the brine seeps during RV ALVINs 1986 visit to the Florida Escarpment (Table 1), which we use to establish the carbon sources and pathways.

The seep methane samples contain between 1.0% and 1.3% modern carbon. However, we cannot distinguish these measured values from 0% modern because they are close to the detection limit of the accelerator-mass-spectrometer technology used (0.4%) (ref. 14) and the extensive sample handling involved in concentrating the methane may have added to the ¹⁴C background.

In our new, larger data set, the δ¹³C and ¹⁴C values of the animal tissues and other organic carbon samples exhibit two trends (Fig. 1). Data lying in envelope A (between ambient methane and average marine organic carbon) indicate that fossil methane mixed with varying amounts of pelagic organic matter is the dominant source of carbon in the seep organisms' tissues; some contain as little as 12.8% modern carbon. Other organic samples with extremely negative δ¹³C values contain appreciable amounts of modern ¹⁴C and exhibit a second mixing trend (B in Fig. 1). These data may indicate that there is a second source of young biogenic methane here⁶, generated by rapid methanogenesis using organic detritus in the sediments around the seeps. Methanogenesis in organic-rich sediments around the seeps should be stimulated by the nearly complete sulphate depletion, as it is in coastal sediments¹⁵. Such depletion results from mixing of pore fluids with sulphate-free seep fluids at the Florida Escarpment¹¹. Dual sulphide sources have been identified by stable-isotope analysis, some advected in with the brines and some produced by microbial sulphate reduction in the sediments¹¹. Thus, there may also be dual methane sources. A less likely, but possible explanation for the second mixing trend, is the contamination of samples by ¹⁴C used in tracer experiments that are periodically conducted in shared laboratory facilities. We note that this trend is dominated by earlier measurements⁶.

The shells of living mussels contained 82 ± 3% modern carbon and a mean δ¹³C (PDB) value of -4.3 ± 1.7‰. These values are similar to those expected for bottom water¹⁶ and confirm that the dominant source of shell carbon is dissolved seawater bicarbonate.

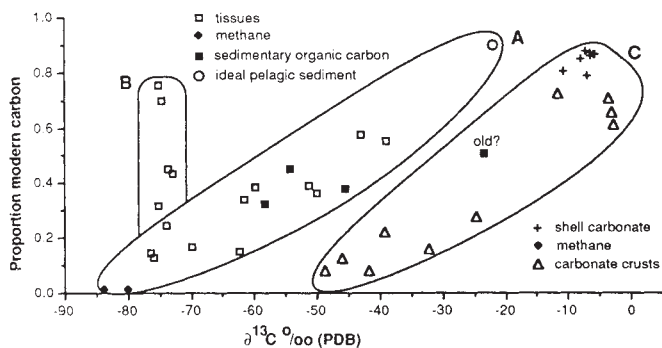


FIG. 1 Plot of the proportion of modern carbon and δ¹³C values of carbon-carrying components associated with abyssal brine seeps at the base of the Florida Escarpment, Gulf of Mexico. Values are given in Table 1. Envelope A contains the organic carbon measurements and indicates the mixing trend between ideal planktonic organic carbon (δ¹³C (PDB) = -22‰ and 90% modern carbon)²¹ and the ambient methane. Envelope B indicates a secondary trend in the organic data. Ageing causes one sedimentary sample to fall below the envelope. Envelope C contains the carbonate data.