

Changes in organic carbon stable isotope ratios across the K/T boundary: global or local control?

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ABSTRACT

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A global shift in carbonate carbon $\delta^{13}\text{C}$ -values from heavier values in the Maastrichtian to lighter values in the early Danian indicates recycling of isotopically light organic carbon to inorganic carbon reservoirs during a period of depressed marine productivity. Comparison of organic carbon $\delta^{13}\text{C}$ -values from globally dispersed K/T sections does not show a similar, globally well-developed pattern. Several factors evidently overwhelm the potential impact of an isotopically lighter inorganic carbon source on organic matter isotopic signatures: (1) species changes in biological assemblages may modify the averaged isotopic fractionation of organic matter; and (2) shifts in the proportion of land/marine organic matter contributions to coastal marine locations may overprint the isotopic record. Local phenomena evidently outweigh global change in determining the isotope signature of organic carbon deposited in K/T boundary sections.

1. Introduction

The end of the Cretaceous Period, ~66 Ma ago, was accompanied by massive reductions in the diversity and numbers of many organisms. The marine fossil record shows marked depletions in calcareous plankton and tropical invertebrates. On land, dinosaurs became extinct at the end of the Maastrichtian (e.g., Sloan et al., 1986), as did many other types of organisms. New genera of plants and animals gradually replaced the decimated species in the early Danian, but an abrupt boundary separates the Cretaceous from the Tertiary virtually everywhere in the geological record.

Several possible explanations for this major

perturbation in biotic evolution have been considered. The general occurrence of Ir enrichment in the boundary clay layer has been interpreted as evidence of an extraterrestrial cause — a massive meteorite impact (Alvarez et al., 1984). Alternatively, this enrichment has been considered prime evidence of extensive volcanism at the Cretaceous/Tertiary transition (Officer et al., 1987). Either scenario might induce a global change in climate and a consequent impact on the biosphere. Another possibility is that of abrupt climate change brought about by the gradual accumulation of non-catastrophic factors (Crowley and North, 1988). Regardless of the cause of this intriguing event in Earth history, the effect was a major change in the nature and abundance of global life.

The change in the fossil record is mirrored in the isotopic record. A shift of 1–2‰ to-

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wards lighter $\delta^{13}\text{C}$ -values has been reported in marine carbonates from the South Atlantic (Shackleton and Hall, 1984), Israel (Magaritz, 1989), the North Pacific (Zachos et al., 1989), Tunisia (Keller and Lindinger, 1989) and Texas (Barrera and Keller, 1990), implying a global perturbation in the carbon cycle. This perturbation is believed to result from a period of globally depressed bioproductivity (Zachos and Arthur, 1986; Magaritz, 1989). During this period, the rate of oxidative recycling of organic matter evidently exceeded the rate of photosynthetic carbon fixation, returning isotopically light carbon to the inorganic carbon reservoir faster than it was removed and producing the isotope shift observed worldwide.

Changes in continental flora evidently also occurred at the time of the K/T boundary. These changes are indicated by shifts from a dominance of angiosperm pollen in Maastriichtian rocks to fern spores in Danian rocks at locations in western North America (Tschudy et al., 1984) and northern Japan (Saito et al., 1986). Forests evidently were destroyed at the end of the Cretaceous. Ferns are hardy plants; they could have been survivors of the K/T biotic perturbations or perhaps the first settlers of devastated areas after this event. A possible agent in the destruction of land plants is a global conflagration initiated by a bolide impact or by volcanism. Evidence of large-scale combustion of plant material is present in the distributions of polycyclic aromatic hydrocarbons (Venkatesan and Dahl, 1989) and in the presence of soot (Wolbach et al., 1985, 1990) commonly found in boundary clay layers. If as widespread as these reports imply, release of CO_2 from destruction of continental vegetation could conceivably compound the impact of depressed marine productivity on the global carbon isotope budget. However, Kump (1991) notes that maintenance of the lightened marine inorganic carbon isotopic signature for the period observed would require continued burial of organic

matter in the oceans. This material was possibly derived from continued continental biosynthesis, although from altered ecosystems.

The documented and implied changes in the compositions and abundance of biota and in the global inorganic carbon supply presumably would leave an imprint on the $^{12}\text{C}/^{13}\text{C}$ ratio of organic matter buried in sediments. This investigation was initiated to determine the nature of any variation in the carbon isotopic composition of organic matter deposited before and after the K/T boundary event and to assess its global expression.

2. Samples and analysis

2.1. Sampled locations

2.1.1. DSDP Site 577, Shatsky Rise, NW Pacific. Deep Sea Drilling Project Site 577 is located in 2700 m of water on the Shatsky Rise in the northwest Pacific Ocean. Samples came from Hole 577, which was hydraulically piston cored and yielded a K/T boundary within a white calcareous nannofossil ooze sequence (Wright et al., 1985). The boundary was identified from nannoplankton stratigraphy (Monechi, 1985), and an Ir anomaly was found in Hole 577B (Michel et al., 1985) at a stratigraphic level equivalent to the nannoplankton K/T boundary in Hole 577. Samples were frozen upon collection.

2.1.2. DSDP Site 605, New Jersey continental rise, W. North Atlantic. Deep Sea Drilling Project Site 605 is located in 2200 m of water on the continental slope of southern New Jersey. Hole 605 was rotary drilled, penetrating a series of ocean margin deposits. The early Paleocene sediments immediately above the K/T boundary contain a large fraction of terrigenous silt, probably as a result of lowered sea-level (van Hinte et al., 1987). Samples were col-

lected specifically for organic geochemical study. They remained frozen until analysis.

2.1.3. Brazos River, Texas, U.S.A. Jiang and Gartner (1986) describe the Brazos River section. It consists of an apparently continuous nannofossil record from the late Maastrichtian to the early Paleocene which was deposited in a continental shelf setting. This may be the most complete K/T section known across the boundary itself. Samples were collected from outcrop in 1989 and remained frozen until analysis. They include one sample from the postulated "tsunami layer" made up of coarse shelly sediment (Bourgeois et al., 1988).

2.1.4. Moreno Formation, California, U.S.A. The Moreno Formation is comprised of a series of shales, siltstones and sandstones deposited in a marine coastal paleosetting. The Lower Dos Palos Shale occurs in the basal Danian and overlies the Marca Shale. Samples used in this study originate from the Exxon Chaney Ranch 1 well located on the western edge of the San Joaquin Valley.

2.2. Analysis

2.2.1. Carbonate and organic carbon determinations. Samples were freeze-dried and coarsely ground in preparation for carbon analyses. Inorganic carbon concentrations were measured by the carbonate bomb procedure (Müller and Gastner, 1971; Dunn, 1980) or with a Coulometrics® Carbonate Carbon Analyzer (cf. Engleman et al., 1985). Analyses of pure calcium carbonate standards and of subsamples of the Brazos samples showed the two procedures to give identical results. The carbon concentrations were converted to calcium carbonate concentrations assuming that the inorganic carbon was present as calcite. Organic carbon concentrations were determined by measuring the carbon remaining after the carbonate bomb determinations with a Coulometrics® Total Carbon Analyzer or by using

a Rock-Eval® TOC module. Standards having a range of organic carbon contents were used to intercalibrate these procedures.

2.2.2. Carbon isotope analysis. Organic carbon isotopic ratios were measured in the carbonate-free residues of the carbonate bomb analyses by The University of Michigan Light Stable Isotope Laboratory using a Finnigan® Delta S mass spectrometer. The data have been standardized to NBS standards and corrected for ^{17}O , and they are presented relative to the PDB standard.

3. Results and discussion

A variety of factors influence the carbon isotopic composition of organic matter (cf. Fry and Sherr, 1984; McKenzie, 1985; Popp et al., 1989). Foremost among these is the difference in isotopic fractionation between C_3 (Calvin-Benson) and C_4 (Hatch-Slack) biosynthetic pathways. Plants utilizing the C_3 pathway strongly select ^{12}C and typically create a fractionation averaging -20‰ from the inorganic carbon source. C_4 plants less selectively incorporate ^{12}C . Their biomass averages 13‰ lighter than their carbon source. Most plants in the biosphere use the C_3 pathway, yet some environments, such as modern intertidal marshes and agricultural areas, are dominated by C_4 plants (cf. Sherr, 1982). Changes in vegetation types can consequently impact the sedimentary organic carbon isotopic record.

Preferential uptake of ^{12}C by biota results in organic carbon being consistently isotopically lighter than inorganic carbon, but the magnitude of isotopic fractionation depends on the availability of CO_2 . As summarized by Popp et al. (1989), high concentrations of CO_2 favor enzymatic rate control over diffusional rate control in isotopic fractionation, with the result that organic carbon synthesized under

these conditions is isotopically more light. Therefore, fluctuations in oceanic p_{CO_2} or atmospheric CO_2 concentrations can potentially appear in the organic matter isotopic record.

The isotopic composition of the inorganic carbon taken up during photosynthesis also affects the ultimate isotopic signature of organic matter. CO_2 in the atmosphere and dissolved in fresh waters is generally $\sim 7\text{‰}$ lighter than the dominant inorganic carbon species, HCO_3^- , in seawater. This difference means that plants on continents and in lakes typically have $\delta^{13}\text{C}$ -values of -28 to -26‰ whereas temperate marine algae have values of -22 to -20‰ . In coastal areas where isotopically light inorganic carbon from freshwater mixes with the marine carbon reservoir, organic carbon with more negative $\delta^{13}\text{C}$ -values can be produced (e.g., Tan and Strain, 1983). Bacterial oxidation of isotopically light organic carbon can also create more negative $\delta^{13}\text{C}$ -values of inorganic carbon in regions of the ocean beneath areas of high organic matter production (e.g., Kroopnick, 1980).

Two additional factors — the temperature during biosynthesis and the molecular composition — influence the isotopic content of organic matter. Cooler water temperatures commonly result in greater fractionation of carbon isotopes with the effect that ^{12}C uptake is enhanced; marine organic matter produced in colder waters has more negative $\delta^{13}\text{C}$ -values (Sackett et al., 1965). Within the mixture of different compounds which comprise bulk organic matter is a range in carbon isotopic ratios. Carbohydrates and proteins are isotopically heavier on average than total organic carbon, whereas lipids are lighter than the bulk isotopic value (Degens et al., 1968). Variations in the proportions of proteins to lipids, for instance, can produce differences in the isotopic composition of organic matter produced at different times by a population of marine algae. Moreover, diagenesis can potentially create isotopic shifts through the preferential degradation of carbohydrates and pro-

teins and selective preservation of lipids. Despite this potential effect, a diagenetic isotopic shift appears not to occur in typical sedimentary accumulations (e.g., Jasper and Gagosian, 1989), although it has been described in sediments rich in organic matter (e.g., Spiker and Hatcher, 1984).

The many environmental variables which influence the carbon isotopic composition of organic matter makes this sedimentary parameter a sensitive indicator of past changes in biota, climate and depositional conditions. Given the documented shift to lighter isotopic ratios found in the inorganic carbon components of Danian deposits from locations distributed around the world, an investigation of organic matter isotopic records across the K/T boundary at four sites was done.

The four sites display a wide range in carbonate carbon concentrations (Fig. 1), which is related to their paleoceanographic settings. The Moreno Formation is comprised largely of clastic, terrigenous sediments which were deposited in a nearshore environment; carbonate percentages are $< 5\%$ in Maastrichtian rocks and are $\sim 3\%$ in Danian samples. Most of the Brazos samples, representing a continental shelf shallow-water marine environment, contain $< 20\%$ carbonate carbon. The shell-hash layer, probably storm-deposited (Gartner et al., 1990), reaches 75% CaCO_3 . Cretaceous sediments from continental slope DSDP Site 605 have carbonate percentages between 30% and 55% , while Tertiary sediments contain only 8% carbonate. Contributions of shallow-water clastic sediment components during sea-level regression are recorded by this compositional change. Open-ocean DSDP Site 577 has carbonate concentrations of $\sim 90\%$ and is virtually free of continental sediment components.

Organic carbon concentrations are generally inversely related to the carbonate concentrations in the data from these four K/T boundary sites. The exception to this generalization is the Moreno Formation. Organic carbon con-

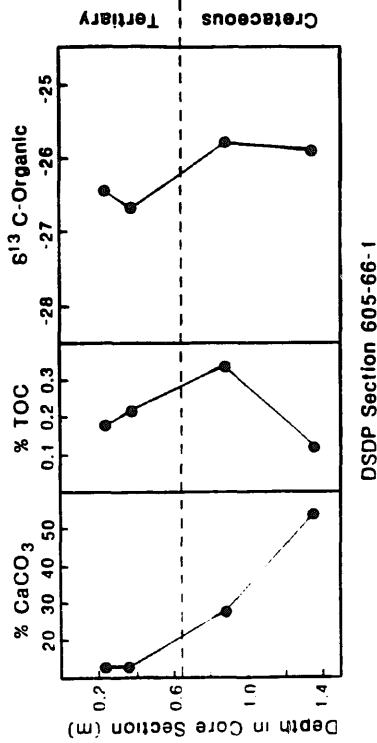
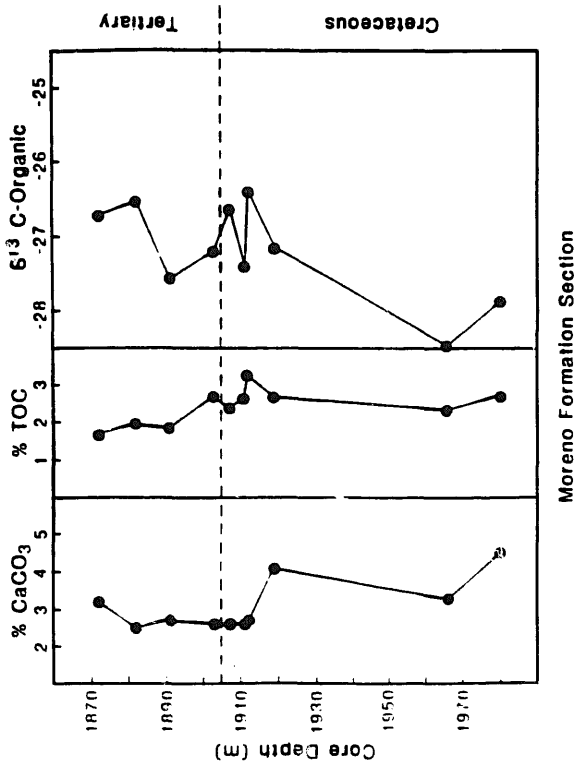
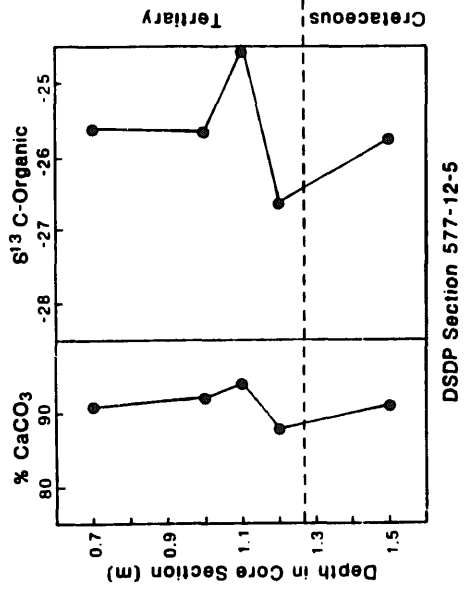
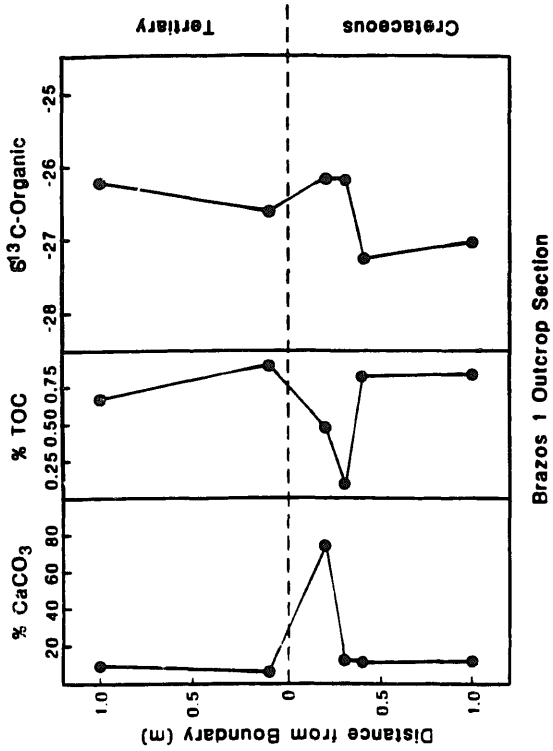


Fig. 1. Comparisons of percentages of calcium carbonate and total organic carbon (TOC) and of $\delta^{13}\text{C}$ -values of organic carbon in four K/T boundary sections. Organic carbon concentrations in samples from DSDP Site 577 are below the 0.05% limit of detection and are not available for presentation (Simoneit and Beller, 1985). A small shift to lighter organic carbon isotopic values appears at the base of the Danian in all four profiles, but none of the isotopic patterns shows the well-developed shift to lighter values typical of the carbonate carbon record.

centrations of 1–3% probably contain substantial proportions of detrital, land-derived material, although this supposition has not been tested. Both the Brazos and the Site 605 samples had low concentrations of organic carbon, <1% (Fig. 1). The lowest percentage measured in the Brazos samples was 0.1% in a limestone layer below the K/T boundary. All of the Site 605 samples contained between 0.1% and 0.3% organic carbon. Attempted measurements of organic carbon in the Site 577 samples showed concentrations to be below the 0.05% limit of detection (Simoneit and Beller, 1985). The general combination of low concentrations of organic carbon and of bacterial biomarker molecules implies that the organic matter in the K/T boundary sections has been heavily reworked by sedimentary microbes (Meyers and Simoneit, 1990).

Organic carbon $\delta^{13}\text{C}$ -values (Fig. 1) do not show the consistent and dramatic shift from heavier ratios in Maastrichtian sediments to lighter ones in Danian deposits that appears in the carbonate carbon record (e.g., Shackleton and Hall, 1984; Keller and Lindinger, 1989; Magaritz, 1989; Zachos et al., 1989; Barrera and Keller, 1990). A small shift to a lighter isotopic ratio is present across the boundary at all four locations, but only when samples from immediately below and above the boundary are compared. When averaged Maastrichtian and Danian values are used, the Cretaceous samples from the Moreno Formation and from Brazos are actually lighter than their Tertiary counterparts. The contrast between the well-developed inorganic carbon isotope shift and the muddled organic carbon isotope patterns indicates that the multiple influences on organic carbon isotopic compositions confuse the sedimentary record.

Comparison of the data from the four sites investigated in this study to published data confirms that the organic carbon isotope story is a complicated one (Table 1). At seven of the eight locations for which measurements are available, a shift occurs from heavier Maas-

trichtian values to lighter values in the Danian. In the K/T section from Hokkaido, Japan, however, the shift is opposite (Saito et al., 1986). Furthermore, the measured $\delta^{13}\text{C}$ -values at Sumbar, Turkmenia, are lighter than at the other locations and the amount of the shift to a lighter ratio is greater (Wolbach et al., 1990). These differences imply that the isotopic ratio of the marine and atmospheric inorganic carbon reservoirs cannot be the only control on variations in the isotopic composition of organic matter deposited around the K/T boundary.

A general characteristic of nearly all K/T boundary occurrences is a depositional hiatus. Sea-level lowering has been postulated to have contributed to this hiatus. Changes in texture between Maastrichtian and Danian sediments at DSDP Site 605 on the New Jersey continental slope, for example, have been interpreted to indicate eustatically lowered sea-level in the early Danian (van Hinte et al., 1987). The type of organic matter deposited in sediments at marine locations close to shore or in shallow water would be modified by sea-level regression. Although an increase in the proportion of organic matter derived from land plants would almost certainly accompany lowered sea-level, the isotopic consequence of this change at a given locality is hard to predict. Modern isotopic compositions average $\sim 7\text{‰}$ lighter in C_3 land plants than in marine algae. Modern intertidal salt marshes, however, are dominated by C_4 plants, which average $\sim 7\text{‰}$ heavier than algal organic matter. Marsh contributions of organic matter to marine sediments are usually minor, but during times of lowered sea-level they might locally become important.

Microfossil assemblages in the sedimentary records indicate major differences between marine phytoplankton and zooplankton communities living before and after the K/T boundary. The record at DSDP Site 577 on the Shatsky Rise, for example, shows a rich Mesozoic nannoplankton assemblage which abruptly terminates at the K/T boundary and is

TABLE 1

Summary of isotopic shifts found in organic carbon at K/T boundaries

Location	Isotopic shift (‰ vs. PDB)	Source
York Canyon (Raton Basin, New Mexico, U.S.A.)	lighter (−27.3 to −25.8)	Schimmelmann and DeNiro (1984)
Stevns Klint (Denmark)	lighter (−26.6 to −25.4)	Hansen et al. (1986)
Sumbar (Turkmenia)	lighter (−25.8 to −22.8)	Wolbach et al. (1990)
DSDP Site 605 (W. North Atlantic)	lighter (−26.4 to −25.8)	this paper
Moreno Fm. (California, U.S.A.)	lighter (−27.3 to −26.4)	this paper
DSDP Site 577 (NW Pacific)	lighter (−26.7 to −25.7)	this paper
Brazos River (Texas, U.S.A.)	lighter (−26.6 to −26.1)	this paper
Hokkaido (Japan)	heavier to (−27.9 to −23.8)	Saito et al. (1986)

replaced by a Tertiary flora (Wright et al., 1985). Because different organisms have different biochemical constitutions and fractionate carbon isotopes to varying degrees, changes in the species compositions of planktonic communities potentially impact the isotopic signatures of the sedimentary organic matter residues of these communities. This factor would influence the isotopic records of both coastal and open-ocean locations, and its consequence would depend on the Maastrichtian and Danian populations present at each locality.

Marine paleoenvironments have provided nearly all the sedimentary records of the K/T boundary event. However, the western edge of the North American Cretaceous seaway contained a number of freshwater marshes at the end of the Maastrichtian and occurrences of the K/T boundary event are recorded at these locations. Schimmelmann and DeNiro (1984) measured the carbon, nitrogen and hydrogen isotopic compositions of organic matter in rocks from below and above the K/T boundary exposure located in the York Canyon of the Raton Basin, New Mexico, U.S.A. They found a shift of 1.8‰ to lighter organic carbon isotopic values in the early Tertiary, which parallels the shift of ~1‰ found in marine carbonates. In contrast to the systematic change in

carbon isotopes, $\delta^{15}\text{N}$ -values vary randomly between +2 and +4‰ in samples from the York Canyon site. Also, δD ratios remain between −125 and −105‰, suggesting no major fluctuations in the water balance or the temperature of this swampy freshwater setting. The absence of recorded hydrologic or temperature perturbations at this locality suggests that regional climate was not perturbed during this period of global change.

4. Summary and conclusions

The carbon isotopic composition of organic matter deposited before and after the K/T boundary has been compared at eight locations. The paleoenvironments of these locations include a freshwater swamp, coastal marine settings and an open-ocean site. Published carbonate-carbon isotope ratios from two of these locations reveal a shift to lighter $\delta^{13}\text{C}$ -values which has been found at other locations and which appears to be a global pattern. This shift is believed to represent a period of depressed biological productivity in the early Danian which allowed organic matter to be remineralized faster than it was biosynthesized.

The consequent return of isotopically light carbon to the inorganic carbon reservoir should have resulted in a shift to lighter carbon isotope ratios in organic matter. This is found at seven of the eight locations compared in this study, but it is weakly developed in comparison to the available inorganic carbon shifts. The difference in the inorganic and organic carbon patterns leads to these conclusions:

(1) Local changes in the proportions of continental and marine contributions of organic matter to sediments may have changed in response to lowered sea-level in the early Danian and complicated the organic carbon isotope record.

(2) Local changes in species compositions of marine and continental flora may have overprinted and masked the change in global carbon isotope composition.

(3) The global isotopic record of organic carbon is more complex than the inorganic carbon record, primarily because of local differences in the biosynthetic pathways, biochemical constitutions and species compositions that influence the organic matter which is contributed to the sedimentary record.

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