Evidence for a small (~0.000 030) but resolvable increase in seawater $^{87}Sr/^{86}Sr$ ratios across the Cretaceous-Tertiary boundary

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ABSTRACT

Previous studies of $^{87}Sr/^{86}Sr$ patterns across the Cretaceous-Tertiary (K-T) boundary have generated inconsistent results. Analyses of samples from Ocean Drilling Program Hole 1049C provide better taphonomic and diagenetic control than has been previously achieved and indicate (1) that there was a rapid increase of ~0.000 030 in seawater $^{87}Sr/^{86}Sr$ ratios across the K-T boundary, (2) that post–K-T Cretaceous foraminifera at this site are reworked, and (3) that subtle diagenetic overprinting affects the basal ~15 cm of the Danian ooze. These conclusions are consistent with the asteroid impact hypothesis. Reworking rather than survivorship confirms nearly complete extinction of Cretaceous Tethyan planktic foraminifera; the $^{87}Sr/^{86}Sr$ excursion can be explained by enhanced continental weathering, perhaps related to acid rain in the aftermath of the K-T impact.

Keywords: Cretaceous-Tertiary boundary, reworking, foraminifera, extinction, strontium, chemostratigraphy.

INTRODUCTION

The reality of the Cretaceous-Tertiary (K-T) Chicxulub impact is generally accepted, but its paleoenvironmental and paleoceanographic consequences continue to be debated (e.g., Norris et al., 1999; Pardo et al., 1999). As an indicator of changing fluxes to the oceans and as a chemostratigraphic signal, $^{87}Sr/^{86}Sr$ ratios could help address outstanding questions. Changes in $^{87}Sr/^{86}Sr$ ratios across the K-T boundary were cited as support for the impact hypothesis even before the Chicxulub crater was discovered (e.g., Macdougall, 1988; Martin and Macdougall, 1991) and have subsequently been used to estimate effects of the impact on continental weathering (Vonhof and Smit, 1997). In terms of biostratigraphy, $^{87}Sr/^{86}Sr$ results were used to argue that post–K-T Cretaceous foraminifera in a controversial high-latitude locality were best explained by reworking, not by survivorship (MacLeod and Huber, 1996). However, the reported pattern of $^{87}Sr/^{86}Sr$ change across the boundary varies considerably among studies (see summary in McArthur et al., 1998), and this uncertainty has limited insights based on $^{87}Sr/^{86}Sr$ results. Pardo et al. (1999) stressed that trans–K-T reworking has been tested with $^{87}Sr/^{86}Sr$ at only one site. McArthur et al. (1998), who reported no resolvable change in seawater $^{87}Sr/^{86}Sr$ ratios across the K-T boundary, stated that previous evidence for steps or spikes in $^{87}Sr/^{86}Sr$ values were analytical or diagenetic artifacts.

To better document seawater $^{87}Sr/^{86}Sr$ evolution across the K-T boundary and to test the fidelity of fossil distributions, we measured the $^{87}Sr/^{86}Sr$ ratios in separates from Ocean Drilling Program (ODP) Site 1049 (Fig. 1A). Site 1049 provides samples in which K-T $^{87}Sr/^{86}Sr$ ratios can be documented at a higher resolution and with better diagenetic and taphonomic control than has previously been achieved. The K-T boundary in Hole 1049C is represented by 10 cm of spherulitic clay capped by a millimeter-scale limonitic layer (Fig. 1B). Maastrichtian and Danian strata on either side of the spherulitic clay are dominantly homogeneous pelagic ooze containing abundant, well-preserved foraminifera, but the basal 5–7 cm of Danian ooze is distinctly mottled and grades upward into ~10 cm of white ooze. The remainder of the Danian interval studied is light gray and homogeneous. Microfossils below the spherulitic clay are typical of late Maastrichtian Tethyan assemblages, whereas Danian samples contain both Danian and Maastrichtian taxa.

MATERIALS AND METHODS

Bulk samples of ooze from 14 horizons spanning the interval from ~2.5 m below to ~1 m above the boundary were collected and processed by using standard techniques. Foraminifera were picked from the coarse residue. Taxa that first appeared below the K-T boundary (referred to herein as Cretaceous taxa) occurred in sufficient numbers for isotopic measurements in all 14 samples, whereas taxa that first appear in the Danian (herein, Tertiary taxa) occurred in sufficient numbers for analysis in the upper 8 of the 9 Danian samples. Picked separates contained a mixed assemblage of planktic species, the proportion of taxa varying among samples. Foraminifera were ultrasonically cleaned and then screened under a binocular microscope. Depending on the size of individuals present, 30–300 (~0.5–3 mg) of the best-preserved specimens from each separate were selected for analysis. In addition, two bulk-sediment samples from the spherulitic layer and one separate of dolomite rhombs from the white interval were analyzed.

Carbonate separates were dissolved in 1.7 *M* acetic acid and centrifuged; the solution was removed. Bulk samples from the clay layer were processed by using a two-stage dissolution. Samples were leached with 1.7 *M* acetic acid for ~2 h until visible reaction stopped. The leachate (assumed to represent carbonate fractions) was removed, and the residue (assumed to represent silicate fraction) was dissolved in HF. Sr was separated using EiChrom SrSpec resin and analyzed on a VG Sector 54 thermal ionization mass spectrometer.

RESULTS

The $^{87}Sr/^{86}Sr$ ratios of Cretaceous foraminifera, of Tertiary foraminifera from the gray interval, and of Tertiary foraminifera from the

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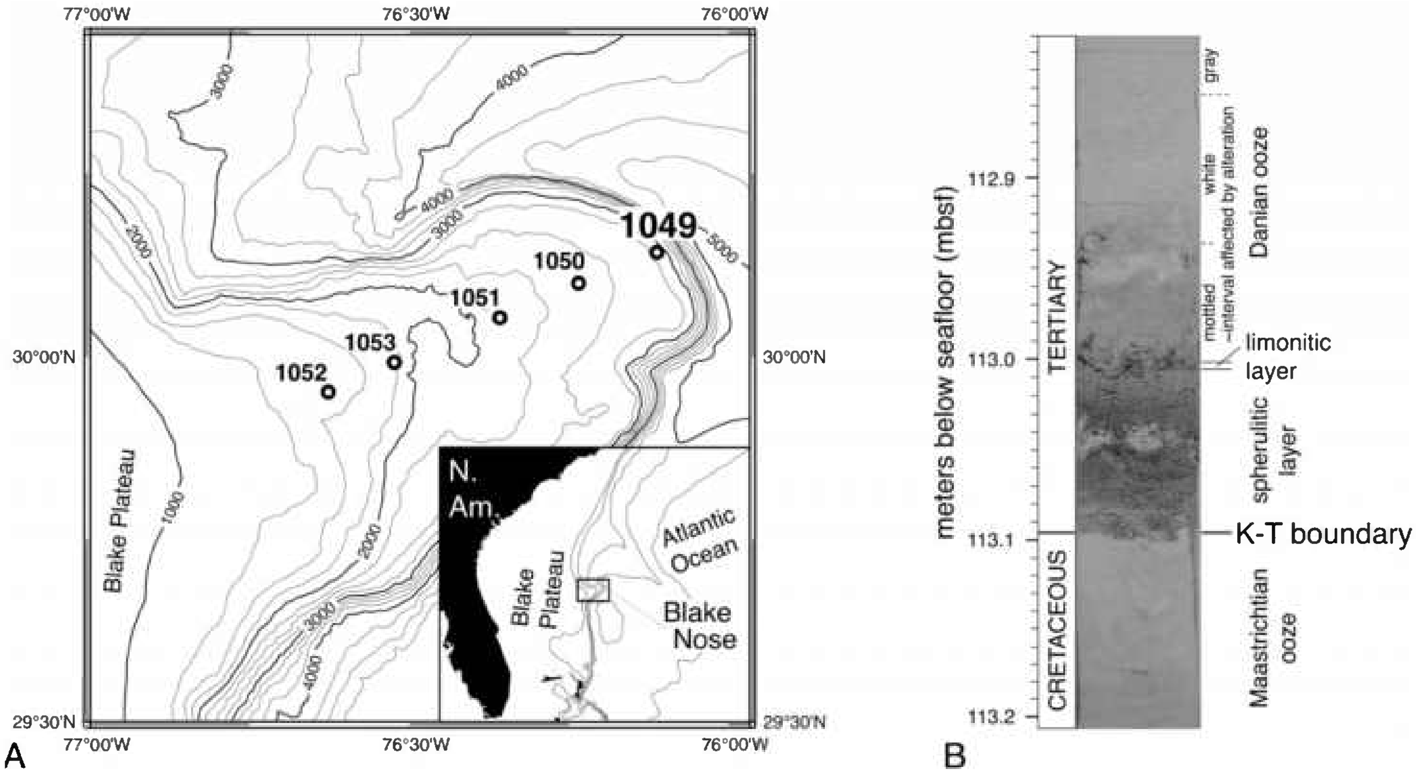

Figure 1. A: Topographic map of Blake Nose showing Site 1049 relative to other Leg 171B sites. Position of Blake Nose relative to southeast coast of North America is shown in inset. B: Cretaceous-Tertiary (K-T) boundary interval in Hole 1049C showing layers discussed in text.

mottled and white intervals seem to represent three geochemical populations (Table 1; Fig. 2). Two samples (Cretaceous taxa from core 1049C, section 8X-5, 60–62 cm and Tertiary taxa from core 1049C, section 8X-5, 80–80.5 cm) yielded anomalous results; both measurements were repeated using independently processed bulk material from the same level. The anomalous Cretaceous sample was small and an insoluble residue remained after dissolution. The replicate sample left no residue and yielded a value matching other Cretaceous values. We conclude that this second analysis provides a better estimate of the $^{87}Sr/^{86}Sr$ value of Cretaceous foraminifera in core 1049C, section 8X-5, 60–62 cm because the sample was larger, dissolution was cleaner, and the value was consistent with 13 other analyses of Cretaceous taxa. The anomalous Tertiary separate was unusual only in that it exhibited a higher value than the two adjacent Tertiary separates. The replicate analysis yielded an even higher value, suggesting that Tertiary foraminifera within the mottled and white intervals have relatively variable $^{87}Sr/^{86}Sr$ ratios.

Values for Tertiary foraminifera from the gray interval (average $0.707\,884 \pm 0.000\,009$, 2 σ standard error [s.e.]) are statistically distinct ($p < 0.001$, t-test for unpaired samples, two tails) from the values for Cretaceous foraminifera regardless of whether the anomalous Cretaceous value is excluded (average $0.707\,856 \pm 0.000\,005$, 2 σ s.e.) or included (average $0.707\,858 \pm 0.000\,006$, 2 σ s.e.). There is no statistical difference between separates of Cretaceous foraminifera from above and below the boundary ($p > 0.75$, t-test for unpaired samples, two tails).

Tertiary foraminifera from the mottled and white intervals exhibit $^{87}Sr/^{86}Sr$ values that are relatively variable (average $0.707\,840 \pm 0.00\,0041$, 2 σ s.e.). Small overgrowths (~1 μm) occur on the inner walls of many foraminifera in the lower ~15 cm of Danian ooze, and these overgrowths are rare to absent both higher and lower in the section (Fig. 3). Dolomite rhombs from sample 1049C, 8X-5, 80–80.5 cm (in the white interval) yield a $^{87}Sr/^{86}Sr$ value similar to the low Tertiary values. Carbonate values from within the spherulitic layer are slightly to markedly higher than values in Cretaceous foraminifera. Silicate separates from this layer have $^{87}Sr/^{86}Sr$ ratios much higher than any other fractions analyzed.

DISCUSSION

Seawater $^{87}Sr/^{86}Sr$ curves provide clues regarding the nature, size, and duration of perturbations to the oceanic Sr budget. Sr isotopes in seawater are controlled by the relative importance of riverine input of weathered continental material (high $^{87}Sr/^{86}Sr$ ratios) and hydrothermal alteration of basalt (low $^{87}Sr/^{86}Sr$ ratios) moderated by the deposition and dissolution of marine carbonates ($^{87}Sr/^{86}Sr$ ratios reflecting former seawater values) (e.g., Hodell, 1994; McArthur, 1994). Because Sr has a long residence time (2–4 m.y.) relative to mixing time of the oceans (~10^3 yr), seawater Sr is isotopically homogeneous at any given time (e.g., Hodell, 1994; McArthur et al., 1998). Thus, shifts in the seawater $^{87}Sr/^{86}Sr$ ratio should be geologically synchronous on a global scale and detectable in any samples that preserve seawater values. Intrinsic factors (e.g., orogeny, seafloor spreading) influencing the oceanic Sr budget generally vary slowly, and their effects appear as gradual changes in the seawater $^{87}Sr/^{86}Sr$ curve. A catastrophic event (e.g., an impact) could have effects large enough and rapid enough to appear as a step in the seawater curve. The seawater reservoir is large, however, and difficult to perturb at a level that can be detected, especially if there are diagenetic or stratigraphic complications (e.g., Vonhof and Smit, 1997).

Comparisons among co-occurring taxa can be used to monitor diagenesis and simultaneously test for reworking (MacLeod and Huber, 1996). Individuals that lived at the same time would have secreted tests with the $^{87}Sr/^{86}Sr$ ratio of contemporary seawater. Reworking could mix populations with distinct $^{87}Sr/^{86}Sr$ ratios, or diagenetic alteration could shift values away from the initial seawater value. Thus, if foraminiferal separates from the same sample have different $^{87}Sr/^{86}Sr$ ratios, either the separates represent individuals that did not live together (i.e., one or both separates contains reworked individuals) or initial

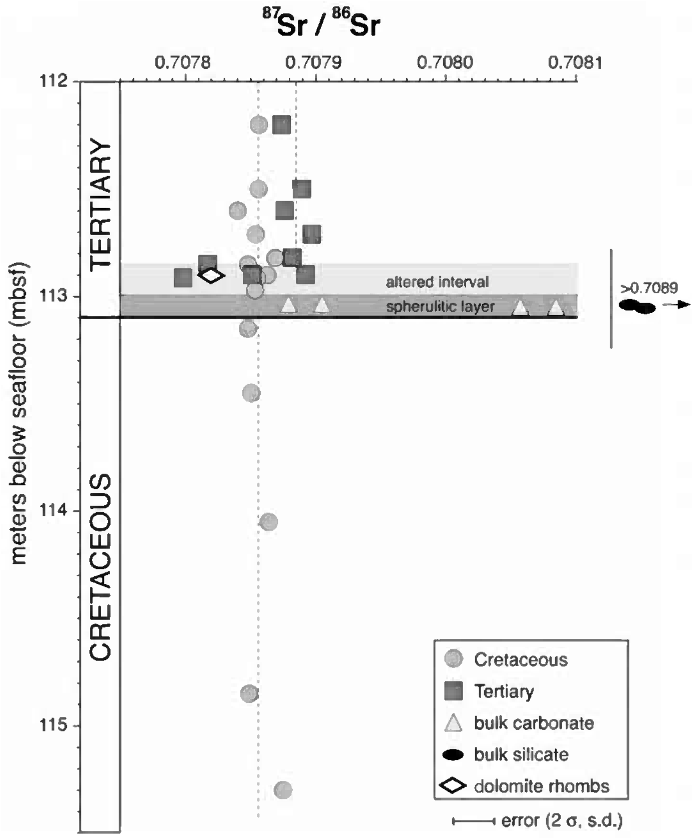

Figure 2. $^{87}Sr/^{86}Sr$ ratios in separates from Cretaceous-Tertiary boundary interval in Ocean Drilling Program Hole 1049C. Error bar is twice standard deviation of results from 22 analyses of NIST 987 run with separates from Hole 1049C. Vertical dashed lines represent average of Tertiary taxa from gray interval and of Cretaceous taxa from across entire interval discussed. Difference between averages is highly significant ($p < 0.001$, t-test for unpaired samples, two tails), and there is no statistical difference between values for Cretaceous taxa from below and from above boundary ($p > 0.75$, t-test for unpaired samples, two tails). Anomalous value for Cretaceous taxa from 112.71 mbsf is not plotted, but inclusion or exclusion of this point does not affect results.

TABLE 1. $^{87}Sr/^{86}Sr$ RESULTS FOR THE CRETACEOUS-TERTIARY BOUNDARY INTERVAL IN HOLE 1049C

Sample level*	Depth†	Type‡	$^{87}Sr/^{86}Sr$§	Std Err (%)
Danian (gray interval)				
1049C, 8X, 5, 10–12 cm	112.20	K	0.707 856	0.0007
1049C, 8X, 5, 10–12 cm	112.20	T	0.707 874	0.0006
1049C, 8X, 5, 40–41 cm	112.50	K	0.707 856	0.0007
1049C, 8X, 5, 40–41 cm	112.50	T	0.707 889	0.0007
1049C, 8X, 5, 50–52 cm	112.60	K	0.707 840	0.0007
1049C, 8X, 5, 50–52 cm	112.60	T	0.707 876	0.0007
1049C, 8X, 5, 60–62 cm	112.71	K	0.707 886	0.0007
1049C, 8X, 5, 61–62 cm	112.71	K	0.707 854	0.0007
1049C, 8X, 5, 61–62 cm	112.71	T	0.707 897	0.0005
1049C, 8X, 5, 72–72.5 cm	112.82	K	0.707 869	0.0007
1049C, 8X, 5, 72–72.5 cm	112.82	T	0.707 882	0.0010
Danian (white and mottled intervals)				
1049C, 8X, 5, 75–75.5 cm	112.85	K	0.707 848	0.0008
1049C, 8X, 5, 75–75.5 cm	112.85	T	0.707 817	0.0007
1049C, 8X, 5, 80–80.5 cm	112.90	d	0.707 820	0.0012
1049C, 8X, 5, 80–80.5 cm	112.90	K	0.707 863	0.0006
1049C, 8X, 5, 80–80.5 cm	112.90	T	0.707 892	0.0011
1049C, 8X, 5, 80–80.5 cm	112.90	T	0.707 851	0.0005
1049C, 8X, 5, 81.5–82 cm	112.92	K	0.707 855	0.0007
1049C, 8X, 5, 81.5–82 cm	112.92	T	0.707 798	0.0007
1049C, 8X, 5, 87–87.5 cm	112.97	K	0.707 854	0.0007
Danian (spherulitic clay)				
1049C, 8X, 5, 93.5–94 cm	113.04	c	0.707 879	0.0008
1049C, 8X, 5, 93.5–94 cm	113.04	c	0.707 905	0.0006
1049C, 8X, 5, 93.5–94 cm	113.04	s	0.708 958	0.0008
1049C, 8X, 5, 95–95.5 cm	113.05	c	0.708 085	0.0007
1049C, 8X, 5, 95–95.5 cm	113.05	c	0.708 058	0.0013
1049C, 8X, 5, 95–95.5 cm	113.05	s	0.709 160	0.0007
Maastrichtian (ooze)				
1049C, 8X, 5, 105–106 cm	113.15	K	0.707 848	0.0006
1049C, 8X, 5, 135–137 cm	113.45	K	0.707 851	0.0008
1049C, 8X, 6, 45–47 cm	114.05	K	0.707 864	0.0006
1049C, 8X, 6, 125–127 cm	114.85	K	0.707 849	0.0006
1049C, 8X, 7, 20–22 cm	115.30	K	0.707 875	0.0007

*Samples are grouped into lithologic divisions as discussed in text; age is based on placement of the Cretaceous-Tertiary (K-T) boundary at base of spherulitic layer.

†Depth in meters below seafloor.

‡K = foraminifera that first appear in the Cretaceous, T = foraminifera that first appear in the Tertiary, d = dolomite rhombs, c = acetic acid leach of bulk sediment (assumed to represent bulk carbonate), s = material insoluble in acetic but soluble in HF (assumed to represent bulk silicate).

§$^{87}Sr/^{86}Sr$ ratios are normalized to $^{86}Sr/^{88}Sr$ ratio of 0.1194. Four or five National Institute of Standards and Technology (NIST) 987 $SrCO_3$ standards were analyzed for $^{87}Sr/^{86}Sr$ with each turret of samples; 0.710 250 was accepted as the correct $^{87}Sr/^{86}Sr$ ratio for this standard, and the $^{87}Sr/^{86}Sr$ value reported for each sample was adjusted by the difference between the average for the analyses of NIST 987 in that turret and 0.702 50. Analytical error based on twice standard deviation of results for NIST 987 run during this study (average = 0.710 248, n = 22) is ±0.000 015.

ratios have been partially altered (pervasive diagenesis would result in convergence of ratios in all components on the diagenetic value).

Within the mottled and white intervals, the differences in $^{87}Sr/^{86}Sr$ values between Tertiary and Cretaceous foraminifera (Fig. 3) are attributed to diagenesis. Diagenetic Sr is likely hosted in the carbonate overgrowths observed on the inner walls of many foraminifera, although partial alteration of test walls cannot be ruled out. Both Tertiary and Cretaceous taxa show overgrowths, but Tertiary individuals in this interval are much smaller and thinner walled than co-occurring Cretaceous individuals (Fig. 3). A thin veneer of secondary carbonate (or partial alteration propagating from the tests' surfaces) should shift the isotopic ratios of small, delicate Tertiary tests more than it would large, robust Cretaceous tests. Neither bulk carbonate nor bulk silicate values from the clay layer are low, so if the clay layer was the source of diagenetic Sr, the source phase does not dominate the bulk ratios of the clay layer today. The $^{87}Sr/^{86}Sr$ value for dolomite rhombs, however,

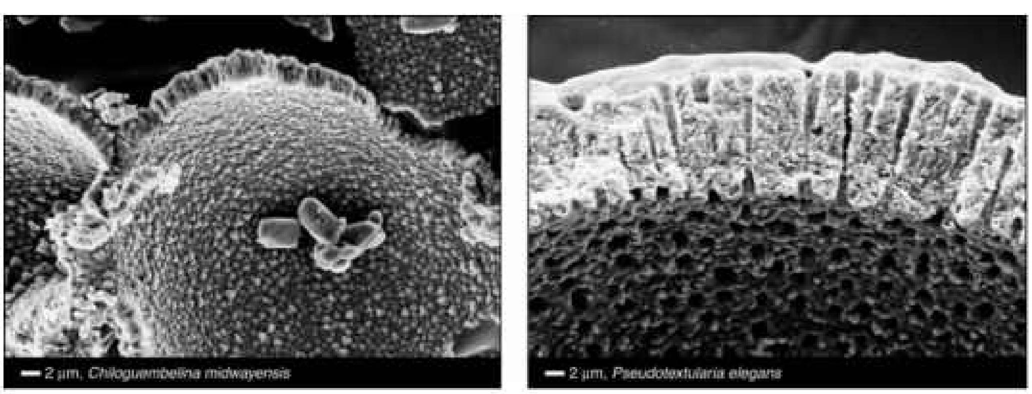

Figure 3. Scanning electron microscope images at same magnification of *Chiloguembelina midwayensis* (Tertiary taxon) and *Pseudotextularia elegans* (Cretaceous taxon) from 112.99 m below seafloor showing scale of diagenetic overgrowths in altered interval relative to test thickness of typical Tertiary and Cretaceous individuals.

confirms that diagenetic fluids with low $^{87}Sr/^{86}Sr$ ratios were present. Mottles suggest that bioturbation-induced heterogeneity (grain size and/or composition) exists on millimeter-length scales in this interval and could affect alteration and explain the variability in $^{87}Sr/^{86}Sr$ ratios observed among Tertiary foraminifera in this interval.

The difference between Tertiary and Cretaceous foraminifera from the gray interval, however, is attributed to an increase in seawater $^{87}Sr/^{86}Sr$ ratio of ~0.000 030 across the boundary, with the Cretaceous foraminifera having been reworked into Tertiary sediments from the upper Maastrichtian. Diagenesis is not a likely explanation for elevated Tertiary values because (1) scanning electron microscope images show minor to absent overgrowths in the gray interval, (2) fluids from which diagenetic carbonates grew appear to have had low $^{87}Sr/^{86}Sr$ ratios, (3) Tertiary foraminifera analyzed from the gray interval are approximately the same size and mass as Cretaceous individuals analyzed, and (4) Tertiary and Cretaceous separates exhibit very consistent but slightly offset values, a pattern that is difficult to reconcile with partial alteration. Point 4 may not be intuitive: alteration, to explain results, would have had to affect some individuals more than others (otherwise Cretaceous and Tertiary separates would all have the same diagenetic ratio). To yield the constant Cretaceous and Tertiary values observed, that differential alteration would have needed to have had a constant effect when averaged across the various individuals and taxa present in each Tertiary and each Cretaceous separate analyzed. Such a coincidence seems unlikely.

The long residence time of Sr in the oceans minimizes uncertainty in our estimate of the size of the K-T $^{87}Sr/^{86}Sr$ excursion. In Hole 1049C, as in all deep-sea sites, the P0 foraminiferal zone is missing. This absence may reflect missing time (MacLeod and Keller, 1991), restriction of the zone P0 assemblage to shallow paleoenvironments (Norris et al., 1999), or bioturbation mixing Pα taxa into P0 strata. However, because alteration has obscured the initial $^{87}Sr/^{86}Sr$ ratios of Tertiary foraminifera in the lower ~15 cm of Danian ooze, arguments regarding biostratigraphic completeness of the basal Danian are irrelevant to this study. The critical consideration is the minimum age of the oldest Danian samples recording seawater $^{87}Sr/^{86}Sr$ ratios. The lowest sample above the altered interval (112.82 mbsf [meters below seafloor]) is just below the first occurrence of *Globoconusa daubjergensis* (112.81 mbsf) and thus is in the lower ~15% of zone Pα as estimated from the record in Deep Sea Drilling Project (DSDP) Holes 528 and 577 (D'Hondt and Keller, 1991). The top of zone Pα projects to a time ~10^5 yr after the K-T boundary (Berggren et al., 1995), suggesting that the temporal gap in our seawater $^{87}Sr/^{86}Sr$ record is 10^4 yr. If there were a step increase at the boundary, the maximum reasonable change in $^{87}Sr/^{86}Sr$ values across the missing interval would occur if the post-step value decayed exponentially to the prestep value (e.g., Martin and Macdougall, 1991; Nelson et al., 1991; McArthur et al., 1998). Assuming a Sr residence time of 2 m.y., a 0.000 030 excursion in $^{87}Sr/^{86}Sr$ would decay by $<<$0.000 001 in 10^4 yr and $<$0.000 002 in 10^5 yr. Thus, even with conservative assumptions (i.e., long temporal gap, short residence time), the size of the K-T step in $^{87}Sr/^{86}Sr$ ratios we found (~0.000 030) is unlikely to have been significantly truncated by an incomplete record.

IMPLICATIONS

Data from Hole 1049C show a small but resolvable positive step (~0.000 030) in seawater $^{87}Sr/^{86}Sr$ ratios across the K-T boundary, suggesting sudden (occurring in 10^4 yr or less) perturbation to the ocean Sr budget. This result contradicts the finding that there was no K-T excursion (McArthur et al., 1998) and reduces by ~50% the size of the excursion estimated by Vonhof and Smit (1997). The size of the excursion in Hole 1049C is similar to the excursion estimated by Martin and Macdougall (1991) and Nelson et al. (1991), but those studies had relatively poor control on potential diagenetic or stratigraphic artifacts. The resolvable difference between earliest Danian and latest Maastrichtian seawater $^{87}Sr/^{86}Sr$ ratios provides an independent chemostratigraphic test of reworking. Because all separates of Cretaceous taxa from above the boundary have $^{87}Sr/^{86}Sr$ ratios statistically indistinguishable from late Maastrichtian values but distinct from values of Tertiary taxa, it is likely that there were few, if any, planktic foraminiferal survivors of the K-T event at Blake Nose.

Enhanced weathering due to acid rain, especially leaching of Sr with high $^{87}Sr/^{86}Sr$ ratios from soils, seems to be the most plausible explanation for the observed excursion. Following the calculations of Vonhof and Smit (1997), a shift of ~0.000 030 in seawater $^{87}Sr/^{86}Sr$ would result if all the Sr were flushed from the upper 1.2 m of the world's soils (9.2×10^{14} mol of Sr at an average $^{87}Sr/^{86}Sr$ ratio of 0.712). The potential contribution from complete dissolution of Sr in material excavated from the Chicxulub crater or soot from global wildfires is too small by one and more than two orders of magnitude, respectively, to have caused the observed shift. Vonhof and Smit (1997) invoked a climatic change correlative with impact-induced acid rain because the ~0.000 060 shift in $\leq 10^4$ yr they reported seemed otherwise too large to explain. The smaller excursion inferred from Hole 1049C data thus strengthens the weathering model (Vonhof and Smit, 1997) by relaxing corollary paleoclimatic requirements.

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