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High-resolution late Maastrichtian–early Danian oceanic $^{87}Sr/^{86}Sr$ record: Implications for Cretaceous-Tertiary boundary events

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ABSTRACT

A high-resolution late Maastrichtian–early Danian seawater $^{87}Sr/^{86}Sr$ reference curve is constructed from two Cretaceous-Tertiary boundary (K-T boundary) sections: Bidart (France) and El Kef (Tunisia). The $^{87}Sr/^{86}Sr$ curve shows maxima at 0.3–0.4 Ma before the K-T boundary and at the K-T boundary. The first maximum could mark the onset of a major outflow of the Deccan Traps. The second maximum, a rapid 0.000 06 $^{87}Sr/^{86}Sr$, shift, extends from ~3–4 m below to ~1 m above the K-T boundary. This profile probably results from diagenetic smoothing of an originally sharp K-T boundary $^{87}Sr/^{86}Sr$ anomaly, rather than from a gradual process. The sharp shift could result from (1) the vaporization of the Chicxulub target rocks, (2) global wildfires, and (3) acid-rain leaching of soils and sialic surface rocks. Of these three possibilities, only Sr release by soil leaching combined with increased rainfall associated with the K-T event appears to be sufficiently large to produce the observed K-T $^{87}Sr/^{86}Sr$ anomaly.

INTRODUCTION

A short-term positive $^{87}Sr/^{86}Sr$ anomaly at the Cretaceous-Tertiary (K-T) boundary was first recognized by Hess et al. (1986) in a reference curve spanning the Cretaceous-Tertiary transition. Several investigations have since been conducted, further exploring the $^{87}Sr/^{86}Sr$ anomaly. The resulting conclusions vary strongly. Some do not observe the $^{87}Sr/^{86}Sr$ anomaly (Jones et al., 1987; McArthur, 1994), and others differ in opinion about its magnitude and position with regard to the K-T boundary (Nelson et al., 1991; Hess et al., 1986; Koepnick et al., 1988; Macdougall, 1988; Martin and Macdougall, 1991; Meisel et al., 1995). Several Sr sources have been proposed to explain this anomaly, such as impact ejecta, the soot from global impact-generated fires, and increased continental weathering by acid rain precipitation.

To evaluate these different possibilities, we have analyzed carbonate Sr isotopic ratios for two continuous late Maastrichtian–early Danian sections with a high sedimentation rate across the K-T boundary,[1] Bidart (France) (Nelson et al., 1991) and El Kef (Tunisia) (Salaj, 1980).

Because a previous $^{87}Sr/^{86}Sr$ stratigraphic study of the Bidart cliff section by Nelson et al. (1991) may have been hampered by some small faults and slumping in the topmost Maastrichtian, we resampled the section in outcrops on the abrasion plain. We were able to reconstruct faults and avoid the slumped interval, allowing us to tie the well constrained Milankovitch-type cyclostratigraphy of the Biscaye region to our sample set (ten Kate and Sprenger, 1993).

In both the El Kef and Bidart sections diagenetic alteration is clearly shown in Electron microscope and cathodoluminescence photographs, and recorded in the elevated Fe and Mn contents of the diagenetic calcite grown in the foram chambers, when compared to the foram tests (Table 1). For analyses, we handpicked, crushed, and ultrasonically cleaned sediment-filled specimens to discard as much of the diagenetic calcite as possible. Clean fragments were dissolved in either 1 *N* hydrochloric acid, or in 5 *N*. acetic acid. Since our pretreatment discarded virtually all clay, and the reaction time was >10 min before centrifuging and isolating the supernatant, we believe to have minimized clay contamination. Tests showed no difference in $^{87}Sr/^{86}Sr$ ratios between splits of samples dissolved in hydrochloric or acetic acid, just like similar tests performed by Nelson et al. (1991). The important interval in the early Tertiary has been treated with both methods, while the Cretaceous interval has predominantly been treated with 1 *N* hydrochloric acid. Since splits of calcite filled foraminifers and thoroughly cleaned sediment-filled foraminifers show no significant difference in

TABLE 1. ELECTRON MICROPROBE DATA OF FORAM TESTS VS. DIAGENETIC CARBONATE

Sample	Mg (ppm)	Mn (ppm)	Fe (ppm)	Sr (ppm)
Bidart forams	938	149	774	1438
Bidart infill calcite	2526	1094	6892	859
El Kef forams	1553	60	1234	1542
El Kef infill calcite	2487	350	12488	1601
Geulhem clay forams	792	46	407	1342

Note: Fe shows an order of magnitude difference between the foram wall and the diagenetic carbonate infill.

TABLE 2. COMPARISON OF Sr ISOTOPIC RATIOS OF WELL PRESERVED VS. POORLY PRESERVED OR CALCITE FILLED PLANKTIC FORAMS

Sample	Sr (ppm)	$^{87}Sr/^{86}Sr$	2 s.e.	Remarks
Kef 131-60	1251	0.707856	0.000008	Well preserved
Kef 131-60	1126	0.707845	0.000010	Poorly preserved
Kef 131-60	1246	0.707842	0.000008	Calcite filled
HVB II-35	N.D.	0.707852	0.000008	Well preserved
HVB II-35	N.D.	0.707839	0.000007	Calcite filled

Note: No significant difference exists for both El Kef and Bidart (HVB samples), indicating diagenetic Sr isotopic ratios close to those of the foram shells. Sr concentrations were analysed by isotope dilution. s.e. is standard error

[1]GSA Data Repository item 9717, the Sr-isotopic data of this study and mass balance calculation, is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301. E-mail: editing@geosociety.org.

Data Repository item 9717 contains additional material related to this article.

$^{87}Sr/^{86}Sr$ ratios (Table 2), we believe that diagenesis did not involve significant Sr redistribution.

Blanks analyzed always contained under 150 pg Sr, and generally under 100 pg; samples contained typically >1 µg Sr.

TIME FRAME

The uppermost Maastrichtian succession of Bidart comprises 68 limestone-marlstone couplets. Assuming these are precession cycles with an average duration of 20.8 k.y., a sedimentation rate of 4.3 cm/k.y. is inferred. At El Kef, estimates for the late Maastrichtian sedimentation rate are based on biostratigraphy and vary considerably. Planktic foraminiferal stratigraphy (A. J. Nederbragt, 1996, personal commun.) results in a sedimentation rate of 4–5 cm/k.y. The last appearance datum (LAD) of Inoceramid body fossils at 190 m compared to their LAD in the Zumaya section (MacLeod and Orr, 1993) indicate 7.5–8 cm/k.y., and the first appearance datum (FAD) of the coccolith *M. Prinsii* (between 26 m and 30 m; J. Pospichal, 1996 personal commun.) indicates a rate of 10–16 cm/k.y. The Danian sedimentation rate of 1.4 cm/k.y. for El Kef is derived from the FAD of *Morozovella pseudobulloides* (at 3.5 m) and *M. trinidadensis* (at 27 m).

MAASTRICHTIAN $^{87}Sr/^{86}Sr$ DATA

If the Bidart time frame is taken as the reference, the El Kef record fits the smoothed Bidart $^{87}Sr/^{86}Sr$ record best at a 8.5 cm/k.y. sedimentation rate (Fig. 1). The detailed correlation of the El Kef and Bidart $^{87}Sr/^{86}Sr$ curves indicates that both sections retained the seawater $^{87}Sr/^{86}Sr$ signal. The combined El Kef–Bidart curve shows increasing $^{87}Sr/^{86}Sr$ ratios in the Maastrichtian, reaching a maximum at 300–400 k.y. before the K-T boundary, followed by decreasing ratios in the topmost Maastrichtian.

Most previous studies have shown a continuous rise of the seawater $^{87}Sr/^{86}Sr$ ratios throughout the Maastrichtian (Hess et al., 1986; McArthur et al., 1992; Sugarman et al., 1995). Two studies also report a late Maastrichtian $^{87}Sr/^{86}Sr$ maximum at 1 m.y. and 2 m.y. before the K-T boundary (Nelson et al., 1991; Martin and Macdougall, 1991). These two studies, however, lack a high-resolution stratigraphic framework.

Martin and McDougall (1991) suggested that Deccan Trap volcanism could have caused the slope break between the steep late Cretaceous $^{87}Sr/^{86}Sr$ curve and the flat curve throughout the early Tertiary. Disregarding the K-T boundary $^{87}Sr/^{86}Sr$ shift (discussed below), our data suggest that this slope break occurs at the late Maastrichtian maximum, 300–400 k.y. before K-T boundary. This correlates well with the magnetostratigraphic age of the major pulse of Deccan Trap outflow, near the 29R/30N chron boundary (Courtillot et al., 1986), ~340 k.y. before the K-T boundary (Berggren, 1985). The rapid outflow of more than 1×10^6 km^3 of basaltic lava ($^{87}Sr/^{86}Sr$ = ~0.705) provided a major source of nonradiogenic Sr to the oceans. The basalt flows covered ~1%–1.5% of the Earth's land surface, which mainly consisted of highly radiogenic (Archean) rocks ($^{87}Sr/^{86}Sr$ = ~0.730). This change of the Indian drainage area may account for a 0.0004 $^{87}Sr/^{86}Sr$ drop of global average runoff. Direct Sr exchange with submarine basalts associated with the Deccan Traps, would decrease oceanic $^{87}Sr/^{86}Sr$ values even more.

K-T $^{87}Sr/^{86}Sr$ DATA

Both El Kef and Bidart record a positive $^{87}Sr/^{86}Sr$ shift just above the K-T boundary. The shift in El Kef reaches a maximum value of 0.707 963; that in Bidart does not exceed 0.707 883. Even when the two highest El Kef $^{87}Sr/^{86}Sr$ values are taken as outliers, the radiogenic $^{87}Sr/^{86}Sr$ shift at the K-T boundary remains significant, the peak $^{87}Sr/^{86}Sr$ values occurring in the first 1 m (~80 k.y.) above the K-T boundary (Fig. 2). In both sections the shift in the $^{87}Sr/^{86}Sr$ ratios is already recorded in the upper few metres of the Maastrichtian. This suggests a gradual change of seawater $^{87}Sr/^{86}Sr$ ratios, starting about 60 k.y. before the K-T boundary, with a maximum duration of ~100–140 k.y.

Because both sections have been diagenetically altered, one can also explain the Maastrichtian part of the $^{87}Sr/^{86}Sr$ shift by diagenetic smoothing of a sharp anomaly, related to the K-T impact event. Comparable smoothing is observed in the Ir distribution across the K-T boundary in El Kef (Rocchia et al., 1992). In both El Kef and Bidart, the detectable vertical diagenetic transport of Danian Sr into the Maastrichtian would then be some 3–4 m. In comparison, in the highly permeable K-T section in Maastricht (The Netherlands), the radiogenic $^{87}Sr/^{86}Sr$ shift starts at 10–15 m below the K-T boundary (Vonhof and Smit, 1996).

Our best-preserved, earliest Danian samples are air-filled foraminifera from the uncompacted K-T boundary clay in the Geulhemmerberg section of

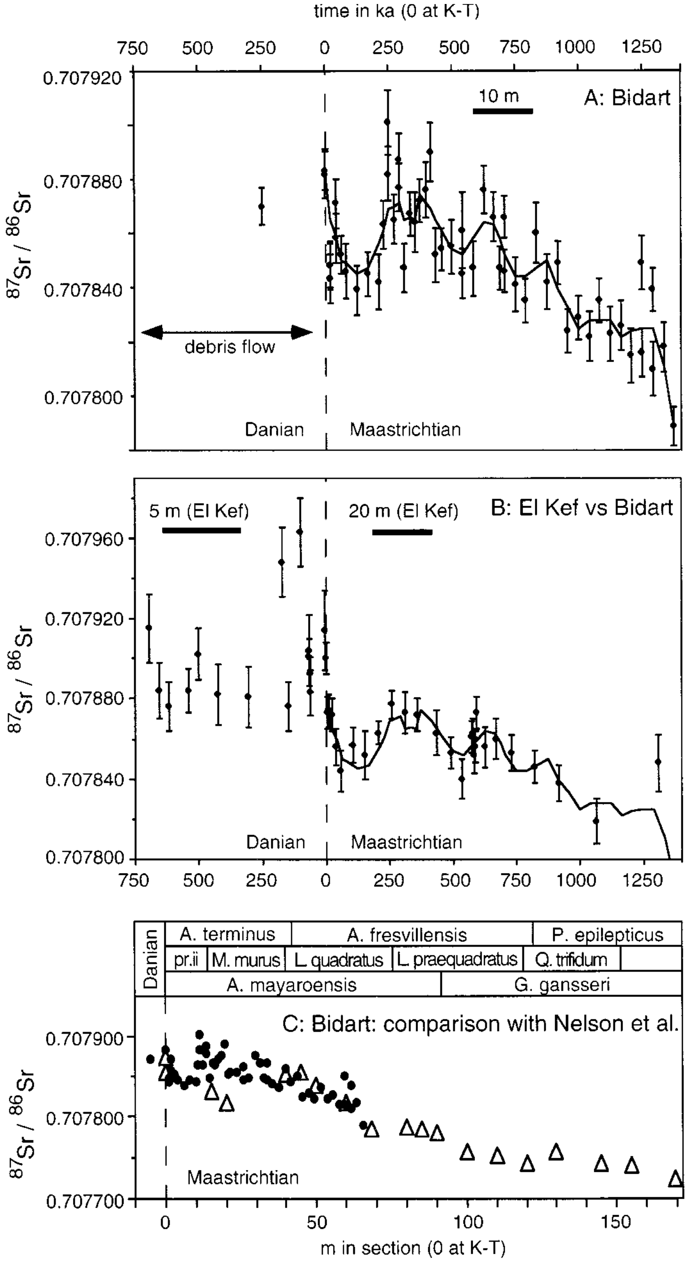

Figure 1: Graphs of $^{87}Sr/^{86}Sr$ data for both sections. A shows Bidart data superimposed on curve of moving average through data (80 k.y. window). Age conversion took place assigning 20.8 k.y. to each precession couplet. B shows best-fit graph of Bidart moving average curve with El Kef $^{87}Sr/^{86}Sr$ data, resulting in an average sedimentation rate of 8.5 cm/k.y. for El Kef. C shows our Bidart data compared to data of Nelson et al. (1991) (triangles), together with the biostratigraphical zonations of ammonites (top), ostracods (middle), and planktic foraminifera (bottom). Sr from handpicked planktic foraminifera and ostracods was separated with standard ion exchange techniques. Analyses on Finnigan 261 mass spectrometer generally yielded a 2 standard error (2 s.e.) of 10×10^{-6} or better. Fifty runs of NBS 987 standard averaged 0.710 275 with standard deviation of 0.000 012. Data are normalized to $^{86}Sr/^{88}Sr$ = 0.1194.

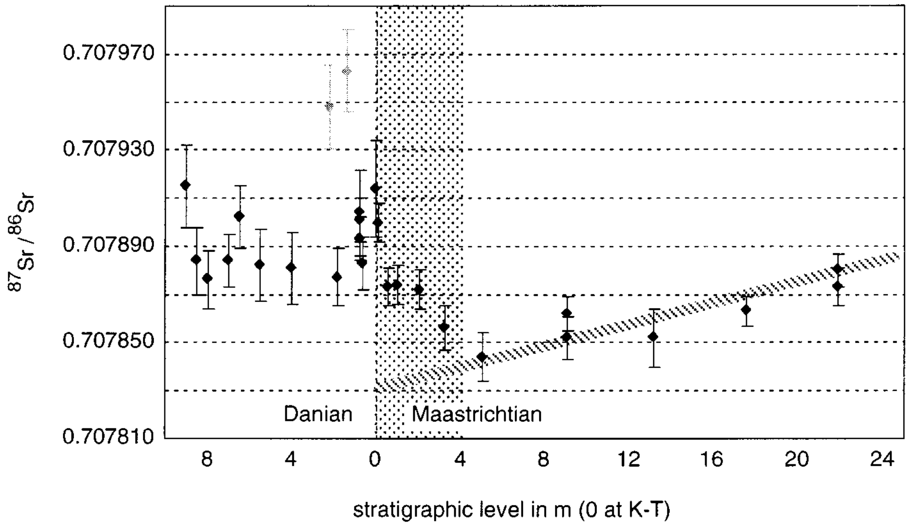

Figure 2: $^{87}Sr/^{86}Sr$ data for K-T interval at El Kef. All Danian samples record higher $^{87}Sr/^{86}Sr$ values than latest Maastrichtian samples. Dashed line drawn through data is inferred seawater Sr curve. Shaded area between 0 and 4 m below K-T, where K-T Sr isotopic shift appears to start, indicates in our interpretation the zone in which diagenetic Sr was transported down from the Danian. Extrapolation of curve in this area represents estimated seawater Sr isotopic ratios for this time interval. Two highest Sr isotopic values in Danian (in gray) are believed to be outliers (see text).

the Maastrichtian stratotype area (Vonhof and Smit, 1996). The 0.707 893 $^{87}Sr/^{86}Sr$ ratio of these foraminifera represents our best estimate of earliest Danian seawater. Because the clay that hosts the forams is relatively radiogenic, we tested the possibility of a highly radiogenic $^{87}Sr/^{86}Sr$ signal in the little, if any, diagenetic carbonate present in the forams. We leached a sample of benthic forams and a sample of planktic forams from this clay layer with a 0.1 *M* NH_4Cl solution for 2 hr under ultrasonic agitation. The $^{87}Sr/^{86}Sr$ ratio of both foram samples appeared no different from that of the leachate, suggesting that the forams have homogeneous compositions. A bulk carbonate sample, presumably in a worse state of preservation, gave a slightly lower $^{87}Sr/^{86}Sr$ ratio, fitting the observed mixture of early Danian carbonate and reworked late Maastrichtian carbonate material. These observations do not support the existence of a highly radiogenic diagenetic phase. We believe therefore, that the K-T $^{87}Sr/^{86}Sr$ anomaly is not a diagenetic artifact, but reflects changes in the ocean water chemistry of that time. On the basis of this 0.707 893 value, we suspect that the two most radiogenic $^{87}Sr/^{86}Sr$ values we obtained for the Danian of El Kef are outliers (Fig. 2).

Extrapolating the latest Maastrichtian $^{87}Sr/^{86}Sr$ curve underneath the zone of diagenetic overprint (Fig. 2) yields a $^{87}Sr/^{86}Sr$ seawater ratio of 0.707 830 for the Maastrichtian just before the K-T boundary, and determines a 0.000 06 $^{87}Sr/^{86}Sr$ shift across the K-T boundary. A gradual steady-state process causing this $^{87}Sr/^{86}Sr$ shift in the 100–140 k.y. available requires a decrease of mid-ocean-ridge (MOR) spreading by more than 100%, or an increase of runoff Sr contribution by more than 50%[2]. We believe this is not very likely and favor a sharp change related to the K-T boundary event, subsequently smoothed by diagenesis.

BOLIDE IMPACT CAUSING A $^{87}Sr/^{86}Sr$ ANOMALY?

Martin and MacDougall (1991) calculated a maximum contribution to the oceans of 1.2×10^{12} mol of Sr with a 0.715 $^{87}Sr/^{86}Sr$ ratio from the soot of global, impact-induced wildfires at the K-T boundary. This is several orders of magnitude too small to cause the $^{87}Sr/^{86}Sr$ shift in our data (Table 3, Table 4, Fig. 3).

Explaining a 0.000 06 K-T boundary seawater $^{87}Sr/^{86}Sr$ shift by the ejecta from the K-T impact crater in Chicxulub would require the complete dissolution in seawater of all the Sr from a hemispheric crater of a 67–83 km radius (1.1×10^{16} mol of Sr; Table 3, Table 4, Fig. 3). The estimated Chicxulub excavation cavity is about 15–20 km in radius (Sharpton et al., 1994), which means that even the dissolution of all the Chicxulub ejecta cannot produce the K-T $^{87}Sr/^{86}Sr$ shift. Hess et al. (1986) already showed that the Sr content of the impacting bolide is too low to significantly contribute to the shift.

Furthermore, there is nitrous and nitric acid at the K-T boundary, produced by shock heating of the atmosphere by the impactor. Prinn and Fegley (1987) calculated that 4×10^{16} mol of acid will rain out on the continents in the first year or so after the impact. This sudden acidification enhances weathering and releases cations from soil complexes, resulting in strongly increased Sr concentrations in the freshwater system that may drain into the oceans in ~10 yr. An additional 1.3-4 $\times 10^{16}$ mol of sulfuric acid, generated by the impact-evaporization of an evaporite succession in the Chicxulub target rock (Sigurdsson et al., 1992), further increases the acidification.

Assuming a global average of 350 ppm Sr in soils (Aubert, 1977), with a 0.712 $^{87}Sr/^{86}Sr$ ratio and 127 10^6 km^2 of soil surface, it requires 1.9×10^{15} mol, or all the Sr flushed out of a 2.5 m soil profile on a global scale to account for the K-T seawater $^{87}Sr/^{86}Sr$ shift. We doubt if this is a realistic scenario. However, soil leaching may be enhanced by a change at the K-T boundary from a semiarid to humid climate, such as documented by Van Valen and Sloan (1977), Johnson and Hickey (1990), and Retallack et al. (1987). Hence, it is conceivable that due to increased rainfall, arid soil areas, which in the Late Cretaceous barely contributed to the Sr runoff to the

TABLE 3. DATA USED FOR CALCULATIONS IN THE TEXT

Variable	Value	Reference
Chicxulub C1-N10 melt Sr ppm	299-324	Blum et al. (1993)
Chicxulub Y6-N17 melt Sr ppm	551	
Chicxulub C1-N10 melt $^{87}Sr/^{86}Sr$	0.7086	Blum et al. (1993)
Chicxulub Y6-N17 melt $^{87}Sr/^{86}Sr$	0.7086	
Average soils Sr ppm	350	Aubert and Pinta (1977)
Arid soil Sr ppm	1000	Aubert and Pinta (1977)
Arid soil surface W Australia (km^2)	2.00E+06	
Soil density (kg/dm^3)	1.5	
Continental crust Sr ppm	375	
Average run off $^{87}Sr/^{86}Sr$	0.712	Palmer and Edmond (1989)
Continental crust density (kg/dm^3)	2.7	
K-T seawater Sr inventory (mol)	1.25E+17	Palmer and Edmond (1989)
early Danian seawater $^{87}Sr/^{86}Sr$	0.707893	This study
late Maastrichtian seawater $^{87}Sr/^{86}Sr$	0.70783	This study

Note: Chicxulub melts are from the melt sheets of the K-T impact crater in Mexico

[2]See footnote 1.

TABLE 4. EXAMPLES SHOWING THE CALCULATED EFFECT ON THE SEAWATER Sr ISOTOPIC RATIO OF Sr FLUXES FROM DIFFERENT SOURCES

Source of Sr	Sr (moles)	^{87}Sr/^{86}Sr of the source	Resulting seawater ^{87}Sr/^{86}Sr	Remarks
Sr in soot from global wildfires	1.2E+12	0.7150	0.707830	Martin and McDougall (1991)
Required Sr from wildfire soot	1.1E+15	0.7150	0.707893	
Sr in Chicxulub ejecta	5.0E+14	0.7086	0.707833	crater radius 29–30 km
Required Sr in Chicxulub ejecta	1.1E+16	0.7086	0.707893	crater radius 80–83 km
Sr leached from soils by acid rain	7.6E+14	0.7120	0.707855	1.0 m soil profile leached
Required Sr from soil globally	1.9E+15	0.7120	0.707893	2.5 m soil profile leached
Required Sr leached in Australia	3.5E+14	0.7300	0.707893	10.3 m soil profile leached

Note: The Cretaceous-Tertiary seawater Sr isotopic shift is in this study is from 0.707 830 to 0.707 890, and would require unreasonable amounts of Sr from wildfire soot or impact ejecta. However, global-scale acid rain soil leaching combined with a short-term change to a wet climate in arid regions like West Australia could produce the required strontium flux.

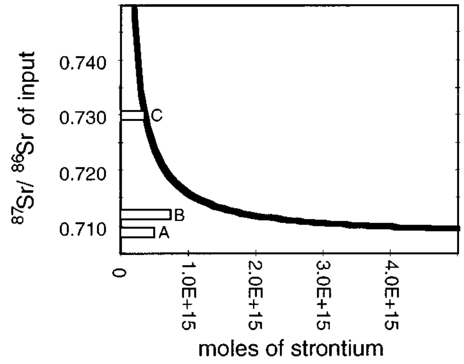

Figure 3: The moles of Sr required to produce 0.000 06 seawater ^{87}Sr/^{86}Sr shift at different ^{87}Sr/^{86}Sr ratios. Bars A, B, and C represent scenarios calculated in text. A: Chicxulub impact ejecta at 30 km crater radius. B: Global top 1 m of soil leaching. C: Top 10.3 m of soil leaching in West Australia desert. Global wildfires (1.2 $\times 10^{12}$ mol of Sr) are not visible at this scale.

oceans and had built up a large reservoir of weathered Sr, would rapidly deliver large quantities of Sr to the oceans. In particular, if in the initial stages of increased rainfall, acid rain predominated. The zone of weathering in an arid region is usually quite deep (>30 m) and rich in Sr (500–2000 ppm Sr). As an example, we take a radiogenic (^{87}Sr/^{86}Sr = ~0.730) arid area of ~2 $\times 10^6$ km^2, representative of the present-day Western Australia desert. It would require the leaching of 3.5×10^{14} mol Sr, which is all the Sr from ~10.3 m soil profile (at 1000 ppm), to supply the necessary amount of Sr (Fig. 3) to the oceans. Assuming that the same process takes place in other arid and semiarid regions and adds up to the global acid rain weathering discussed above, the required depth of the leached soil profile becomes significantly thinner. We therefore propose a scenario of global acid rain soil leaching in combination with a change to a wet climate in formerly arid areas to have produced the K-T oceanic ^{87}Sr/^{86}Sr anomaly.

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