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Acid Trauma at the Cretaceous-Tertiary Boundary in Eastern Montana

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ABSTRACT

Acid is a likely consequence of many of the catastrophic events postulated for the Cretaceous-Tertiary boundary: nitric acid from atmospheric shock by bolide and from burning of trees, sulfuric acid from volcanic aerosols and from impact vaporization of evaporites, hydrochloric acid from volcanic aerosols, and carbonic acid from carbon dioxide of volcanoes and fires. The amount of acid is here estimated from base cation leaching of boundary beds and paleosols in eastern Montana. The thin boundary claystone consumed so much more acid than the overlying impact layer and associated paleosols that strong acids are indicated. Vigorous early neutralization of hot acid by silicate ejecta may explain the distinctive kaolinitic composition, and the microspherulitic and vuggy texture of the boundary bed, in which impact evidence such as shocked quartz has been destroyed by profound chemical leaching. This early buffering of impact-generated acid was fortunate for life in terranes with high acid-buffering capacity such as the calcareous and smectitic floodplains of Montana, which were not acidified to less than pH 4, thus sparing small mammals, amphibians, and fish, but affecting plants, non-marine molluscs, and dinosaurs.

INTRODUCTION

Catastrophic impact of a large bolide at the Cretaceous-Tertiary (K-T) boundary is now established from evidence of iridium anomalies (Alvarez et al., 1980; Orth et al., 1990), shocked quartz (Izett, 1990), dramatic changes in fossil plants (Wolfe and Upchurch, 1987; Nichols et al., 1990; Johnson and Hickey, 1990), and a large impact crater in Yucatán (Hildebrand et al., 1991; Sharpton et al., 1993; Kring, 1995). Also occurring at this time were flood-basalt eruptions of the Deccan Traps in India (Duncan and Pyle, 1988; Courtillot et al., 1990) and widespread wildfires (Wolbach et al., 1988; Tinus and Roddy, 1990). Acid is a likely consequence of all

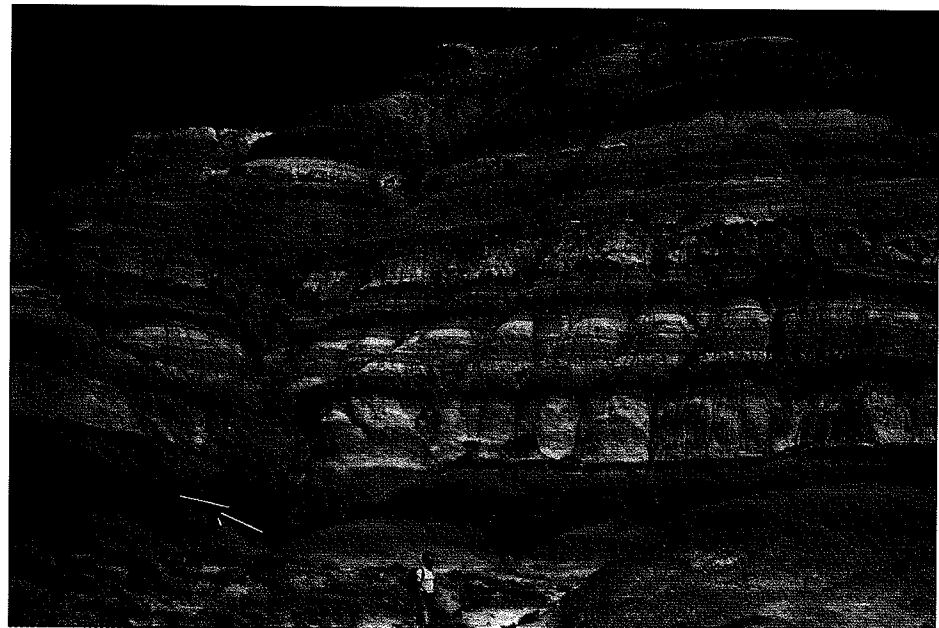


Figure 1. The K-T boundary (arrow) in the sequence of paleosols in Bug Creek (NW¼NW¼SE¼, sec. 17, T. 22 N., R. 43 E.), McCone County, Montana. The iridium anomaly is weak here because of bioturbation into a moderately developed paleosol beneath the dark gray band exposed in the trench excavated low in the bluffs to the left.

these events: nitric acid from atmospheric shock by the bolide and from burning of trees (Zahnle, 1990), sulfuric acid from volcanic aerosols and impact vaporization of evaporites (Hildebrand et al., 1991; Sigurdsson et al., 1992; Brett, 1992; Sharpton et al., 1993), hydrochloric acid from volcanic aerosols (Caldeira and Rampino, 1990), and carbonic acid from carbon dioxide of volcanoes and fires (Wolbach et al., 1988; Tinus and Roddy, 1990). All this acid should have left a record in paleosols or boundary beds. This study has been a search for direct evidence of acid leaching and an exploration of the role of acid in the still-controversial topic of selective extinctions at the K-T boundary (Williams, 1994; Ward, 1995).

PALEOSOLS AND K-T BOUNDARY BEDS IN MONTANA

Paleosols in the Bug Creek and Brownie Butte areas of eastern Montana

(Retallack et al., 1987; Retallack, 1994) are a remarkably complete fossil record of K-T boundary events (Smit et al., 1987; Rigby and Rigby, 1990; Swisher et al., 1993). Only a weak iridium anomaly and no distinctive boundary beds have been found in Bug Creek, but the K-T boundary can be located there by means of unusually abundant fern spores and fossil plant extinctions. The "zone of death" in Bug Creek is the carbonaceous surface of a moderately developed paleosol into which the thin ejecta layers were presumably mixed by the action of later roots and burrows (Fig. 1). At Brownie Butte, the K-T meteoritic ejecta include an impact bed, which is 1 cm thick, gray, smectitic, and layered, with shocked quartz and an iridium anomaly (Figs. 2 and 3). It directly overlies the boundary bed, which is 2 cm thick, pink to white, kaolinitic, micro-

K-T Boundary continued on p. 2

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spherulitic, and vuggy. These two distinctive thin beds have been discovered at 30 sites from Alberta and Saskatchewan south to New Mexico at the radiometrically and palynologically determined K-T boundary (Izett, 1990). The boundary bed at Brownie Butte has been interpreted as a paleosol (Fastovsky et al., 1989; Izett, 1990), but is now regarded as an early-settling fraction of altered ejecta from bolide impact (Alvarez et al., 1995). By either interpretation, its distinctive kaolinitic composition requires reaction with acid, quantification of which is contingent on the exact origin of these boundary layers. Alvarez et al. (1995), explained the distinctive composition of the boundary bed by postulating a glassy parent material or high temperature. These factors would kinetically favor base leaching, but there remains a need for acid to carry out this marked chemical mass transfer.

The boundary and impact beds have been interpreted as fallout from separate impacts within months of one another because the boundary bed has plant remains interpreted as root traces truncated by the impact bed (Fastovsky et al., 1989; Izett, 1990). By this view the boundary bed represents ejecta from the Chicxulub crater, and the source of the impact bed was thought to be the Manson crater, Iowa (Izett, 1990). However, the Manson crater is now known to be about 10 m.y. older than the K-T boundary (Izett et al., 1993). In addition, shocked zircons from the K-T impact layer have crystallization ages much younger than found near the Manson crater, and they are compatible with the age of target rocks around Chicxulub (Kamo and Krogh, 1995). Furthermore, isotopic measurements of Sr, O, and Nd on K-T impact glasses are similar to Chicxulub, rather than Manson melt (Blum et al., 1993). In view of this evidence against two impacts, Alvarez et al. (1995) interpreted the root traces as truncated plant stalks and proposed that the

boundary bed is altered glassy ejecta from an early ejecta blanket of melt, shocked rocks, and admixed sea water, followed within hours by fallout from a warm fireball with volatiles, rocks, and shocked quartz.

Decisive evidence for either view is the nature of fossil plant debris in the boundary layer. The concertinalike deformation of the plant material is an indication that it was there in life position before burial and compaction of the sediments (Fastovsky et al., 1989), as would be true for either plant stalks or root traces. However, the structures in the boundary bed are plant stalks, because they are 5 mm or more in diameter and lack the fine rootlets that accompany large roots. Decisive evidence that these are not roots is the way some of these carbonaceous structures branch and are frayed upward (Figs. 2 and 3). My preference is to interpret both impact and boundary beds as different phases of a single impact, but acid consumption for double impact and local derivation also has been calculated.

COMPUTING ACID CONSUMPTION

Both weak acids of weathering and strong acid rain have the effect of displacing basic cations (Ca²⁺, Mg²⁺, K⁺, Na⁺) with hydronium (H⁺) by hydrolysis. Using procedures from studies of modern soil acidification (Fölster, 1985), the loss of basic cations can be used to calculate the moles of hydronium consumed from weight percent analytical values and bulk density compared with parent materials of K-T boundary beds and paleosols (Appendix 1; data from Fastovsky et al., 1989; Retallack, 1994). Units of equivalents to hydronium were used rather than moles because of the differing hydronium contents of the likely acids (HNO₃; H₂SO₄).

These estimates of acid consumption are conservative for the following four reasons. First, acid consumption by C horizons was not included, because these sediments were considered parent materials.

Second, allowances were not made for acid-induced aluminum loss, because the paleosols retained clay and had little variation in alumina/silica ratios, with no chemical or petrographic indications of podzolization (Retallack, 1994). Third, potential loss of soil material by landscape denudation was not included because the paleosols were in a large sedimentary basin (Smit et al., 1987; Rigby and Rigby, 1990). Fourth, coaly horizons were not included, because their low mineral content was probably original rather than due to hydrolytic destruction of minerals (Retallack, 1994).

The most critical assumption of these calculations is parent-material composition, against which base loss was assessed. Separate samples were taken as parent materials for Cretaceous and Paleocene paleosols (Table 1), because of changes in alluvial source areas (Retallack, 1994). Because the boundary claystone paleosol formed on an airfall deposit whose ultimate origin can be interpreted in several ways (Izett, 1990; Fastovsky et al., 1989; Alvarez et al., 1995), all conceivable parent materials were estimated: local Cretaceous and Paleocene sediments (Retallack, 1994), the impact bed at Brownie Butte (Fastovsky et al., 1989; Izett, 1990), melt rock from Chicxulub crater, Mexico (Hildebrand et al., 1991), impact glasses from Beloc, Haiti (Sigurdsson et al., 1991, 1992) and Mimbral, Mexico (Smit et al., 1992), and glasses, microbreccias, and target rocks from the Manson crater, Iowa (Koeberl and Hartung, 1992). These various calculations were done to cover a variety of potential interpretations.

BACKGROUND ACID CONSUMPTION

Potentially exceptional acidification at the K-T boundary must be compared with background acidification due to normal weathering. The calculations (Fig. 4) show that the total amount of acid consumed by mineral horizons of the paleosols was not much different from Late Cretaceous to early Paleocene. This result is supported by Bell (1965), who found base-rich clay near the boundary (Fig. 4). It is also supported by the lack of change in paleosol depth functions of barium/strontium and base/alumina ratios, of trace metals such as Cu, Ni, and Zn, and of rare earths across the K-T boundary (Retallack, 1994). There is no significant difference between four Cretaceous paleosols analyzed that used on average 5297 ± 3758 keq/ha acid (error of 1σ) and nine Paleocene paleosols that used 2069 ± 1481 keq/ha.

Estimated total acid consumption of the paleosols does not take into account their different times for formation. Some paleosols retained clear relict bedding, and are effectively sediments disrupted by only a few seasons of root growth. Other paleo-

TABLE 1. TOTAL ACID CONSUMPTION OF K-T BOUNDARY AND IMPACT BEDS AT BROWNIE BUTTE, MONTANA, FOR VARIOUS ASSUMED PARENT MATERIALS AND HYPOTHESES

Assumed parent material	Number of analyses	Bulk density g/cm ³	Acid consumption, boundary claystone keq/ha	Acid consumption, impact bed keq/ha	Data source*
<i>Hypothesis of single impact (favored here)</i>					
Chicxulub melt, Mexico	2	2.5†	299.7	158.5	1
Tektite, Beloc, Haiti	19	2.8†	647.8	318.5	2
Tektite, Mimbral, Mexico	3	2.5†	327.9	144.5	3
<i>Hypothesis of multiple impact</i>					
Glass, Manson, Iowa	6	2.5†	159.9	74.6	4
Country rock, Manson, Iowa	16	2.5†	276.5	132.9	4
<i>Hypothesis of local derivation</i>					
Paleocene, Montana (R513)	1	1.93 ± 0.05	136.0	62.6	5
Cretaceous, Montana (R610)	1	2.07 ± 0.05	158.7	73.8	5
<i>Relative acidification for all hypotheses</i>					
Impact bed, Montana	2	2.02 ± 0.08	5.4	0	6
Boundary bed, Montana	2	1.92 ± 0.01	0	-10.8	6

*1: Hildebrand et al. (1991); 2: Sigurdsson et al. (1992); 3: Smit et al. (1992); 4: Koeberl and Hartung (1992); 5: Retallack (1994); 6: Fastovsky et al. (1989).
†Estimated values; other densities were measured (Retallack, 1994).

sols had well-mixed clayey subsurface horizons of the kind that form over thousands of years. Estimates of the rate of acid consumption (in keq · ha⁻¹ · yr⁻¹) used maximum values for duration of ancient soil formation estimated by comparison with studies of morphological (*not chemical*) differentiation of Quaternary soils. These time estimates are discussed elsewhere (Retallack, 1994). The calculated minimal rates of acid consumption of Late Cretaceous and early Paleocene paleosols are not appreciably different from each other or from those of Holocene soils (Föhlster, 1985), which generally fall between limits of 0.2 and 2.3 keq · ha⁻¹ · yr⁻¹. The four latest Cretaceous paleosols had an average rate of acid consumption of 2.0 ± 1.7 keq · ha⁻¹ · yr⁻¹, and the nine earliest Paleocene paleosols had a rate of 0.9 ± 0.4 keq · ha⁻¹ · yr⁻¹.

These unsurprising rates and total acid consumption for paleosols above and below the boundary are evidence against a long-term volcanic or meteoritic contribution to paleosol acidity in Montana. In addition, paleosols near the boundary in Bug Creek are somewhat less calcareous but more smectitic than paleosols earlier in the Cretaceous or later in the Paleocene (3-26 m in Fig. 4). Eruption of the Deccan Traps has been proposed to have released 5×10^{17} moles CO₂, 1.7×10^{17} moles H₂SO₄, and 7.4×10^{15} moles HCl (Caldeira and Rampino, 1990), but the effect of this acid was mitigated by smaller doses spread out over about 600,000 yr (Courtillot et al., 1990).

MINIMAL ACID CONSUMPTION AT THE K-T BOUNDARY

An estimate of minimum acid consumption from the boundary bed indicates that strong acid was involved, rather

than merely weak acids such as carbonic acid. The unique arrangement of impact over boundary bed allows assessment of minimal acid use of the boundary bed in excess of that used by the overlying impact bed at Brownie Butte (Table 1). The boundary claystone and its plant debris is more acidified by at least 5.4 keq/ha than the sharply overlying, well-bedded, smectitic impact layer (Figs. 1 and 2, Table 1). This significant acidification could not have been created by deposition or alteration early during burial, for the following reasons. There are no local kaolinitic source beds or diagenetic mechanisms that would form the boundary bed in so many separate depositional basins (Izett, 1990). The boundary claystone may have been leached downward from overlying lignitic paleosols at Brownie Butte, as argued for other kaolinitic coal partings (Staub and Cohen, 1978; Demchuck and Nelson-Glatiotis, 1993), but this would have affected the overlying impact bed as well. The boundary bed was much more profoundly leached than the overlying impact bed.

This minimal value of 5.4 keq/ha for the boundary claystone is evidence for strong acid leaching. It is significantly greater than for paleosols at the boundary, which could have consumed as little as an unexceptional 0.2 ± 0.006 keq · ha⁻¹ · yr⁻¹. The boundary bed is an order of magnitude thinner than the paleosols, yet this small volume consumed more than twice as much acid. This leaching would have been in place within a soil over a period of months by the two-impact model of Fastovsky et al. (1989), and Izett (1990), but it is more likely that it was leached during emplacement and within hours

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before the later-settling bed of high-energy ejecta from the same impact (Alvarez et al., 1995). For comparison, a modern soil from near Unadilla in upstate New York, after experimental application of rain of pH 3.5, maintained a pH of 4.1 in mineral horizons and lost $7.8 \text{ keq} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ from these horizons (Cronan, 1985), which is comparable to the loss estimated here for the boundary bed in Montana and about three times the loss from weak acids (Fölster, 1985). The calculated 5.4 keq/ha spread out over a year proposed by the two-impact model (Fastovsky et al., 1989) is comparable to modern soils locally polluted by mine waste or industrial acid. By the single-impact model (Alvarez et al., 1995), this is a dramatic short-term acidification.

MAXIMUM ACID CONSUMPTION AT THE K-T BOUNDARY

There are several ways of assessing upper limits to acid consumption of soils and boundary beds at the K-T boundary in Montana. A direct calculation for two paleosols at the boundary in Bug Creek gives an average consumption of $6585 \pm 199 \text{ keq/ha}$. These paleosols show profile differentiation and little remaining relict bedding compatible with some 15 ka of soil formation, which would give a rate of acid consumption of $0.2 \pm 0.006 \text{ keq} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$. These values are normal for Late Cretaceous, early Paleocene, and late Quaternary soils, as already mentioned. There is no indication of podzolization in the petrographic or chemical composition of the boundary bed or other paleosols of Montana (Retallack, 1994), so that pH is likely to have been buffered to above 4. This figure is supported by the pattern of extinction of different kinds of organisms across the K-T boundary in Montana. Considering acid tolerances of related living creatures (Howells, 1990; Weil, 1994), groundwater pH in Montana was probably less than 5.5 but no less than 4.

A dramatically different view emerges from calculations based on the impact and boundary beds at Brownie Butte, for which a maximal acid consumption of 986 keq/ha can be calculated by using parent material with the composition of tektites from Beloc, Haiti (Table 1). By the model of Alvarez et al. (1995), this amount of acid would have been consumed within hours; by the model of Fastovsky et al. (1989), it would have been consumed within a year. This more serious acid load is compatible with prior theoretical estimates of acid production. Estimates on the production of NO_2 by a bolide capable of creating K-T geochemical anomalies have varied from 1×10^{14} to 1.2×10^{17} moles (Lewis et al., 1982; Prinn

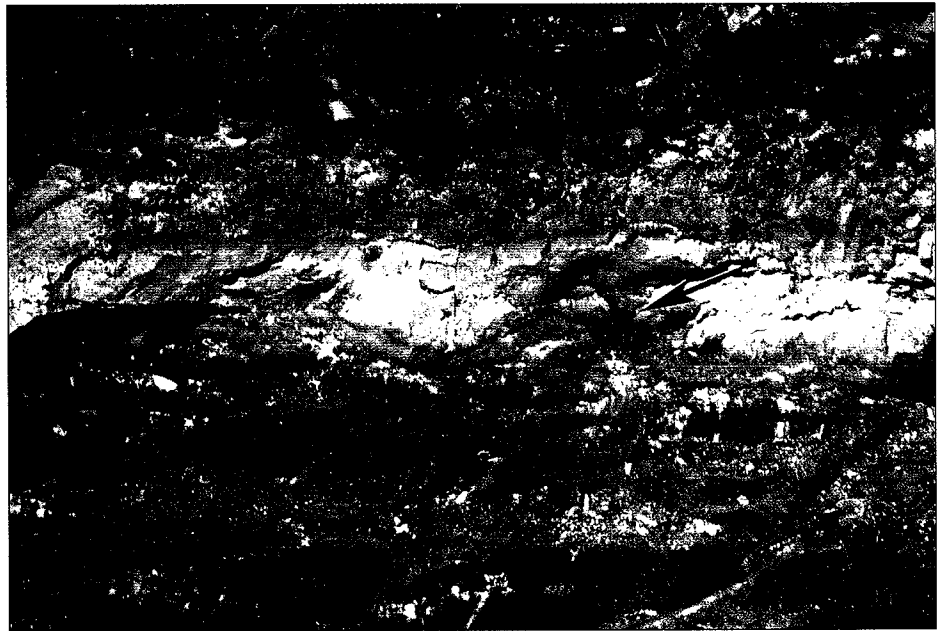


Figure 2. The K-T boundary and impact beds near Brownie Butte (SE $\frac{1}{4}$ SW $\frac{1}{4}$ SW $\frac{1}{4}$, sec. 32, T. 21 N., R. 37 E., Garfield County), Montana. The branching and upward-forked brown carbonized material in the pink boundary bed (arrow) is a frayed shoot, as predicted by the model of Alvarez et al. (1995), not a root, as interpreted by Fastovsky et al. (1989).

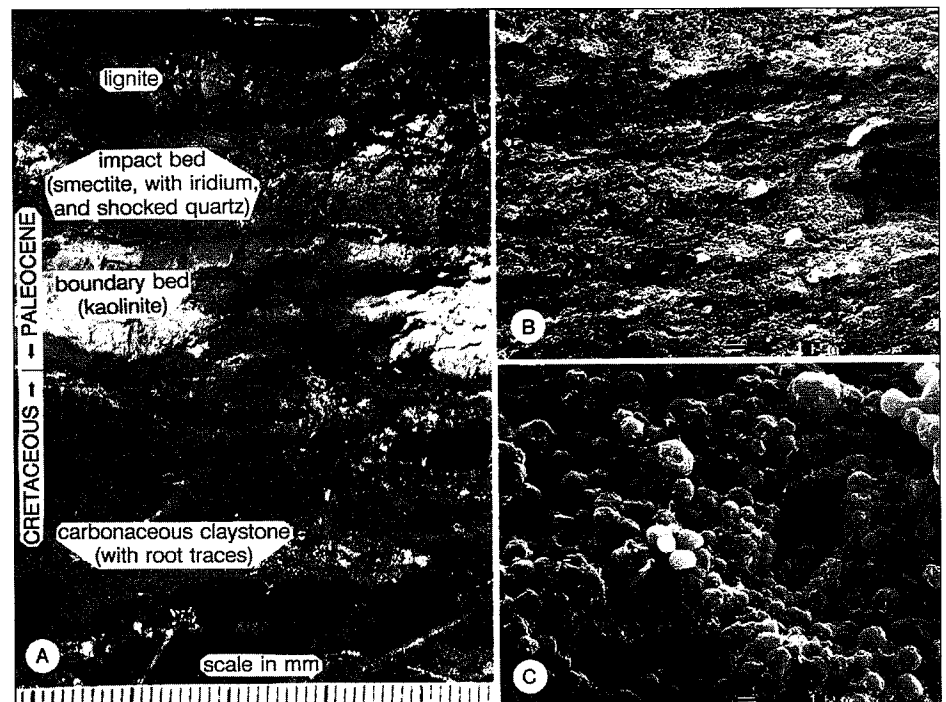


Figure 3. Annotated field photograph of the K-T boundary beds at Brownie Butte (A), with scanning electron micrographs of the impact bed (B) and boundary claystone (C). The pelletoidal and vuggy microstructure of the boundary claystone reflects vigorous acid leaching, which the later-settling laminated impact bed largely escaped. Scales are in millimeters for the field photograph and in micrometers for the micrographs.

and Fegley, 1987; Zahnle, 1990) or some $2\text{--}2350 \text{ keq/ha}$ of Earth's surface area. An additional source of acid on short time scales is vaporization of anhydrite evaporites under the impact crater of Chicxulub, Mexico (Hildebrand et al., 1991; Sigurdson et al., 1992; Brett, 1992; Sharpton et

al., 1993). This may have produced 4×10^{17} to $1.3 \times 10^{19} \text{ g SO}_2$ (Brett, 1992; Sigurdson et al., 1992), which is 6.2×10^{15} to 2.0×10^{17} moles, or $254\text{--}7840 \text{ keq/ha}$ globally. Wildfires would produce comparable amounts of NO and CO_2 (Zahnle, 1990), perhaps focused at the boundary

(Wolbach et al., 1988; Tinus and Roddy, 1990). An additional estimate from hypothetical oceanic titration (d'Hondt et al., 1994) is a total acid load of no more than 5×10^{16} moles, or 980 keq/ha globally. The maximal acidification of the boundary bed estimated here indicates that the lower estimates of acid load are reasonable, but the higher estimates are excessive.

If generated, then where did all this acid go to leave associated paleosols only mildly acidified? One possibility is consumption by reaction with impact ejecta. Assuming the single-impact model of Alvarez et al. (1995), rock of the low-energy ejecta curtain would have been attacked by acid generated from entrained SO_2 and NO_x as it cooled during or shortly after ballistic emplacement, unless quenched and diluted by fallout in the deep ocean (as for glasses of the Caribbean area described by Sigurdsson et al., 1992; Smit et al., 1992). The volume of ejecta thrown up by the impact at Chicxulub has been estimated to be $1-2.2 \times 10^4$ km³ (Kring, 1995). If all of this were leached to the degree seen in the boundary bed from a composition similar to Haitian tektites, it would consume 7.1×10^{11} eq of acid. The remainder of the broadcast acid could easily be accommodated by the mild acidification seen in the paleosols at Bug Creek. This is the most optimistic atmospheric scrubbing of acid imaginable, so some of the hypothetical estimates of acid production cited above can be still regarded as excessive.

A short burst of acidic leaching not only explains the base-poor, kaolinitic composition of the boundary bed, but also its anomalously low Ni, Co, and Ir content for either meteoritic or volcanic material (Izett, 1990) and its peculiar spherulitic and vuggy microtexture (Fig. 3). Because iridium concentrations would be dispersed and shocked quartz, spherules, and other indicators of impact origin obliterated by this chemical leaching, their absence in the boundary bed does not require hypotheses of ballistic sorting (of Alvarez et al., 1995). Such acidic leaching of iridium and shocked quartz from impact ejecta could explain weak to nonexistent geochemical and mineralogical signatures at other major extinction events (Orth et al., 1990). Thus, acid generated by impact could make some impacts geochemically "self cleaning."

Increased weathering induced by acid has been invoked to explain anomalous enrichment of crustal strontium in marine foraminifera at the K-T boundary (MacDougall, 1988; Martin and MacDougall, 1991). Crustal strontium also could have been leached to the ocean from the hot fallout preserved as the boundary bed.

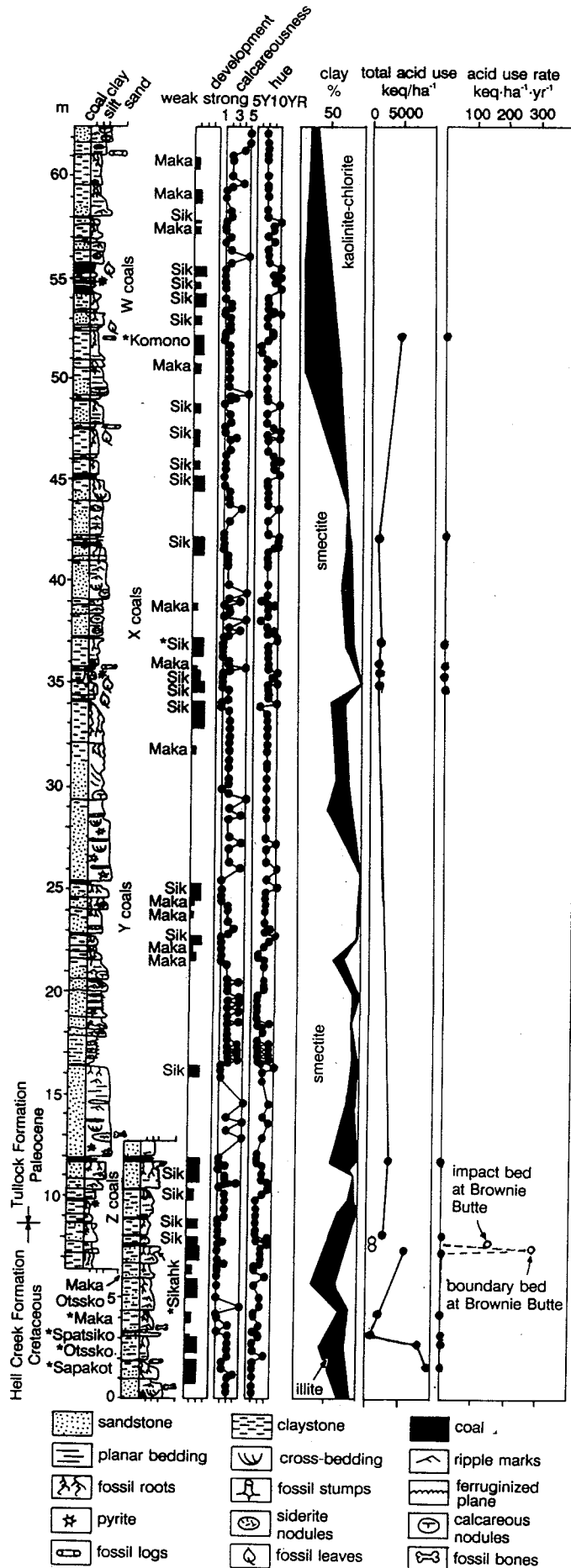


Figure 4. Measured section of paleosols and selected indices of weathering across the K-T boundary in cliffs and a low knoll north of Bug Creek (NW¼ NW¼ SE¼, sec. 17, T. 22 N., R. 43 E., McCone County), Montana. Black boxes indicate positions of the paleosols; lengths of boxes correspond to degree of development (Retallack, 1990). The calcarousness scale is for field reaction with 1.2M HCl (Retallack, 1990), hue data are from Munsell charts, and clay mineral data are by X-ray diffraction (Bell, 1965). The impact bed and boundary claystone (open circles) were not preserved in Bug Creek, but are known from Brownie Butte, Montana. Their acid use is plotted assuming derivation from Chicxulub melt, but calculated use varies with other assumed parent materials (Table 1).

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BIOTIC EFFECTS

The amount of NO₂ produced by the bolide at the K-T boundary has been estimated at globally averaged atmospheric concentrations of about 0.5 ppm V (Lewis et al., 1982) or 21 μmol/m³. A comparable amount of SO₂ is known to injure leaves directly (Whitmore, 1985; Howells, 1990). Doses near the source would have been much higher than this globally mixed average. The high-temperature acid vapor and melt ejecta proposed by Alvarez et al. (1995) would have been particularly lethal, its effects tapering off with distance from the impact.

In Montana, reconstructed at 3330 km from the Chicxulub crater (Kring, 1995), noxious gases, acid, and warm leached ejecta raining out to a 2 cm layer would still have had a significant effect on large plants and animals. Scalding by later cool acid rain, darkening of the sky by dust, chilling of the atmosphere by dust shielding, and then warming by a greenhouse effect (Prinn and Fegley, 1987; Zahnle, 1990) would thus have been additional insults to a biota already traumatized by acidic fluids and ejecta. Acidic trauma may explain the transition in Montana from eutrophic angiosperm-dominated semievergreen forests to a fern-dominated recovery flora and then to oligotrophic conifer-dominated swampland (Wolfe and Upchurch, 1987; Nichols et al., 1990; Johnson and Hickey, 1990), and from herbivorous to insectivorous vertebrates (Sheehan and Fastovsky, 1992).

Even within a single area such as Montana, different organisms fared differently across the K-T boundary. The aquatic molluscs were severely affected (Morris, 1990), but amphibians and fish were little affected (Archibald and Bryant, 1990; Weil, 1994). Molluscs would have been excluded by pH less than 5.5, but greater losses of fish and amphibians would have been expected at pH less than 4 (Howells, 1990; Weil, 1994). Acid buffering by calcareous smectitic soils may have been important to the survival of small birds, mammals, reptiles, amphibians, and fish. Similarly, oceanic mixing and buffering may have diluted acid to no less than pH 7.6, so that many of the ammonites and coccolithophores died, but other molluscs, radiolarians, and acid-sensitive dinoflagellates survived (d'Hondt et al., 1994). Biotic effects of acid rain would have been more severe in less well buffered soils of humid granitic terrains and in shallow seas receiving runoff from such regions. Thus, impact-generated acid could have been a selective kill mechanism from place to place, and within the same ecosystem, if buffered to reasonable levels by wide dispersal and reaction with ejecta, soils, and the ocean.

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APPENDIX 1. FORMULAE FOR CALCULATING ACID CONSUMPTION OF SOILS AND PALEOSOLS

$$B = 2\rho(0.01783C + 0.02481M + 0.01062K + 0.01613N)/100$$

$$T_i = [(D_i + D_{i-1}) - (D_i + D_{i+1})]/2$$

$$A = \sum T_i(B_p - B_i)$$

Symbols:

- A = acid consumption of profile (eq/cm²)
- B = base content of sample (eq/cm³)
- C, M, K, N = CaO, MgO, K₂O, Na₂O, respectively (wt%)
- D = depth to sample (cm)
- T = thickness represented by sample (cm)
- ρ = bulk density (g/cm³)

Subscripts:

- i = for sample i
- i + 1 = for sample or surface above i
- i - 1 = for sample or parent material below i
- p = for parent material

Constants:

- 0.01738, 0.02481, 0.01062, 0.01613 = element in oxide (mole)
- 2 = equivalence adjustment
- 100 = weight percent adjustment

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