

# Reports

## Cretaceous Extinctions: Evidence for Wildfires and Search for Meteoritic Material

**Abstract.** Clay samples from three Cretaceous-Tertiary boundary sites contain 0.36 to 0.58 percent graphitic carbon, mainly as fluffy aggregates of 0.1 to 0.5 micrometers—apparently a worldwide layer of soot. It may have been produced by wildfires triggered by a giant meteorite. This carbon, corresponding to a global abundance of  $0.021 \pm 0.006$  gram per square centimeter, could have greatly enhanced the darkening and cooling of the earth by rock dust, which has been suggested as a cause of the extinctions. The surprisingly large amount of soot (10 percent of the present biomass of the earth) implies either that much of the earth's vegetation burned down or that substantial amounts of fossil fuels were ignited also. The particle-size distribution of the soot is similar to that assumed for the smoke cloud of "nuclear winter," but the global distribution is more uniform and the amounts are much greater, suggesting that soot production by large wildfires is about 10 times more efficient than has been assumed for a nuclear winter. Thus cooling would be more pervasive and lasting. No trace of meteoritic noble gases and no meteoritic spinel were found in these carbon fractions. Accordingly, limits can be set on the mass fraction of the meteorite that escaped degassing ( $\leq 3 \times 10^{-5}$ ) or vaporization ( $\leq 0.04$ ). Thus it seems unlikely that comets contributed significant amounts of prebiotic organic matter to the primitive earth.

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The mass extinctions at the end of the Cretaceous apparently were caused by the impact of a meteorite (~10 km) that deposited a worldwide layer of iridium and other meteoritic elements (1-4). To learn more about the nature of the meteorite (2), we looked for meteoritic noble gases in clay from the Cretaceous-Tertiary (K-T) boundary. Though the major part of the meteorite undoubtedly vaporized on impact, a small amount may have survived, and might be recognized or even identified by its primordial noble gas pattern.

Clay samples from the K-T boundary at Stevns Klint, Denmark; Caravaca, Spain; and Woodside Creek, New Zealand, were treated with a succession of reagents that dissolve silicates, soluble organics, and kerogen, leaving elemental carbon and certain resistant minerals (rutile and spinel) unaffected (5). Previous work has shown that the primordial noble gases in meteorites largely reside in such a carbon residue (6).

**Undegassed material.** Six carbona-

ceous samples were analyzed for xenon, one also for neon, and one for all other noble gases. To remove the adsorbed atmospheric component, we extracted the gases in four to eight steps, from 400° to 2000°C (Table 1). Atmospheric gases evolve below 800°C, and meteoritic gases, mainly above 800°C (6). In further, but largely unsuccessful, attempts to deplete adsorbed gases, we etched some samples with HNO<sub>3</sub>, or isolated a colloidal carbon fraction.

More than 99 percent of the gas was released between 400° and 800°C and had terrestrial isotopic composition. Only small amounts were evolved above 800°C, slightly above blank levels. We have conservatively interpreted them as upper limits. The most stringent limits come from xenon.

From these data we can estimate upper limits to the mass fraction  $F$  of the meteorite that was not degassed. Let  $I_m$  and  $X_m$  be the iridium and <sup>132</sup>Xe concentrations of the meteorite;  $I_b$  and  $X_b$  the corresponding surface concentrations [in nanograms per square centimeter or in cubic centimeters at standard temperature and pressure (STP) per square centimeter] at the boundary; and  $\alpha$  the fraction of meteoritic gas remaining after etching. Then  $F = I_m X_b / X_m I_b \alpha$ . Assuming CI chondrite composition (7) and

$\alpha = 0.1$  (6), we obtain the mass fractions in the last column of Table 1.

The most interesting value is the lowest one (D5Af). Apparently less than  $3 \times 10^{-5}$  of the meteorite escaped degassing—equivalent to a 5-cm skin on a 10-km body.

This is a surprising result. Stony meteoroids, especially those of cometary origin, are very friable and hence spall or break up in the atmosphere; this sometimes happens even for kilometer-sized bodies, as shown by the spontaneous splitting of comets and the presence of twin craters on the earth and moon (8). Moreover, the rear surface of a meteorite usually is torn off on impact by a rarefaction wave. Surely more than  $3 \times 10^{-5}$  of the K-T meteorite must have been spalled off in flight and on impact. Perhaps we missed these spalls because they fell later (or more locally) than the main ejecta. But a more likely reason is that the spalls were engulfed by the fireball and vaporized or at least outgassed. The dust train left by the meteorite could have been no longer than ~200 km (except for a very oblique impact), and at least the forward part must have been vaporized by the 10<sup>30</sup>-erg fireball. Still, the solid ejecta would provide some shielding, and as complete degassing of noble gases from meteoritic carbon takes minutes at 1600°C and hours at 1200°C or lower, one would expect some material to survive with only partial degassing. We have yet to find it.

**Prebiotic organic matter from comets?** This negative result casts further doubts on the popular notion that comets contributed a significant part of the earth's prebiotic organic matter. It has been clear all along that only bodies in the meteoritic size range (a few centimeters to a few meters) could survive atmospheric entry; larger bodies would be destroyed on impact, with organic compounds reverting to CO and H<sub>2</sub> (9). But our new data suggest that such destruction would be remarkably complete. As organic compounds in meteorites volatilize more readily than do noble gases (~400°C compared to 800° to 1600°C), the fraction of organic compounds surviving the impact must have been even less than  $3 \times 10^{-5}$ . For this reason, it is also unlikely that toxic substances from the meteorite, such as HCN, were a major cause of the extinctions (10).

**Unvaporized material.** Two potential survivors are spinel, MgAl<sub>2</sub>O<sub>4</sub> (melting point 2135°C) and graphitic carbon (melting point 3550°C, but combustible). Both are present in carbonaceous chondrites in amounts of 0.1 to 0.3 percent. An acid-insoluble fraction from New

Zealand was treated with  $\text{HClO}_4$  to destroy carbon, and the residue (0.053 percent of the original clay) was examined for spinel on the scanning electron microscope. No spinel grains were seen (<1 percent); the principal mineral was (detrital) rutile (11). Proceeding as before, we derive an upper limit of  $\leq 0.04$  for the unvaporized mass fraction of the meteorite.

An analogous limit can be derived from carbon, but meteoritic carbon must be identified isotopically, to distinguish it from the more abundant terrestrial carbon (12).

**A worldwide carbon layer.** All K-T boundary samples gave about 1 to 2 percent of insoluble black carbonaceous matter. After removal of oil and kerogen, surprisingly large amounts (0.36 to 0.58 percent) of elemental carbon remained, 4 to 5 times more than that in the marls above or below the Danish boundary and 4 to 25 times more than that in modern marine sediments (Table 2). Actually, the excess is far greater still if the differences in sedimentation rate are taken into account. The impact glass from which the boundary clay is derived must have settled in less than 1 year, as the ejecta cannot have stayed aloft much longer than a few months. This implies a sedimentation rate of more than 1 cm/year (before any spreading by bioturbation and so on). The adjacent strata, on the other hand, have deposited at mean rates of 0.012 cm/year (upper Maastrichtian) and 0.005 cm/year (lowest Danian), judging from their thickness and age (13).

Table 1. Noble gases in carbon from K-T boundary clay.

Sam- ple*	Xenon-132†		F‡
	$10^{-10} \text{ cm}^3/\text{g}$		
	$\leq 800^\circ$	$> 800^\circ$	
D5Af	425	0.26	$16 < 3 \times 10^{-5}$
D5Afn	264	0.21	$12 < 2 \times 10^{-4}$
D5Ac	322	0.23	$20 < 4 \times 10^{-5}$
D5Acn	176	1.9	$9 < 2 \times 10^{-4}$
N5An	182	2	$160 < 6 \times 10^{-3}$
S5A	5	0.7	$250 < 8 \times 10^{-4}$

\*D, Stevns Klint, Denmark; N, Woodside Creek, New Zealand; S, Caravaca, Spain. The four Danish samples were all prepared from the same residue. Lower-case letters indicate further separation or treatment of the original residue: c, coarse; f, fine (colloid); and n, etched with 16M  $\text{HNO}_3$  for 2 hours at  $85^\circ\text{C}$ . †Italicized values are  $\leq 2$  times the normal procedural blank, which in these cases has not been subtracted. For the remaining high-temperature fractions, the normal blank was subtracted. However, the actual blank may have been larger, owing to residual gas from the preceding, large, low-temperature fractions. Isotopic compositions of the  $\leq 800^\circ\text{C}$  fractions were atmospheric; those of the  $> 800^\circ\text{C}$  fractions nearly so, implying that less than half the gas was meteoritic. ‡Fraction of meteorite that was not degassed. These values are too large by at least a factor of 2 because it was assumed that all the xenon above  $800^\circ\text{C}$  was meteoritic (although less than half was).

Thus the mean carbon abundance at the K-T boundary ( $0.021 \text{ g/cm}^2$ ) implies a worldwide flux  $10^4$  times the present one or some  $10^3$  times the Late Cretaceous–Early Tertiary value suggested by the two Danish samples below and above the boundary.

All samples consist mainly of elemental carbon, with little or no kerogen. This is indicated by the lack of color in the final NaOH extracts (5) and by the high combustion temperature ( $\geq 500^\circ\text{C}$  in 20

torr  $\text{O}_2$ , in contrast to marine kerogens, which combust at  $\leq 300^\circ\text{C}$  under these conditions) (12).

Under the scanning electron microscope, the carbon from all three K-T sites shows a bimodal size distribution. Submicrometer-sized particles dominate by number and even by mass (82 percent in the Danish sample). Most show the characteristic morphology of carbon deposited from flames, such as soot or carbon black: irregular, fluffy, often chainlike clusters of spheroids (Fig. 1, A and C), but some are compact and platy (Fig. 1B); these predominate above and below the boundary. Large, micrometer-sized particles of irregular shape (Fig. 1D) are present in lower abundance (1 to  $3 \mu\text{m}$  at 3 percent;  $> 3 \mu\text{m}$  at 15 percent). They resemble charcoal from forest fires or marine sediments (14), which is also derived from wildfires. Tschudy *et al.* (15) had found fusinite (carbonized woody material) in a thin coal seam above the K-T boundary at Starkville North, Colorado, and attributed it to “periods of fire consuming the vestigial or dead organic matter.”

**Origin of carbon.** The spheroid clusters (Fig. 1, A and C) presumably formed in a hot gas or flame, as no other process is known to produce this morphology. The K-T impact could certainly give high temperatures, but where did the carbon come from? Three possibilities come to mind.

The meteorite itself is not a likely source. Though the most primitive meteorites, with 3.5 percent carbon, contain about half the total carbon needed to yield 0.02 g of carbon per square centimeter along with 100 ng of iridium per square centimeter, more than 97 percent of this carbon is in the form of organic matter or carbonate rather than elemental carbon. It is unlikely that the organic carbon in the fireball, as primitive meteorites—even before mixing with crustal rock, ocean water, or air—are too oxidized to permit survival of carbon at high temperatures. This argument is further supported by isotopic data (12). A comet would serve no better: although the total carbon content is somewhat higher, so is the oxygen-carbon ratio (16), which would drive the equilibrium toward CO rather than carbon.

A second source is fossil carbon from the crater. The mean crustal abundance of fossil organic carbon is  $2300 \text{ g/cm}^2$ ; a crater of 100 to 150 km in diameter, as expected for a 10-km meteorite, in average terrain would excavate two to four times the required amount of carbon. The problem again is how to convert this

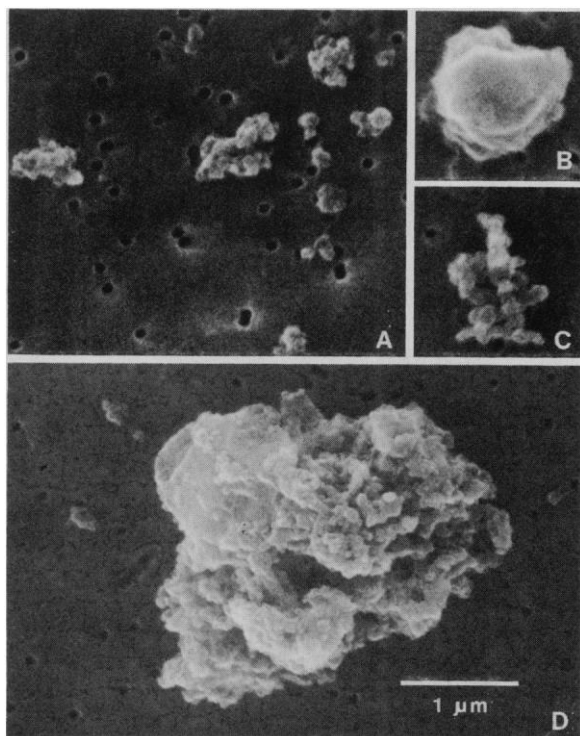


Fig. 1. Carbon from the K-T boundary shows a bimodal size distribution. Most particles are submicrometer-sized and show the characteristic clusterlike morphology of soot. (A) Stevns Klint, Denmark. (B) Dense, platy particles are less abundant in boundary clay, but dominate in the adjoining marl layers (upper marl; Nye Kløv, Denmark). (C) Clusterlike particles from Caravaca, Spain. (D) Micrometer-sized particles form a second population; they are irregular but also fall within the morphologic range of soot (New Zealand).

organic carbon to elemental carbon of the right morphology, in an oxidizing fireball. The K-T impact apparently took place in the ocean (17, 18) and so the oxidizing character of rock vapor would be further enhanced by water vapor.

**Wildfires.** A third possibility is a major wildfire. Even an impact in the ocean can ignite wildfires on continents more than  $10^3$  km away, by radiation from the fireball and especially the expanding cloud of rock vapor (19). The radiation and fallout would ignite vegetation and pyrolyze organic matter in the topsoil. Once started, such a fire could spread over an entire continent, and the resulting winds may disperse the soot worldwide. If the impact happened in the Bering Sea (17), then both Eurasia and North America would be within ignition range.

On balance, wildfires seem to be the most plausible source of the K-T soot layer, although fossil carbon cannot be ruled out without a detailed understanding of carbon condensation chemistry in the fireball.

The scale of the putative wildfires must have been enormous. The surface abundance of carbon at the K-T boundary,  $0.021 \text{ g/cm}^2$ , is 10 percent of the total present biomass carbon on the earth,  $0.2 \text{ g/cm}^2$  (20), or 4 percent of the precivilization level of  $0.5 \text{ g/cm}^2$  (21), which may be more appropriate for the Cretaceous.

**Extinction mechanisms.** The K-T carbon, if dispersed in the atmosphere, would contribute to three of four proposed extinction mechanisms. First, it would absorb sunlight far more efficiently than does rock dust, which alone has been considered thus far (1). An atmospheric load of  $0.021 \text{ g}$  of carbon per square centimeter, with an absorption coefficient typical of smoke ( $1$  to  $6 \text{ m}^2/\text{g}$ ) (22) would give an optical depth of 200 to 1200, thus absorbing virtually all the light and blocking photosynthesis. Second, the pyrotoxins formed during combustion would harm most land life. Carbon monoxide alone, if produced in the same amount as soot, would reach 50 ppm in the atmosphere, a distinctly toxic level. Third, the soot would cool the earth by the "nuclear winter" mechanism (22–24), until most of the carbon (>99 percent) had settled out of the atmosphere.

The soot in the K-T clay thus is an ancient analog of the smoke cloud predicted for nuclear war. It may therefore help determine some important parameters for nuclear winter calculations.

1) The size distribution of K-T carbon is log-normal (Fig. 2), with parameters similar to the average values used in a

Table 2. Elemental carbon at the K-T boundary.

Locality*	Content		Flux‡ ( $\mu\text{g cm}^{-2}$ $\text{year}^{-1}$ )
	Parts per million†	Grams per square centimeter	
Denmark, K-T	3600	0.027	27,000
New Zealand, K-T	5700	0.015	15,000
Spain, K-T	5800	0.022	22,000
Mean		$0.021 \pm 0.006$	21,000
Denmark			
Above K-T	1040		10
Below K-T	950		22
North Pacific, Recent (14)	$580 \pm 260$		$1.8 \pm 1.9$

\*See Table 1 for sample locations. The North Pacific value is the average of 23 samples between  $10^\circ$  and  $60^\circ\text{N}$  (14); the error is the standard deviation of the population. †All concentrations refer to the bulk, dry sediment. Replicate samples generally agreed to within 20 percent: New Zealand values, 5600 and 5700; Denmark above K-T, 1150 and 1140, and below K-T, 1170 and 950 ppm; only the Danish K-T samples gave large scatter: 2800, 3600, and 6500 ppm. ‡Fluxes were obtained as follows. K-T boundary clays: assumed deposition time of 1 year. Danish samples above and below boundary: sedimentation rates of 0.005 and 0.012  $\text{cm/year}$  (13). North Pacific sediments: mean of 9 flux values between  $10^\circ$  and  $60^\circ\text{N}$  (14).

National Academy of Sciences report on nuclear winter (22): modal particle radius  $0.11$  compared to  $0.10 \mu\text{m}$  and dispersion index  $\gamma = 1.6$  compared to  $2.0$  (25). The smaller  $\gamma$  implies a more sharply peaked distribution, but is within the range for smoke,  $1.5$  to  $2.5$  (22).

2) The geographic distribution of soot seems to have been worldwide and fairly uniform, as the two European sites and the antipodal New Zealand site differ by only a factor of 2 (Table 2).

3) The soot production efficiency was an order of magnitude higher than the  $0.24$  (22) or  $0.6$  percent (24) assumed in recent studies of postnuclear fires. As noted above, the K-T carbon layer represents some 4 percent of the total land

biomass, so that the soot yield was 4 percent if the whole earth burned down or even greater if only part of it did. Soot yields increase when the supply of oxygen is limited, and this would certainly be the case when Amazon-sized areas ignite at once. On the other hand, perhaps much of the soot came from fossil carbon rather than biomass.

These data suggest that some of the assumptions used in nuclear winter scenarios were too optimistic. The soot yield is higher and the global soot distribution is more uniform, so that cooling would be more pervasive and lasting. On the other hand, the effects on the biosphere, though disastrous, were not apocalyptic: about half the known gen-

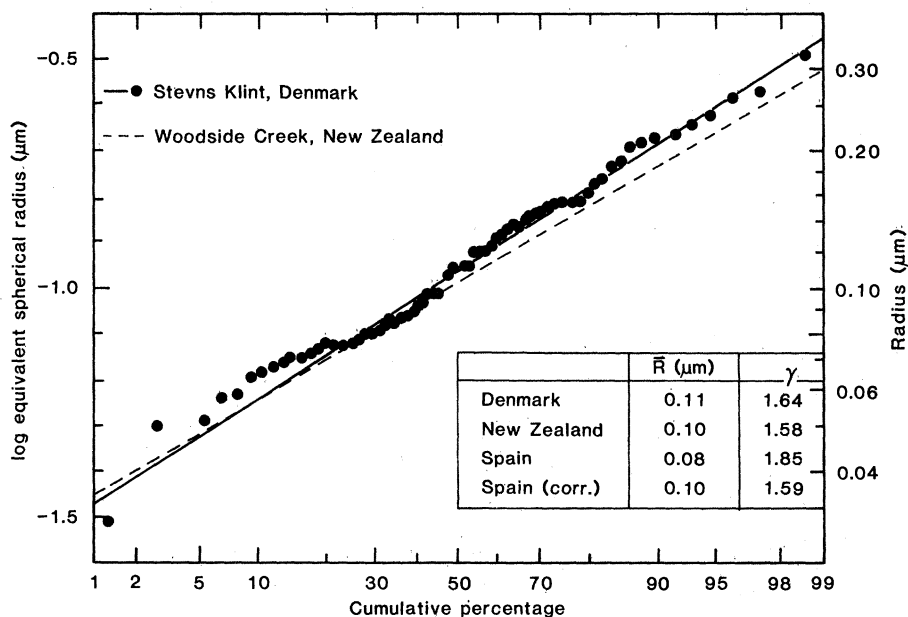


Fig. 2. Submicrometer-sized carbon particles from three K-T boundary sites show similar, log-normal size distributions, with modal radius  $\bar{R}$  and dispersion  $\gamma$  in the range for soot (25). The Spanish site gives a steeper distribution displaced to smaller sizes, but this may reflect surficial oxidation of carbon in the  $\text{Fe}^{3+}$ -rich sediment. When an assumed, constant correction of  $0.017 \mu\text{m}$  is added to the radius of each grain, both  $\bar{R}$  and  $\gamma$  approach the value for the other two sites.

era did survive (26), even though the K-T impact, at  $10^{30}$  ergs, far exceeded the total explosive power of  $3 \times 10^{26}$  ergs assumed for nuclear winter. Evidently the geologic record can provide data not only on an ancient cataclysm but also on similar disasters, manmade or natural, that threaten the earth in the future.

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of 0.2W/ml. Only the primary, flame-welded aggregates are expected to survive this treatment; any secondary aggregates formed in the atmosphere, sediment, or laboratory should have broken up.

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27. This report is dedicated to the memory of Serge Regnier, principal organizer of the Bordeaux meeting of the Meteoritical Society that provided much of the impetus for the work reported here. We gratefully acknowledge the donors of the samples: H. J. Hansen, R. R. Brooks, and R. Ganapathy. We are also indebted to E. D. Goldberg for valuable advice and a manuscript copy of his book and to O. Eugster and J. Geiss for unpublished data on an early search for noble gases in material from the Danish K-T boundary. A. M. Davis, A. Hashimoto, and T. K. Mayeda kindly provided advice and assistance in the identification of minerals. J. P. Bradley of Walter C. McCrone Associates provided Fig. 1D. Supported in part by NSF grant EAR 8212355 and NASA grant NAG 9-52.

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## Halite Particles Injected into the Stratosphere by the 1982 El Chichón Eruption

**Abstract.** Halite particles about 2 micrometers in size were collected by a quartz crystal microbalance cascade impactor from the El Chichón eruption cloud in the lower stratosphere during April and May 1982. These particles are probably derived from the erupted chloride-rich, alkaline magma. Enrichments of hydrogen chloride and increases in optical depolarization in the eruption cloud observed by lidar measurements may reflect the influence of the halite particles. There is evidence that the halite particles reacted with sulfuric acid after about 1 month, releasing gaseous hydrogen chloride, which can influence the catalytic destruction of ozone in the stratosphere.

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The El Chichón volcano in southern Mexico (17.3°N, 93.2°W) injected large amounts of gases and particles into the stratosphere during eruptions between 28 March and 4 April 1982. Mass and size distributions of the aerosol were measured in situ in a layer of eruption

material in the lower stratosphere (altitude, 18 to 21 km) by a quartz crystal microbalance (QCM) cascade impactor (1) on a U-2 aircraft. Size-fractionated aerosol particles were collected by the QCM and were analyzed after the flight. The general character of the aerosol particles was similar to that in other volcanic clouds in most respects (2, 3), but scanning electron microscopy (SEM) and x-ray energy spectroscopy (XES) revealed the presence of NaCl particles (halites), which are rarely if ever seen at these altitudes. These halite particles are a natural source of chloride that, in addition to anthropogenic sources (such as chlorofluoromethanes), could have a major impact on the stratosphere through the catalytic destruction of ozone (4). The particles may have also caused the enhanced optical depolarization in the El Chichón cloud observed by lidar measurements (5).

The monodisperse nature of the halite aerosol was evident because almost all the particles appeared on impactor stage 5 [corresponding to a geometric mean diameter (GMD) of 2  $\mu$ m], except for a few that appeared on stage 4 (GMD, 4.5  $\mu$ m). On the basis of the SEM data from impactor stage 5 and the mass-loading rate inferred from the QCM data, we

Table 1. U-2 aircraft sampling flights from Moffett Field, California (~37°N, 122°W).

Date (1982)	Destination	
15 April	23°N	111°W
4 May	23°N	111°W
20 July	23°N	111°W
23 July	23°N	111°W
4 November	32°N	106°W
5 November	41°N	106°W
15 December	23°N	111°W
17 December	40°N	105°W