LBL-9666
Preliminary Copy
Revised 12-12-79

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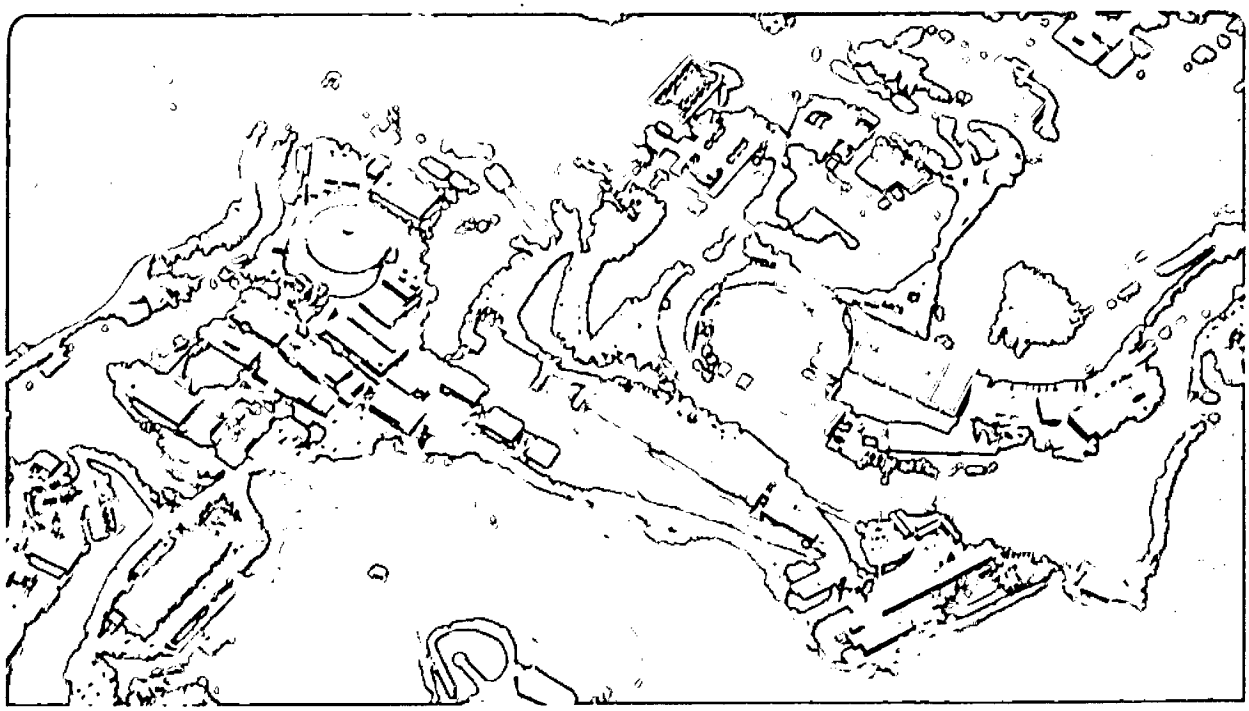
Luis W. Alvarez, Walter Alvarez, Frank Asaro,
and Helen V. Michel

November 1979



Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

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EXTRATERRESTRIAL CAUSE FOR THE CRETACEOUS-TERTIARY EXTINCTION: EXPERIMENT AND THEORY

Luis W. Alvarez*+

Walter Alvarez**

Frank Asaro*

Helen V. Michel*

University of California
Berkeley, California 94720

*Lawrence Berkeley Laboratory

**Department of Geology and Geophysics

+Space Sciences Laboratory

ABSTRACT

Platinum metals are depleted in the earth's crust relative to their cosmic abundance; concentrations of these elements in deep-sea sediments may thus indicate influxes of extraterrestrial material. Deep-sea limestones exposed in Italy and Denmark show iridium increases of about 30 and 160 times, respectively, above the background level at precisely the time of the Cretaceous Tertiary extinctions, 65 m.y. ago. Reasons are given to indicate that this iridium is of extraterrestrial origin, but did not come from a nearby supernova. We suggest a new hypothesis to account for the extinctions and the iridium observations. Impact of a large Apollo object would inject about 100 times the object's mass into the atmosphere as

pulverized rock; about 20 percent of this dust would stay in the stratosphere for 3-5 years and be distributed world-wide. The resulting darkness would suppress photosynthesis and the expected biological consequences match quite closely the extinctions observed in the paleontological record. One prediction of this hypothesis has been verified: the chemical composition of the boundary clay is markedly different from that of clay mixed with the Cretaceous and Tertiary limestones, which are chemically similar to each other. Four different and quite independent estimates of the diameter of the asteroid give values that lie in the range 10 ± 3 km. This hypothesis has not been proven, but deserves further testing.

INTRODUCTION

Abundant fossil remains provide a record of organic evolution over the last 570 million years of Earth history. During this time there have been five great biological crises, during which many groups of organisms died out. Numerous crises of less severity are also recorded.

The most recent of the great extinctions is used to define the boundary between the Cretaceous and Tertiary Periods, about 65 million years ago. At this time, the marine reptiles, the flying reptiles, and both orders of dinosaurs died out (1). Extinctions occurred at various taxonomic levels among the marine invertebrates, for example, the ammonoid and belemnoid cephalopods and various groups of gastropods, pelecypods, brachiopods, and echinoderms. Dramatic extinctions occurred among the microscopic floating animals and plants; both the calcareous planktonic foraminifera and the calcareous nannoplankton were nearly exterminated, with only a few species surviving the crisis. On the other hand, some groups were little affected, including the land plants, crocodiles, snakes, birds, mammals, and many kinds of invertebrates.

Many hypotheses have been proposed to explain the Cretaceous-Tertiary extinctions (2), and two recent meetings on the topic (3,4) produced no sign of a consensus. Suggested causes include gradual or rapid changes in oceanographic, atmospheric, or climatic conditions (5) due either to a random (6) or a cyclical (7) coincidence of causative factors, the effect of a magnetic reversal (8) or a nearby supernova (9).

Various mechanisms specific to a single group, such as disease or destruction of dinosaur eggs by mammals, are incapable of explaining why the extinctions affected many groups.

A major obstacle to determining the cause of the extinction is that virtually all available information on events at the time of the crisis deals with biological changes seen in the paleontological record, and is therefore inherently indirect. Little physical evidence is available, and it, also, is indirect. This includes variations in stable oxygen and carbon isotopic ratios across the boundary in pelagic sediments, which may reflect changes in temperature, salinity, oxygenation, and organic productivity of the ocean water, and which are not easy to interpret (10). These isotopic changes are not particularly striking and, taken by themselves, would not suggest a dramatic crisis. Changes in minor and trace element levels at the Cretaceous-Tertiary boundary have been noted from limestone sections in Denmark and Italy (11), but these data also present interpretational difficulties. It is also noteworthy that in pelagic marine sequences, where nearly continuous deposition is to be expected, the Cretaceous-Tertiary boundary is almost invariably marked by a hiatus (12). Because of the lack of clear evidence outside the field of paleontology, it is still possible to argue that the various extinctions were not exactly synchronous, that they were produced by a fortuitous combination of ordinary environmental stresses, and that there really was no Cretaceous-Tertiary boundary crisis (6).

In this paper we present, for the first time, direct physical evidence for an unusual event at exactly the time of the extinctions in the planktonic realm. None of the current hypotheses adequately accounts for this evidence, but we have developed a new hypothesis that appears to offer a satisfactory explanation for nearly all the available paleontological and physical evidence.

In this paper, we first discuss the use of anomalous concentrations of platinum-group elements in deep-sea sediments as indicators of influxes of extraterrestrial material; the technique is based on the extreme depletion of these elements in the earth's crust. We then present the results of our measurements by neutron activation analysis of iridium in Upper Cretaceous-Lower Tertiary marine limestones from two areas. The first area is in the Umbrian Apennines of Italy, where recent paleomagnetic studies have established the timing of the planktonic extinction in the sequence of geomagnetic polarity reversals (13). Analysis of the acid-insoluble clay fraction of the limestone shows Ir levels of about 0.3 ppb in the uppermost Cretaceous. The Cretaceous-Tertiary boundary is marked by a 1 cm clay layer in which Ir suddenly jumps to about an average of 6 ppb in the acid-insoluble residue, then gradually decreases to the previous background level in less than 1 m above the boundary. To test whether this is a local phenomenon, we have analysed samples from a classic exposure of the Cretaceous-Tertiary boundary near Copenhagen and found Ir levels nearly ten times higher than are seen in the Italian sections, a 160 fold increase above the background level.

These results lead to the hypothesis that the anomalous Ir was probably introduced into the Earth's atmosphere as part of an abnormal influx of extraterrestrial material at the time of the extinctions. One possible extraterrestrial cause of the extinctions, a nearby supernova, has been debated for some time. We report three lines of evidence for rejecting the supernova hypothesis: (a) ^{244}Pu (80×10^6 yr half life) should be detectable along with Ir in the boundary clay if a supernova is responsible for the Ir anomaly, but if present, ^{244}Pu is at least a factor of 10 below predicted levels. This fact alone is not conclusive, as oceanic chemistry could alter the Ir/Pu ratio. (b) The ratio $^{191}Ir/^{193}Ir$ might well be significantly different in supernova material at the boundary and in normal terrestrial material, but no difference was found. (c) To account for the Ir seen in the boundary clay, a hypothetical supernova would have to be so close that the probability of such an event becomes vanishingly small. We conclude that the extraterrestrial material introduced at the time of the extinctions came from a solar system source, and not a galactic source, e.g., it was not caused by a supernova or passage of the earth through the galactic arms. (Note added Dec. 12, 1979: W. M. Napier and S. V. M. Clube suggested the latter in Nature, 282, 455-9 (1979)).

After consideration and rejection of a number of possible explanations for the extraterrestrial material and the extinctions, we have developed the following hypothesis, which seems to account for most or all of the relevant observations: A number of asteroids have

orbits with perihelions inside the Earth's orbit; these are called "Apollo Objects." Collisions of Apollo objects with the Earth are inevitable, and the many known meteor craters are the result. We suggest that impact of a large Apollo object was the cause of the Cretaceous-Tertiary extinctions. Four independent calculations indicate that the impacting Apollo object had a diameter of approximately 10 km. Such an impact would produce a crater approximately 175 km in diameter. The material expelled from the crater would weigh about 100 times as much as the asteroid, and much of the combined mass would stay aloft for several years as dust in the stratosphere. Comparisons of this effect with the much smaller explosion of Krakatoa in 1883 indicate that the atmosphere would become opaque to visible light until the dust settled, although enough heat would reach the surface to prevent a major drop in temperature.

The resulting suppression of photosynthesis appears to be capable of explaining the major features of the extinctions. The marine phytoplankton would die out through loss of light with which to carry on photosynthesis. This would cause the collapse of the food chain supporting the planktonic foraminifera and the large open marine fauna, including ammonites, belemnites, and marine reptiles. Terrestrial plants would cease to grow, but would not die out, because their seeds could lie dormant until the dust settled and light returned. Small terrestrial animals including the mammals as well as the birds, could survive on a food chain based on seeds and nuts. The herbivorous dinosaurs, and the carnivorous dinosaurs that preyed on

them, depended on a food chain based on growing vegetation; this chain would have collapsed. The survival of many marine invertebrates and aquatic animals such as crocodiles may be due to their ability to utilize food chains based on nutrients derived from decaying land plants carried by rivers to the shallow seas.

Thus it appears that a highly probable sequence of events could produce the observed extinctions, and independent physical evidence for this mechanism has now been found.

IDENTIFICATION OF EXTRA-TERRESTRIAL PLATINUM METALS IN DEEP-SEA SEDIMENTS

This study began with the realization that the platinum group metals and some related elements (Pt, Ir, Os, Rh) are 10^3 - 10^4 times more abundant in chondritic meteorites than they are in the earth's crust and upper mantle (14). Carbonaceous chondrites are thought to come from undifferentiated bodies giving a close approximation to average solar system elemental abundance. Depletion of the platinum group elements in the earth's crust and upper mantle is probably the result of concentration of these elements in the earth's core.

Platinum group elements are apparently difficult to detect without chemical separation in most sedimentary rocks, judging from the few published data (15). This is reasonable, because in addition to their scarcity in rocks from which sediments are derived, they are also extremely insoluble and should be present only at very low levels in river and sea water (again, data are not available).

These considerations suggested to us that much of the iridium to be found in sedimentary rocks comes from ablated meteoritic dust, deposited at a roughly constant rate. We suspected that measurement of the platinum group metals might shed light on the time interval represented by the 1 cm thick clay layer that marks the Cretaceous-Tertiary boundary in the Umbrian Appennines. We therefore undertook an investigation of the abundance of iridium, which can easily be determined at low limits by neutron activation analysis (NAA) because of its large capture cross section for slow neutrons, and because the gamma rays given off during de-excitation of the decay product are not masked by other gamma rays. The other platinum group elements are more difficult to determine by NAA.

After we had begun our work, we learned that this method of identifying extraterrestrial material had been suggested in the early 1950's by Pettersson and Rotschi and by Goldschmidt (16) and actually carried out in the 1960's by Barker and Anders (17). Subsequently the method was used by Ganapathy et al. (14) to demonstrate an extra-terrestrial origin for silicate spherules in deep-sea sediments. Sarna-Wojcicki et al. (18) suggested that meteoritic dust accumulation in soil layers might enhance the abundance of iridium sufficiently to permit its use as a dating tool.

We have found that iridium <u>does</u> show anomalously high abundances at exactly the stratigraphic position of the Cretaceous-Tertiary boundary in two areas, Italy and Denmark, where marine limestones preserve a complete or nearly complete record of this stratigraphy.

THE ITALIAN STRATIGRAPHIC SECTIONS

Many aspects of earth history are best recorded in sedimentary rocks deposited in the relatively quiet waters of the deep sea as individual grains settle to the bottom. These slowly but continuously deposited pelagic sediments are present in many parts of the ocean and have been sampled by the Deep Sea Drilling Project (19). Although pelagic sedimentary sequences normally remain below the sea bottom, they have in a few places been uplifted and exposed at the surface as a result of deformation during mountain building. Such exposures are valuable because the rocks can be examined over large areas, and have not suffered the disturbances sometimes produced by drilling.

In the Umbrian Apennines of northern peninsular Italy there are exposures of pelagic sedimentary rocks representing the time from Early Jurassic to Oligocene, ca. 185-37 m.y.B.P. (20). The Cretaceous-Tertiary boundary occurs within a portion of the sequence formed by pink limestone containing a variable amount of clay. This limestone, the Scaglia rossa, has a matrix of coccoliths and coccolith fragments (calcite platelets on the order of 1 micron in size, secreted by algae living in the surface waters) and a rich assemblage of foraminiferal tests (calcite shells, generally in the size range 0.1-2.0 mm, produced by single-celled animals that float in the surface waters).

In some Umbrian sections there is a hiatus in the sedimentary record across the Cretaceous-Tertiary boundary, sometimes with signs of soft-sediment slumping. Where the sequence is apparently complete, foraminifera typical of the Upper Cretaceous (notably the genus

Globotruncana) disappear abruptly, and are replaced by the basal Tertiary foraminifer Globigerina eugubina (21). This change is easy to recognize because G. eugubina, unlike the Globotruncanids, is too small to see with the naked eye or the hand lens (Plate 1). The coccoliths also show an abrupt change, with disappearance of Cretaceous forms, at exactly the same level as the foraminiferal change, although this was not recognized until more recently, because these fossils can only be studied with the scanning electron microscope (22). No other types of fossils are present in the Umbrian limestones near the Cretaceous-Tertiary boundary in numbers sufficient to be useful.

In well-exposed complete sections there is a bed of clay about 1 cm thick between the highest Cretaceous and the lowest Tertiary limestone beds (23). This clay contains little or no original $CaCO_3$, although secondary calcite crystallized during deformation is present in at least one section. Thus there is no record of the biological changes during the time interval represented by the clay. The boundary is further marked by a zone in the uppermost Cretaceous in which the normally pink limestone is white in color. This zone is 0.3-1.0 m thick, varying from section to section. Its lower boundary is a gradational color change; its upper boundary is abrupt and coincides with the faunal and floral extinctions. In one section (Contessa) we can see that the lower 5 mm of the boundary clay is gray and the upper 5 mm is red, thus placing the upper boundary of the zone in the middle of the clay layer.

The best known of the Umbrian sections is in the Bottaccione Gorge near Gubbio. Here some of the first work on the identification of foraminifera in thin section was carried out (24); the oldest known Tertiary foraminifer, Globigerina eugubina, was recognized, named, and used to define the basal Tertiary biozone (21); the geomagnetic reversal stratigraphy of the Upper Cretaceous and Paleocene was established, correlated to the marine magnetic anomaly sequence, and dated with foraminifera (13); and the extinction of most of the nannoplankton was shown to be synchronous with the disappearance of the genus Globotruncana (22).

In the present study we have analyzed 15 samples from the Bottaccione section, supplemented by 4 samples from the Contessa section, 2 km to the northwest, where details of the Cretaceous-Tertiary boundary are clearly exposed. Samples from 3 sections about 30 km to the north of Gubbio have been analyzed to test the possibility that the iridium anomaly is a local feature restricted to the Gubbio area (25).

RESULTS FROM THE ITALIAN SECTIONS

Our first experiments involved NAA of 9 samples from the Bottaccione section (2 limestone samples from immediately above and below the boundary plus 7 limestone samples spaced over 325 m of the Cretaceous). This was supplemented by 3 samples from the nearby Contessa section (2 from the boundary clay and one from the basal Tertiary bed). The stratigraphic positions of these samples are shown in Fig. 1.

Twenty-eight elements were selected for study because of their favorable nuclear properties, especially neutron capture cross-sections, half lives, and gamma-ray energies. The results of these analyses are presented in Fig. 2 on a logarithmic plot to facilitate comparison of the relative changes in elements over a wide range of concentrations. The only preparation given to these samples was removal of the $CaCO_3$ fraction by solution in dilute nitric acid. Figure 2 shows elemental abundances as gram of element per gram of insoluble clay residue. The limestones generally contain about 5 percent clay. The boundary clay layer contains about 50 percent $CaCO_3$, but this is coarse grained calcite that probably crystallized during deformation long after deposition; no primary $CaCO_3$ is present.

One of the results of this work is that although 27 of the 28 elements show very similar patterns of abundance variation, iridium shows a grossly different behavior; it increases by a factor of about 30 in coincidence with the Cretaceous-Tertiary boundary, while none of the other elements shows as much as a doubling with respect to an "average behavior" shown in the lower right panel of Fig. 2. This diagram is a best "eyeball fit" to the seven rare earth curves (slid left or right to overlap to the greatest extent). The "flags" on the 12 points in that diagram do not indicate errors, but instead denote a "plus or minus" change of a factor of 1.3. All of the rare earth points fit within these limits, and so do most of the points corresponding to the abundances of the other elements. This "average

curve" fits very well to the small changes in relative abundances of most of the observed elements, extending over a time period of more than 40 million years. This makes the behavior of iridium all the more extraordinary, since it is the only one of the 28 elements that departs by more than a factor of two from the average behavior, and then only in the immediate vicinity of the Cretaceous-Tertiary boundary. Such a specific enrichment of iridium, if it occurs world-wide, would be difficult to obtain from terrestrial materials and strongly suggests an extraterrestrial origin. This interpretation is justified in detail in a later section.

In follow-up experiments we analyzed 17 more samples for iridium. This second group included 5 samples from the Bottaccione section, 8 from Gorgo a Cerbara (28 km north of Gubbio), and 4 large samples of the boundary clay from the two sections near Gubbio and two sections about 30 km to the north.

Figure 3 shows the results of the 29 Ir analyses completed to date on Italian samples. Note that the section is enlarged and the scale is linear in the vicinity of the Cretaceous-Tertiary boundary, where details are important, but changes to logarithmic to show results from 350 m below to 50 m above the boundary. It is also important to note that analyses from 5 stratigraphic sections are plotted on the same diagram on the basis of their stratigraphic position above or below the boundary. Because of slight differences in sedimentation rate that probably exist from one section to the next, the chronologic sequence of samples from different sections may not be exactly

correct. Nevertheless the figure gives a clear picture of the general trend of iridium concentrations as a function of stratigraphic level.

The pattern, based especially on the samples from the Bottaccione Gorge and Gorgo a Cerbara shows a steady background level of a few tenths of a ppb (~0.3×10^{-9}) throughout the Upper Cretaceous, continuing into the uppermost bed of the Cretaceous. This level increases abruptly by a factor of over 20, to 6.35×10^{-9} (average in the clay residues of the four large samples) in the 1 cm clay bed at the boundary. Ir levels are high in clay residues from the first few beds of Tertiary limestone, but fall off to background levels by 1 m above the boundary. For comparison, the upper dashed line in Fig. 3 shows an exponential decay from the boundary clay Ir level with a half height of 4.6 cm. Figure 4 shows a typical gamma-ray spectrum used to measure the abundance of Ir.

In a later section we will argue that this iridium pattern is indicative of a sudden influx of extraterrestrial material at exactly the time of the Cretaceous-Tertiary extinctions. First, however, it is necessary to show that the iridium anomaly is not a local feature confined to the Umbrian limestones. Complete sections across the boundary are quite rare, but we were able to sample a nearly complete section near Copenhagen, the type area of the Danian Stage (lowest stage of the Tertiary).

THE DANISH SECTION

The sea cliff of Stevns Klint, about 50 km south of Copenhagen is a classical area for the Cretaceous-Tertiary boundary and for the

Danian, or basal stage of the Tertiary. A collection of up-to-date papers on this and nearby areas has recently been published, which includes a full bibliography of earlier works (26).

Our samples were taken at Højerup Church (27). At this locality the Maastrichtian, or uppermost Cretaceous, is represented by white chalk marked by undulating layers of black chert nodules with amplitudes of a few meters and wavelengths of 10-50 m (28). These undulations are considered to represent bryozoan banks (29). The Cretaceous-Tertiary boundary is marked by the Fiskeler, or fish clay, up to 35 cm thick in the deepest parts of the basins between bryozoan banks (28), but commonly only a few cm thick, thinning or disappearing over the tops of the banks. The fish clay at Højerup Church was studied in detail by Christensen et al. (28), who subdivided it into four thin units; we have analyzed a sample mixing the two internal layers (units III and IV of Christensen et al.). These layers are black or dark gray, and the lower one contains pyrite concretions; the layers below and above (II and V) are light gray in color. Undisturbed lamination in bed IV suggests that no bottom fauna was present during its deposition (28).

Above the fish clay, the Cerithium limestone is present to a thickness of about 50 cm in the small basins, disappearing over the banks. It is hard, yellowish in color, and cut by abundant burrows. Above this is a thick bryozoan limestone. The Maastrichtian carbonates are soft chalks, while the Danian carbonates are indurated limestones. As a result, the Danian forms an overhanging cliff, with the fish clay making a recess at the base of the overhang.

The presence of a thin clay layer at the Cretaceous-Tertiary boundary in both the Italian and Danish sections is quite striking. However, there are notable differences as well. The Danish sequence was clearly deposited in shallower water. Stevns Klint is located on a zone of structural uplift related to the Ringkøbing-Fyn High (30), and the Danish limestones preserve an extensive bottom-dwelling fauna of bivalves (31), echinoderms (32), bryozoans (33), and corals (34).

Foraminiferal (35) and coccolith (36) zonation indicates that the Cretaceous-Tertiary boundaries at Gubbio and Stevns Klint are at least approximately contemporaneous, and they may well be exactly synchronous. However, no paleomagnetic results are available from Stevns Klint, so synchroneity cannot be tested by reversal stratigraphy.

RESULTS FROM THE DANISH SECTION

Seven samples were taken from near the T/C boundary as shown in Fig. 5. Fractions of each sample were treated with dilute nitric acid, and the residues were filtered, washed and heated to 800°C. The acid treatment should have dissolved all carbonates, and some oxides, sulfides and phosphates, but not much of the silicate minerals. The yield of acid-insoluble residue was 44.5 percent for the boundary fish clay and varied from 0.62 percent to 3.3 percent for the pelagic limestones as shown in Table I.

Neutron activation analyses (37) and X-ray fluorescence measurements (38,39) were made on all 7 samples both before and after the acid treatment. This measurement regime was more sophisticated than that used for the Italian sections studied earlier, and 48 useful elements were determined.

The major element abundances in the acid-insoluble residues of the three Cretaceous limestones are very similar to each other as shown in Fig. 6. The three Tertiary limestones are likewise very similar to each other in chemical composition. There are distinguishable differences (e.g., Na and Ti) between the Cretaceous and Tertiary patterns, but they still are very much alike. The boundary layer residue, however, is much different from the Cretaceous and Tertiary pattern in that it is much lower in Na and K and much higher in Fe and Mg. This difference also appears in most of the trace element abundances as shown in Table II. Of 38 trace and minor elements which were usefully detected, 15 were enhanced in the fish clay residue compared to the pelagic residues, 19 were depleted and 4 were comparable. Most of these effects were very marked, and examples are shown in Fig. 7.

Although it might be possible to explain the abundance of some of the enhanced elements by oceanic chemistry effects involving sulfide-producing organisms, this would not explain the depleted elements. This suggests that a different source of clay material was introduced during the boundary formation.

The abundance of Ir in the Danish boundary layer (whole-rock) is $(28.6 \pm 1.3) \times 10^{-9}$. Some Ir dissolved in the nitric acid, and its abundance in the insoluble residue was 41.6±1.8 ppb. The Ir in the residue from 0.5 meters below the boundary was 0.73±0.08 ppb, and the amount in the other two Cretaceous residues had a weighted average of 0.26±0.08 ppb. In the residue from 0.7 meters above the boundary the abundance was 0.36±0.06 ppb, and it was less than 0.3 ppb in the other

two Tertiary residues. Thus the Ir in the boundary layer residue rises by about a factor of 160 over the background level (~0.26 ppb). A one centimeter thickness of this layer would have about 72×10^{-9} gm Ir/cm^2. To test if there was enough Ir in the sea water to contribute to this value, we made a measurement of the Ir in the ocean off the central California coast. None was detected in water passed through a .45 micron filter with an upper limit of 8×10^{-13} gm Ir/gm sea water. If the depth of the shallow ancient Danish sea is assumed to be less than 100 meters and our limit on Ir in sea water is applicable, then the maximum Ir in the 100 meter column of water would be 8×10^{-9} gm/cm^2, almost a factor of 10 lower than the observed value. So there was probably not enough Ir stored in the sea water to explain the amount observed in the Danish boundary.

As most terrestrial basic rocks have Ir abundances of less than 1 ppb, and ultrabasic rocks have values of less than 20 ppb (40), extraterrestrial sources with abundances of about 400 ppb seem more likely to produce a sediment with 29 ppb Ir than do terrestrial sources. It is not possible to rule out terrestrial sources completely. There are nickel sulfide and chromite ores (40) for example which have Ir in the hundreds and thousands of ppb, respectively. The Danish boundary layer, however, does not have enough nickel (506 ppm) or chromium (165 ppm) to explain its Ir in this fashion, unless the oceanic chemistry concentrated Ir preferentially and disposed of the other elements elsewhere. The probability of these effects occurring on a world-wide basis seems less likely than an extraterrestrial

origin for the Ir. If the Ir arose from such a source, the Ni in the Danish boundary layer is close to the value expected if the Ir were chondritic and there were no Ni-Ir fractionation. All the elements which we measure and which appear in meteorites are at least as abundant in the boundary layer as would be expected from their abundance in carbonaceous chondrites and our Ir value. There are no apparent chemical conflicts with an extraterrestrial explanation for the Ir. As mentioned in the next section, however, the Ir abundance is higher than would be expected by the mixing of a carbonaceous chondrite with a hundred times its mass of terrestrial material.

THE BOUNDARY LAYERS

The T/C boundary layer in the Contessa section near Gubbio is about 1 cm thick and contains a variable amount of recrystallized calcite. The acid-insoluble or clay component averages about 50 percent of the total mass. The whole rock composition (a mixture of red and gray clay) is shown in Table III. There are two detectable layers, each about 0.5 cm thick, the upper being red in color and the lower gray. The iron content which may explain the color, is significantly higher in the residue of the red layer (7.7 vs. 4.5 percent) than in the gray and so is the Ir (9.1±.6 vs. 4.0±.6 ppb). Boundary samples were analyzed from the Bottaccione Gorge nearby and two other areas about 30 km to the north. All of these samples were roughly similar in composition with respect to all measured elements.

It was indicated earlier that in samples taken near the Italian boundary layer, the chemical compositions of all clay fractions were

roughly the same except for the element Ir. There are some discernible differences, however, between the boundary layer and the limestones above and below as shown in Fig. 8. These differences suggest that the boundary layer clay fraction or a component in it may have had a different origin than the Cretaceous and Tertiary clay fractions.

The Danish T/C boundary layer is somewhat thicker than 1 cm and has been classified into 4 sections as mentioned in the Danish Section of this paper. Only a single mixed sample from the center two sections was measured so no information is available on the chemical variations within the boundary. The average Ir abundance is 29 ppb in the whole rock or 65 ppb based on the weight of acid-insoluble residue.

The whole rock abundances and mineral composition are shown in Table III, and pertinent trace elements were shown in Table II. The major silicate minerals were not detected so the other mineral abundances were normalized to give the amount of calcite expected from the Ca measurement. The boundary clay fraction is far different chemically from the limestone clay fractions above and below, which are similar to each other. Pyrite was known to be present in the boundary and was detected in our measurements. Those elements which are greatly enhanced in the boundary relative to the limestone clay fractions are generally those which form water-insoluble sulfides. The trace elements which are depleted are those which often appear as clay components. The element Mg is an exception and may be related to a different, more mafic source for the boundary layer clay than the Tertiary and Cretaceous clays. If the boundary is considered to be a

mixture of two such clays and a third component, the depletion of the alkali elements indicates more than 80 percent of the boundary clay fraction is due to the third component.

It has been shown then that the Danish and, to a lesser extent, the Italian boundary layers are different in composition from the clays which were deposited along with the Cretaceous and Tertiary limestones. This is to be expected in the asteroid impact theory. The asteroid and the terrestrial impact material (if the asteroid impacted on land) could blanket the earth with about a 1 cm thick layer of material.

There are many problems which remain unsolved. There are great differences in chemical composition between the boundary layers in Italy and Denmark. One would expect from the simplest considerations of our hypothesis that the boundary layer is mostly crustal material (enriched in certain elements by the asteroidal matter) which was distributed world-wide in the stratosphere and then fell into the ocean, and that the boundary clay in Italy and Denmark would be indistinguishable. The enhancements of sulfide-insoluble metals in the Danish T/C boundary compared to the Italian might be related to anerobic conditions in the former and aerobic conditions in the latter. H_2S can be produced by bacteria in oxygen deficient waters, and this would precipitate those metals if they were available. The striking depletion of some of the trace elements found in the Danish boundary also remains as one of the unsolved problems. Others are the difference in Ir abundance between the two layers (about an order of

magnitude) and the high absolute abundance of Ir in the Danish layer. Chondritic Ir at 400 ppb mixed with a hundred times its mass of earth should produce a mixture with 4 rather than the 65 ppb of Ir found in Denmark.

A SUDDEN INFLUX OF EXTRATERRESTRIAL MATERIAL

The reasons for considering iridium in pelagic sediments to be largely of extraterrestrial origin have been set out above, and we interpret the iridium in the Italian and Danish limestones in this way.

To test the extraterrestrial origin of the anomalous iridium at the boundary in the Gubbio sections, we have considered the increases in 27 of the 28 elements measured by NAA that would be expected if the iridium in excess of the background level came from either of two possible sources. Fig. 9 shows that if the source had an "average earth's crust" composition (41), increases significantly above those observed would be expected in all 27 elements. However, for a source with "average carbonaceous chondrite" composition (41), only nickel should show an elemental increase greater than that observed. As shown in Fig. 10 this increase in nickel was not observed, but the predicted effect is small and the discrepancy is probably not important. We conclude that the pattern of elemental abundances in the Gubbio sections is compatible with an extraterrestrial source for the anomalous iridium, and incompatible with a crustal source.

Could the anomalous iridium have come from the upper mantle in a large volcanic eruption? It might be expected that the upper mantle would be richer in platinum metals than the crust, since the only

commercial sources of these elements are in mafic rocks (42). However, the few Ir analyses from rocks of upper mantle origin show levels only slightly above those typical of crustal rocks (40).

The anomalous iridium is thus apparently of extraterrestrial origin, and we must now ask whether it is due to an abnormal influx of extraterrestrial material at the time of the extinctions, or whether it is from the same source as the normal background iridium, that is, the steady accumulation of meteorite ablation dust (17), and was concentrated in the boundary rocks by some identifiable mechanism.

There is prima facie evidence for an abnormal influx in the observations that the excess iridium occurs exactly at the time of one of the extinctions, that the extinctions were extraordinary events which may well indicate an extraordinary cause, that the extinctions were clearly world wide, and that the iridium anomaly is now known from two widely-separated areas in western Europe. Furthermore, we will show in a later section that impact of a 10-km Apollo object, an event that probably occurs with about the same frequency as major extinctions, would probably produce the observed physical and biological effects. Nevertheless, one can invent two other scenarios which might lead to concentration of normal background iridium at the boundary. These appear to be considerably less likely than the sudden-influx model, but we cannot definitely rule out either one at the present time.

The first scenario requires a physical or chemical change in the ocean waters at the time of the extinctions leading to extraction of

iridium resident in the sea water. This would require that iridium be present at fairly high concentration in sea water. In addition, it suggests that the positive iridium anomaly should be accompanied by a compensating negative anonaly immediately above, but this is not seen.

The second scenario postulates a reduction in the deposition rate of all components of the pelagic sediment except for the meteoritic dust that carries the concentrated iridium. It is not difficult to account for cessation of $CaCO_3$ deposition: something certainly killed off the $CaCO_3$-fixing nannoplankton during the crisis, and this would stop the rain of $CaCO_3$ particles and produce a rise in the calcite compensation depth (CCD). It is difficult to stop the normal rain of clay particles, but this is also required since the iridium anomaly is seen at Gubbio as about a 30-fold increase in the Ir/clay ratio. One could postulate that little or no clay was carried to the sea by rivers during the extinction event, but we are unable to think of a mechanism that would have this effect; we consider instead a mechanism that might prevent deposition of the clay.

In this scenario an unspecified Cretaceous-Teritary crisis event kills off the nannoplankton and foraminifera and generates bottom currents of an optimum velocity. The rain of $CaCO_3$ stops, the CCD rises, the sea floor suffers some dissolution and a "hard ground" or indurated bottom forms. The bottom currents are sufficient to keep the light, platy clay particles in suspension, sweeping them away toward the deep ocean and not replenishing the areas with clay brought in from elsewhere. Only the Ir-bearing meteorite dust reaches the

bottom; it must occur as particles whose size, shape, and density allow them to sink through currents capable of transporting clay particles. When deposition of $CaCO_3$ and clay resumes at the end of the crisis, bioturbation smears the iridium upward through the first 30 cm of the Tertiary limestone; the hard ground at the top of the Cretaceous prevents burrowing organisms from smearing the iridium downward. At this point there is no boundary clay layer; it is produced much later by dissolution of the uppermost Cretaceous and basal Tertiary limestone, and this dissolution is unrelated to the boundary crisis events.

We feel that this scenario is too contrived. The bottom currents must have been fast enough to transport clay but not fast enough to transport meteoritic dust; the meteoritic dust must occur in a size range that makes this possible; clay must have been swept away from the areas we sampled but not toward them; these specially adjusted currents must have endured for approximately 230,000 years based on Gubbio sedimentation rate calculations, but not have occurred at any of the 18 other time horizons we have sampled in the Italian sections; and all of these features are required in Denmark as well. In addition, in Denmark the chemical composition of the boundary layer is very different from the acid-insoluble fractions of the Cretaceous and Tertiary periods.

We have discussed this scenario at some length since our original reason for undertaking Ir measurements was to develop a sedimentation-rate indicator; is it possible that the boundary iridium anomaly is a

sedimentation-rate effect? Although we cannot completely eliminate this possibility at present, the arguments given above make it seem quite unlikely.

In summary, we conclude that the anomalous iridium concentration at the Cretaceous-Tertiary boundary is best interpreted as indicating an abnormal influx of extraterrestrial material. We now proceed to a consideration of the various hypotheses that might explain our data and account for the mass extinctions.

NEGATIVE RESULTS OF TESTS FOR THE SUPERNOVA HYPOTHESIS

Considerable attention has been given to the hypothesis that the Cretaceous-Tertiary extinctions were the result of a nearby supernova (9), and our first thoughts, after we saw the Ir anomaly, and suspected that it was of extraterrestrial origin, were that if the supernova theory of the extinction was correct, we could verify it. It was possible to make a rough calculation of the distance from the assumed supernova to the solar system by using the measured surface density of iridium in the Gubbio boundary layer and two assumed quantities relating to the supernova. The first is the 1 solar mass usually assumed to be blown off in a type II supernova. The second is the fraction of that material which is iridium, and here one has nothing but untested theories as a guide. C. F. McKee gave us his best estimate of the distance (from the Italian data) to be about 0.1 light year (L.Y.). That is much closer than those who have favored a supernova theory of the extinctions have ever suggested, even though most everyone would agree that such a close explosion would have killed most of the

organisms living on the earth by the mechanism of greatly increased cosmic radiation. The reason no one had ever suggested such a close supernova is simply that its a priori probability is so low. The probability varies with the cube of the distance, and depending on the assumptions one makes, the value lies in the range of 10^{-6} times R^3, per 10^8 years, where R is the distance, measured in light years (43). So for R = 0.1 L.Y., the probability is about 10^{-9} that in the last 100 million years a supernova event occurred within this distance from the sun. That is an uncomfortably small probability. It is doubly so because any viable theory of the extinction mechanism should probably apply to earlier extinctions as well. Any mechanism with such a low a priori probability is obviously a one-time-only theory. Nevertheless because the theory could be subjected to a direct experimental test, it was treated as a real possibility until two other independent pieces of evidence effectively ruled it out.

Ordinary charged particle thermo-nuclear reactions in stars cannot produce elements with atomic numbers higher than those having the maximum binding energy per nucleon--iron and nickel. In order to make heavier elements such as iridium, uranium or plutonium, stars must produce neutrons, which build heavier isotopes by being captured (44). The most copious source of neutrons known is that unleashed in a supernova, when the stored gravitational energy of a star is suddenly released by the star's collapse, with the formation of enormous neutron fluxes. One heavy isotope in particular offered the possibility of testing the supernova hypothesis; this is ^{244}Pu, with a half-life of 80.5×10^6 years, which is close to the age of the extinction event.

The explosion of a supernova should send out an expanding shell of newly created heavy elements, with a predictable ratio of Ir atoms to ^{244}Pu atoms. Any ^{244}Pu incorporated in the earth at the time of the creation of the solar system, about 4.7 billion years ago, would have decayed by 58 half-lives, or by a factor of 10^{17}, which would make it quite undetectable in the Gubbio limestone by the most sensitive techniques available. As was explained above, the absolute amount of Ir expected to be expelled in a supernova could not be tested experimentally, but came instead from theoretical models. But the expected ratio of ^{224}Pu to Ir has a relatively solid experimental basis (45). The amount of ^{244}Pu present at the formation of the solar system can be inferred from the existence of an anomaly in the meteoritic abundance of heavy Xe isotopes that has been found to be due to the fission of ^{244}Pu. From the experimentally determined ratio of ^{244}Pu to ^{238}U in the meteorites, plus the known Ir to U ratio in solar system material, one calculates that each Ir atom should have been accompanied, in the supernova explosion, by about 10^{-3} ^{244}Pu atoms. This is the origin of the ratio used in one of two tests of the supernova theory of the extinction. It is of course independent of the calculated distance to the assumed supernova.

^{244}Pu emits alpha particles, as does its more familiar isotope, ^{239}Pu, which has a half-life of 24,000 years. The specific radioactivity of ^{244}Pu is therefore smaller than that of ^{239}Pu by the ratio of their half-lives, or a factor of about 3000. We considered and rejected the possibility of detecting the alpha radioactivity of

the ^{244}Pu. If the Ir in the Cretaceous-Tertiary boundary layer had come from a supernova, the expected alpha particle counting rate from a highly concentrated sample of the ^{244}Pu would be in the range of several counts per year. But fortunately, ^{244}Pu is easily detectable both by mass spectrometry and by NAA. In the latter method, which we utilized, ^{244}Pu is converted to ^{245}Pu, which has a half-life of 10 hours, and emits many characteristic gamma rays and X-rays.

The plutonium was chemically separated from 25 and 50 g batches of boundary material and a 50 g batch of bedding clay from below the T-C boundary, and nearly "mass-free" samples were obtained--no carriers were added. Chemical separations were also performed on the plutonium fraction after the neutron irradiation. No significant gamma radiation was observed, other than that associated with the Pu isotopes. In order to measure our chemical yields, some Gubbio acid-soluble and acid-insoluble residues were "salted" with small amounts of ^{238}Pu. This Pu isotope is easily detectable through its alpha decay as its half-life is only 87.7 years. In addition, one of the samples was salted with ^{244}Pu. Figure 11 shows the gamma ray spectrum of a sample salted with about 20 picograms of ^{244}Pu, and indicates both the sensitivity of NAA to the detection of ^{244}Pu, and also the freedom of the purified sample from other elements that might interfere with the detection of ^{244}Pu. The dotted line is not drawn to pass through the measured points, but is computed from the measured height of the 327 KeV gamma ray line, and the known energies and relative abundances of the other most intense gamma ray and X-ray

lines from ^{245}Pu, and its 2-hour ^{245}Am daughter. A comparison of Fig. 11 with that of the sample not salted with ^{244}Pu (Fig. 12) showed that the Gubbio samples contain less than 0.1 of the amount that would be expected to accompany the measured iridium, if a supernova were responsible for the latter. The ocean, however, can produce chemical and physical changes in depositing materials as well as diagenetic alterations in the deposited sediments. The absence of measurable ^{244}Pu in our samples suggests that the anomalous iridium did not come from a supernova 65 m.y. ago, but it is not a conclusive argument. The NAA calibration for ^{244}Pu was confirmed by a measurement of the Pu isotopic ratio in the sample salted with ^{244}Pu. The 5-foot-radius single-direction focusing mass spectrometer used in this measurement, which employs thermal ionization at the source and single ion counting detection, will be used to search for much lower ^{244}Pu levels than is practical by NAA.

The second method that was used to test whether a supernova was responsible for the iridium anomaly stemmed from a suggestion by J. R. Arnold; it involved a measurement of the isotopic ratio of iridium in the boundary material. Iridium has two isotopes, 191 and 193, which would be expected to occur in about the same abundances, 38.5 percent and 61.8 percent, respectively, in all solar system material, because of mixing in the proto-solar gas cloud. However, different supernovae should produce iridium with different isotopic ratios, because of differences in the rapid ("r-process") to slow ("s-process") production contributions occasioned by variations in neutron fluxes,

reaction times, etc., from one supernova to the next. According to this generally accepted picture, solar system iridium is a mixture of that element produced by all the supernovas that ejected material into the gaseous nebula that eventually condensed to form the sun and its planets. Any new supernova would be expected to produce Ir with an isotopic ratio different from that of solar system material, with possible differences of a factor of 2, according to C. F. McKee.

We therefore compared the isotopic ratio of Ir from the C-T boundary clay with that of ordinary Ir, by NAA. In our earlier analytical work, we used only the 74-day ^{192}Ir, made from ^{191}Ir by neutron capture. But in this new work, we also had to measure the 18-hour ^{194}Ir made from the heavier Ir isotope, and extensive chemical separations before and after the neutron irradiations were necessary. The chemical purity requirements for this work were even more severe than those necessary in the plutonium search, and Fig. 13 shows a typical gamma-ray spectrum of the kind used in the isotopic ratio measurement. Also shown is one which demonstrates the need for chemical purification of the iridium fraction.

It is hard to think of a measurement that is simpler in principle, and apparently less sensitive to systematic error. One might expect to take the iridium from the boundary layer and an ordinary sample of iridium, irradiate them simultaneously in a reactor at any unknown flux, and measure the relative peak heights of the gamma rays of the two Ir isotopes. The two samples could be placed at any unmeasured (and even different) distances from the gamma ray detector. Then the

ratio of the two peak heights should be the same in the two samples, if the isotopic ratio was the same in each. If, by chance, the isotopic ratio was different in the two samples, the "ratio of the ratios" would give the desired answer. But this picture was found to be too simplistic. Two important systematic errors were tracked down and eliminated. The first involved a greater loss of counts by ^{192}Ir, when the solid angle of detection was increased; ^{192}Ir has more gamma rays emitted in coincidence than ^{194}Ir. The second involved small changes in the neutron flux spectrum between two points spaced a few mm apart in the reactor.

The final result was that the isotopic ratio of the boundary Ir differed by only (0.03 ± 0.65) percent from that of the standard. From this, we think it is safe to conclude that the boundary layer Ir and the standard did not differ significantly by more than 1.5 percent and therefore the Ir was very likely of solar system, rather than supernova, origin. So from that point onward, the iridium anomaly was assumed to be of extraterrestrial, but solar system, origin (46).

THE ASTEROID IMPACT HYPOTHESIS

After obtaining negative results in our test of the supernova hypothesis, we were left with the question of what extraterrestrial source within the solar system could supply the observed iridium and also cause the extinctions. We considered and rejected a substantial number of hypotheses. Several sources of hydrogen and iridium were examined, because hydrogen could combine with the atmospheric oxygen, to reduce its partial pressure, and therefore kill animals by anoxia.

Before the supernova was eliminated from consideration, passage through a gaseous nebula appeared to be able to supply the right amount of hydrogen, at the proper intervals, but the hydrogen accumulated too slowly. The sun appeared as an attractive source for some time, but it was eventually eliminated by several energetic considerations. Jupiter was more favorable energetically, but no reasonable scenario using Jupiter could be constructed; however, consideration of Jupiter had a useful catalytic effect in making us look more closely at asteroids.

Finally, one hypothesis, which we believe has not previously been suggested, did seem to account quite well for most of the relevant observations. In brief, this hypothesis suggests that an Apollo object--an asteroid in an earth-crossing orbit--struck the earth 65 m.y. ago. From the Italian data, the object was about 8 km in diameter and produced an impact crater about 140 km in diameter. Much of the dust-sized material ejected from the crater reached the stratosphere, was spread around the globe, and effectively prevented sunlight from reaching the surface for a period of several years, until the dust settled to earth. Loss of sunlight suppressed photosynthesis, and as a result, most food chains collapsed and the extinctions resulted.

Several lines of evidence support this hypothesis, as discussed in the next few sections. We can calculate the size of the impacting object, and comparison with the distribution of known Apollo objects in orbit indicates that such bodies should strike the earth with about

the same frequency as that of the major extinctions. The volume of dust-sized material that would be ejected from the crater produced by such an impact is easily sufficient, if spread world wide, to darken the earth, and atmospheric mixing rates are fast enough to spread the dust over the whole globe. The general pattern of extinctions that would be produced by several years of darkness matches quite well the pattern of extinctions that occurred at the end of the Cretaceous.

APOLLO OBJECTS

One might expect the moon and the earth to have swept up long ago all objects capable of scarring their surfaces, and a simple calculation would support that conclusion. But the well-known 25,000 to 50,000 year old crater in Arizona indicates that there are still large objects capable of making impact craters on the earth. The explanation that has been given for this apparent discrepancy is that an equilibrium exists between the loss of earth-orbit-crossing objects by collisions with Mars, the Earth and the Moon, and the injection of new material (now called Apollo objects and meteoroids) into earthcrossing orbits. The injection mechanism has been studied by a statistical-mechanical investigation of the perturbation of orbits of out-gassed comets and ordinary asteroids, by gravitational interactions (47), and has also been simulated on computers, using a Monte Carlo technique (48). Both methods predict a steady state population of Apollo objects of about the correct density to have produced the moon's craters, and those on Mercury, Mars and the Earth. Most of the Earth's craters have been erased by erosion, but in the past few years, large numbers have been found on satellite and aerial photographs, particularly in Canada.

Additional convincing evidence in support of the nearly constant number of Apollo objects with time has been the discovery, on astronomical photographic plates, of about the predicted number of such objects. The first Apollo object (Apollo itself) was discovered in 1933; now 28 are known, with a present day discovery rate of about 4 per year. The latest article in the astronomical literature on such objects is by Chapman, Williams and Hartman (49), and an excellent summary article by G. W. Wetherill (50) appears in the March 1979 issue of the Scientific American.

Apollo objects are difficult to find and study. They are so far away as to appear pointlike, so their diameters must be inferred from their scattered light intensity and their assumed albedos; these are known to vary (for solar system debris) from 0.05 to 0.20. (Note added Dec. 12, 1979. Eugene Shoemaker now determines the photographic albedo by spectrophotometric means and thereby reduces this uncertainty). From statistical arguments involving the rate of discovery of new objects, the "rediscovery" of known objects, the observed lunar and terrestrial cratering, and the known inventory, Wetherill (50) estimated that there are about 700 objects equal to or larger than 1 km in diameter (compared to the 28 now known, with a largest known diameter of 8 km). Shoemaker, Helin and Gillett (49,50) estimate the number above 1 km in diameter to be 750 ± 300.

FREQUENCY OF IMPACTS

Careful counts of lunar impact craters (52) larger than a diameter, D, show that this number, N, per unit area, varies about as D^{-3}; in other words, $\log N = -3.0 \log D + C$. This is true for

craters with diameters between 1 meter and 100 km, where one "runs out of statistics." This relationship should break down when the craters have diameters less than about 3 meters, because at that size, the calculated probability that every circle with such a diameter has been hit by a meteorite large enough to make such a crater approaches unity. Extensive experimental and theoretical studies of crater formation (47) indicate that for a constant impact velocity, the diameter of a lunar crater should vary with the mass of the impinging object according to the relationship

$$\log D = 0.30 \log m + K.$$

If we assume a constant density and a constant velocity for the incoming object, and a spherical shape, with diameter, d, the slope of the log D vs log d plot is 3 times 0.30 = 0.90. (This is because mass is proportional to d^3.) The slope of the log N vs log d curve is then 0.90 x (-3.0) = - 2.7. The last sentence says in effect that the number of objects impacting the moon, per unit time and per unit area, with diameters greater than the value d, is given by the power law

$$N_d = kd^{-2.7},$$

where the constant k depends upon the units of area and time, and is therefore unimportant. The important constant is the "index", which in this case is -2.7. So far, all of this information concerning moon

craters has come from an examination of the moon's surface, plus a theoretical extrapolation from terrestrial cratering experience.

If one plots the known number of Apollo objects against their inferred diameter, on log paper, the maximum slope (index) is about -1.6. Sagan (54) says, "The sizes of objects in the asteroid belt, and indeed anywhere else that collisions determine sizes, are understood by comminution physics" (the study of fragmentation processes). "The number of objects of a given size range is proportional to the radius of the object to some negative power, usually in the range from 2 to 4." Since there must be a bias against finding smaller Apollo objects at a given range -- they reflect less light -- the observed maximum slope of -1.6 should probably be changed to about -2.7, to lie within Sagan's theoretical range, from -2 to -4, as well as to agree with the index observed from moon crater data. (A steeper true slope, compared to the observed slope, would arise naturally from the selection bias in any search for Apollo objects; one misses a larger fraction of the passing Apollo objects, as they become smaller and fainter.)

In our earliest calculations of the mean time between collisions of objects larger in diameter than 5 or 10 km with the earth, we inserted estimated numbers of Apollo objects into Öpik's equation (47,54) relating the orbit parameters of the object (relative to the earth's orbit), and the lifetime of that object against collisions with the earth. None of the known Apollo objects, on their presently known orbits, could impact either the earth or the moon. But as Wetherill

explains, (50) the orbital planes of the objects precess under the gravitational influence of Jupiter, so that once every 5000 years or so, a typical Apollo object will be in an orbit that exactly intersects the earth's orbit, so there is then an easily calculable probability of a collision -- larger than that predicted by the physical cross sectional area of the earth, because of "gravitational focusing."

The most straightforward way to understand the mean time between collisions of an Apollo object of, say, 10 km diameter with the earth is to quote from the caption to Wetherill's map (50) showing the ancient craters in Canada: "Richard Grieve of the Canadian Department of Energy, Mines and Resources has made a count of the well-established ancient craters in North America and Europe; on the basis of his count one can estimate that in the past 600 million years 1,500 Apollo objects one kilometer in diameter or larger have struck the earth, about 70 percent of which landed in the oceans." By using an index of -2.7, one calculates that 3 Apollo objects 10 km or more should have struck the earth in that time, or one every 200 M.Y. The importance of the index can be seen if the time is recalculated using an index of -2.5; collisions then happen on the average every 130 M.Y.

The mean time between collisions can also be calculated from the inventory of Apollo objects and their mean orbit parameters, using Öpik's formula. Rather than doing this from first principles, using input data from various sources, it is probably more valid to scale from the value obtained by Wetherill when he uses his best judgement

in carrying out that same exercise. (50) He finds the probability that any typical Apollo object will collide with the earth, per year, is 5×10^{-9}. Its inverse, 200 M.Y., is the mean time to a collision with the earth, for that object. If there are 750 Apollo objects larger than 1 km, and the index is -2.7, there should be 1.5 objects larger than 10 km diameter. The mean time between collisions for such objects is then 200 M.Y./1.5 = 130 M.Y. Again, the use of a different index changes the time as it did in the last paragraph.

The two time intervals calculated above, one from purely geological data (Grieve's), and the other from purely astronomical data (assumed number of Apollo objects plus Öpik's collision rate formula), are both in the 100 M.Y. range expected from paleontological evidence. The only adjustable parameter is the index, which is not known for Apollo objects. Three reasons have been given for choosing an index near -2.7, and an example was given to show the sensitivity of the time scale to the change in index.

(Note added Dec. 12, 1979. Just before the first set of preliminary copies of this report were sent out for review we learned of an article by William K. Hartmann [Sci. Am. 236, Jan. 84-99 (1977)] that contained much more data on moon craters than was available in our reference (52). Hartmann gives reasons for believing that the correct index to use in relating large crater density to crater diameter is -2, which corresponds to an index for crater density vs. Apollo object diameter of -1.8. Subsequent conversations with Eugene Shoemaker confirmed that value. Shoemaker also believes that the

index increases for Apollo objects above about 10 km, and that the largest Apollo object is approximately 10 to 20 km in diameter. We must therefore recalculate our predicted times between collisions, based on the smaller index of -1.8. From Grieve's data, the average time between impacts of 10 km diameter objects is reduced from 200 MY to 25 MY. Impacts with the 60 percent of the earth's surface that is covered by more than 2000 meters of water would probably not lead to the scenario described in this paper, so the time between "effective collisions" is increased to 42 MY. And any increase in the index at large diameters would further increase the time between collisions, to make it agree more closely with the canonical 100 MY required by the proposed hypothesis. Eugene Shoemaker reports that on hearing of the qualitative aspects of our hypothesis, he independently calculated that the required asteroid should have a diameter of approximately 10 km).

KRAKATOA

The largest well-studied terrestrial explosion in historical times is that of the island volcano, Krakatoa, in the Sunda Strait, between Java and Sumatra (55). Since this event provides the best available data on injection of explosion products into the stratosphere, we give here a brief summary of relevant information.

On August 26 and 27, 1883, Krakatoa underwent volcanic eruptions that shot an estimated 18 cubic kilometers of material into the atmosphere, of which about 4 cubic kilometers ended up in the stratosphere, where it stayed for 2 to 2-1/2 years. Dust from the explosion

circled the globe so quickly that many scientific observers wouldn't believe that the obscured sunsets seen world-wide, a few days later, were related to the explosion; they proposed alternate theories in which the earth almost simultaneously passed through a meteor shower on the far side of the globe. But the Krakatoa Committee of the Royal Society collected hundreds of reports on the unique meteorological conditions, and proved that the upper air circulation was far more rapid than anyone had expected. (One can call this the discovery of the jet stream).

Many famous names appear in the list of almost 500 men who reported observations that are recorded in the committee's report (55); in addition to Cornu, Helmholtz, Langley, Lockyer, Pockels, and Tyndall, 90 ship captains reported on entries from their logs. From this mass of data, the committee was able to reconstruct the way the material fell out of the atmosphere as a function of distance -- from the thickness of pumice on ships' decks or covering city streets within hundreds of miles of the explosion. Some nearby ships reported that it was quite dark for days.

The world at large was fascinated by the extraordinarily dusty and highly colored sunsets that persisted for two and one-half years. The dust spread quickly to the west, and then, after circling the globe, spread relatively uniformly over all latitudes. (Recent measurements of the ^{14}C injected into the atmosphere by the nuclear bomb tests confirm the rapid -- 1 year -- mixing between hemispheres (56).) At noon, observers generally classed the sky as "clear blue," when no

water vapor clouds were in evidence. Photo cells were unknown in this period, and accurate photometers were apparently not available, so no quantitative measurements seem to be available to help in evaluating the actual absorption of the dust as a function of path length.

If we take the estimated mass in the stratosphere (4 cubic kilometers) times the assumed low density of 2 gm per cm^3, and spread it uniformly over the globe, it amounts to 1.6×10^{-3} gm/cm^2. The layer was effectively 0.0003" thick, and was spread over a range of several miles in height. It is known that the layer did not absorb much of the incident radiation, on a "straight through" basis, but it did lead to the dusty sunsets when viewed through a much longer path, roughly parallel to the horizon.

But if the amount of very fine dust in the stratosphere were increased by a factor of 1600, it is most probable that the sunlight would be attenuated to a high degree, and the scenario outlined in our hypothesis could well ensue. Since the scaling laws involved in the extrapolation to explosive cratering in this energy range are apparently not known, it will not be useful to try to extract more information from this scenario than is warranted. One final observation may be useful: Contrary to what one might think, the fallout of the suspended material should not be exponential with time, but more nearly linear; each dust grain falls at its own speed, as determined by Stoke's law, unless it coalesces with a neighboring grain, in which case it falls faster. Since the time for the dusty sunsets to disappear after Krakatoa is frequently given as 2 to 2.5 years, we have

arbitrarily assumed that the boundary layer was deposited in about 3 to 5 years. Optical observation of the suspended particle sizes (by measuring diameters of lunar diffraction halos) together with Stoke's law, gave a reasonable account of the way the material fell out of the stratosphere in the mid 1880's. It is therefore quite possible that 65 million years ago, day could have been turned into night for a period of perhaps three to five years, after which time, the atmosphere would return relatively quickly to its normal transparent state.

What happened during the Krakatoa explosions can be expected to happen to a much greater extent during the impact of a large asteroid. Wetherill (50) has the following to say concerning the impact of a 1 km diameter asteroid with the earth (our canonical 10 km diameter object has 10^3 times the energy -- 100 million megatons of TNT -- and should make a crater with about 8 times the diameter he mentions -- 175 km);

> "When these rare but statistically inevitable collisions take place, the consequences are dramatic. In the reference frame of the earth the kinetic energy of a one-kilometer Apollo object of typical density (3.5 grams per cubic centimeter) and moving in a typical orbit is about 4×10^{27} ergs. This is 100,000 times the energy released by the detonation of a one-megaton nuclear warhead. All this energy must be dissipated in one way or another on impact with the earth. Only a small fraction of the total energy is required to vaporize the impacting body, so that most of the energy will be expended in pulverizing the material of the earth's surface and ejecting it outward at high velocity. Hence, the impact will excavate a crater similar to the craters that dominate the landscapes of the moon, Mars and Mercury.

> There are no direct data on the dimensions of craters produced by explosions of 10^5 megatons. One can, however, extrapolate empirical scaling laws based on much smaller explosions and carry out computer simulations of explosions of asteroidal magnitude. Such calculations indicate that an impact releasing 4×10^{27} ergs of energy will make a crater about 22 kilometers in diameter."

Although these paragraphs stress the inevitability of an event of the kind considered in this paper, the scenario stops as the earthly material (plus the Apollo object) is ejected "outward at high velocity". It seems plausible to imagine that in the case of a 10^3 times more energetic explosion, a mushroom cloud would form, and sweep the material into the stratosphere quite efficiently.

SIZE OF THE IMPACTING OBJECT

If we are correct in our hypothesis that the Cretaceous-Tertiary extinctions were due to the impact of an Apollo object, there are four independent ways to calculate the size of the object. All four ways, using the Italian data, give a diameter close to 7 km.

(1) The postulated size of the incoming asteroid was first computed from the iridium measurements in the Italian sections, the tabulated elemental abundances in carbonaceous chondrites, which are considered to be typical solar system material, and the fraction of erupted material estimated to end up in the stratosphere. If we neglect the latter fraction for the moment, the asteroid mass is given by

$$M = sA/f$$

where s is the surface density of Ir (measured at Gubbio to be 1×10^{-8} gm/cm^2), and f is the Ir mass fractional abundance in carbonaceous chondritic meteorites (0.40×10^{-6}). This preliminary value of the asteroid mass, 1.3×10^{17} gm, is then divided by the estimated fraction staying in the stratosphere, 0.22, to give $M = 5.8 \times 10^{17}$ gm. (We use the "Krakatoa fraction", 0.22, simply because it is the only relevant number we know, and we don't know how it scales.) At a density of 3.5, the diameter of the Apollo object would be 6.8 km.

(2) The second estimate comes from data on Apollo objects, and from the observed craters they have made on the earth's surface. In a sense, the second estimate comes from two quite different data bases -- one from geology and the other from astronomy. We will calculate the asteroid diameter from both data bases but they will not really be independent, since the two data bases are known to be consistent with each other. As we showed in an earlier section, the most believable calculation of the mean time between collisions of the earth and Apollo objects equal to or larger than 8 km in diameter is about 100 million years (MY). The smaller the diameter, the more frequent are the collisions, so our desire to fit not only the Cretaceous-Tertiary extinction, but earlier ones as well, sets the mean time between extinctions to be about 100 MY, and therefore sets the diameter at about 8 km.

(3) The third method of estimating the size of the asteroid comes from the possibility that the 1 cm boundary layer at Gubbio and Copenhagen is composed of material that fell out of the stratosphere,

and is not related to the clay that is mixed in with the Cretaceous limestone below it, or with the Tertiary limestone above it. This is quite a surprising prediction of the hypothesis, since the most obvious explanation for the origin of the clay is that it had the same source as the clay impurity in the rest of the Cretaceous and Tertiary limestone, and that it is nearly free of limestone because the extinction temporarily destroyed the calcite-producing plankton for about 5000 years. But as discussed above, the material in the boundary layer is of a quite different character from the clay above and below it; the latter two clays are very similar. To estimate the diameter of the asteroid, one can use the surface density of the boundary layer (about 2.5 gm/cm^2), together with an estimate of the fraction of that material which is of asteroidal origin. The asteroid diameter is calculated to be 7 km. The numbers used in this calculation are the following: density of the asteroid = 3.5; mass of crustal material thrown up per unit mass of asteroid = 100 (from cratering experiments plus scaling laws); fraction of excavated material delivered to the stratosphere = 0.22 (from the Krakatoa measurements). If one uses different numbers than those just listed the diameter obviously changes only by the cube root of the change in input value.

The first and the third methods are independent, even though they both depend on measurements made on the boundary material. This can best be appreciated by noting that if the Ir abundance was about the same in the earth's crust as it is in meteorites, the "iridium anomaly" seen in Fig. 3 would not exist. Therefore, method 1 would

not exist either. The fact that method 3 could still be used is the indicator of the relative independence of the two methods.

(4) The fourth method is not yet able to set as close limits on the mass of the incoming asteroid, but it leads to consistent results. This method derives from the need to make the sky much more opaque than it was in the years following the Krakatoa explosion. If it is assumed (for illustrative purposes and because it is probably not far from the correct value) that the Krakatoa dust cloud absorbed 3 percent of the vertically incident sunlight, then an explosion involving 33 times as much material would reduce the light intensity to 1/e. An explosion of the magnitude calculated in the four earlier methods -- about 1600 times that of Krakatoa -- would then reduce the sunlight to exp (-48) = 10^{-21}. This is of course much more light attentuation than is needed to stop photosynthesis. But if the mass of the asteroid were dropped by a factor of 3 (diameter dropped by a factor of 1.44, to 5 km), the sunlight would be attenuated to exp (-16) $=10^{-7}$ which is about one-tenth the light at full moon. This low illuminance would subject plants to a severe stress, but still permit most animals to locate food supplies. We can conclude this discussion of the "fourth method" of computing the size of the asteroid by saying that our input data concerning the vertical attenuation of sunlight after Krakatoa are not well enough known to give much weight to the calculation. But a reasonable guess (from the historically documented "dusty sunsets") tends to set the diameter in the same range as the 7 to 8 km found in the other three methods. The

lower limit is obvious; if there is not enough dust in the atmosphere to cut out the light, the plants will not stop growing. If on the other hand, the attenuation associated with an Apollo object of a certain assumed diameter is much greater than that required to do the job, there should have been many similar extinctions in the past 100 MY. These would have made use of the many smaller (but still adequate), asteroids that exist for every large one. The small number of such extinctions therefore sets an upper limit on the asteroid size. This upper limit argument is based on our qualitative knowledge (from Apollo objects and craters) that the number decreases rapidly as the size increases. In effect, it says that the upper limit is not too far from the lower limit. So in the absence of good measurements of the solar constant in the 1880's, it can only be said that the fourth method leads to asteroid sizes that are consistent with the other three.

Until we understand the reasons for the factor of 7 difference in Ir content, of the boundary clay, between Denmark and Italy, we will be faced with a different value for the asteroid diameter, based on the first method. The "Danish diameter" is then 6.8 km x $7^{1/3}$ = 13 km. The second and third estimates are unchanged; the second does not involve measurements made on the boundary layer, and the third uses the thickness of the clay, which is only slightly greater in Denmark than in Italy The fourth method is based on such an uncertain input attenuation value, from Krakotoa, that it is not worth recalculating.

It is perhaps best to conclude this section by saying that all known data are consistent with an impacting asteroid with a diameter about equal to 10 ± 3 km.

BIOLOGICAL EFFECTS

A temporary absence of sunlight would effectively shut off photosynthesis and thus attack food chains at their origins. In a general way the effects to be expected from such an event are what one sees in the paleontological record of the extinction.

The food chain in the open ocean is based on microscopic floating plants, such as the coccolith-producing algae, which show a nearly complete extinction. The animals at successively higher levels in this food chain were also very strongly affected, with nearly total extinction of the foraminifera and complete disappearance of the belemnites, ammonites, and marine reptiles.

A second food chain is based on land plants. Among these plants, existing individuals would die, or at least stop producing new growth, during an interval of darkness, but after light returned they would regenerate from seeds, pollen and existing root systems. However, the large herbivorous and carnivorous animals that were directly or indirectly dependent on this vegetation would become extinct. Dale Russell states that "no terrestrial vertebrate heavier than about 25 kg is known to have survived the extinctions" (57). Many smaller terrestrial vertebrates did survive, including the ancestral mammals, and they may have been able to do this by feeding on nuts, seeds, insects, and decaying vegetation.

The situation among shallow-marine, bottom-dwelling invertebrates is less clear; some groups became extinct and others survived. A possible base for a temporary food chain in this environment is nutrients originating from decaying land plants and animals and brought by rivers to the shallow marine waters.

We will not go further into this matter here, but we refer the reader to the proceedings of the 1976 Ottawa meeting on the Cretaceous Tertiary extinctions. This volume reproduces an extensive discussion among the participants of what would happen if the sunlight were temporarily "turned off" (58). Those involved in the discussion seemed to agree that many aspects of the extinction pattern could be explained by this mechanism, although a number of puzzles remained. The discussion apparently moved on to other topics without establishing any way in which the light could have been turned off.

As one aspect of future testing of the asteroid-impact hypothesis it will be very important to consider in detail the pattern of extinction. This should be done in terms of the interpreted environmental habitat or food-chain membership of the organisms, rather than by taxonomic grouping.

We must note, finally, an aspect of the biological record which, on the face of it, is not in accord with the asteroid-impact hypothesis, or with any sudden, violent mechanism. Extinction of the foraminifera and nannoplankton occurs within reversed geomagnetic polarity zone Gubbio G- in the Gubbio section (13). R. F. Butler, E. H. Lindsay, and colleagues (59) have studied the non-marine

sequence of the San Juan Basin of New Mexico, and have found a polarity sequence that appears to correlate with the reversal sequence at Gubbio. In the San Juan Basin, the highest dinosaur fossils are found in the normal polarity zone (anomaly 29) that follows the Gubbio G- Zone. It thus appears that the dinosaur and foram-nannoplankton extinctions were not synchronous. (Extinctions occurring in the same polarity zone in distant sections would not establish either synchroneity or diachroneity.) Three comments on the San Juan Basin work have been published, including one by one of us (60), calling attention to the possibility of an unconformity at the boundary, in which case the correlation of the magnetic polarity zones could be in error, and the extinctions might still be synchronous. Lindsay, Butler and Johnson (61) argue strongly against a major hiatus, but admit "that the case is not completely closed."

Resolution of the question of whether or not the extinctions could have been synchronous will depend on further paleomagnetic studies. In the meantime we can state that the asteroid-impact hypothesis predicts that the apparently diachronous timing of the foramnannofossil and dinosaur extinctions will eventually be shown to be incorrect.

IMPLICATIONS

Among the many implications of the asteroid-impact hypothesis, if it is correct, two stand out prominently. First, if the Cretaceous-Tertiary extinctions were caused by an impact event, the same could be true of the earlier major extinctions as well. There have been five

such extinctions since the end of the Precambrian, 570 m.y. ago, which matches well with the probable interval of about 100 m.y. between collisions with objects this size, (~10 km). Discussions of these extinction events generally list the organisms affected according to taxonomic groupings; it would be more useful to have this information given in terms of interpreted ecological or food-chain groupings. It will also be important to carry out iridium analyses in complete stratigraphic sections across these boundaries.

Second, it may be possible to associate large impact craters on earth with particular extinctions. There are, at present, one suggested and five proven impact craters greater than 50 km in diameter. The largest is the partial ring structure marked by the accurate shoreline of southeastern Hudson Bay, which was once suggested as a possible impact crater (62). Its diameter would be about 400 km, and if it is an impact feature, it is older than the flat-lying Precambrian rocks on the Belcher Islands within the ring. We know of no further work on this site. The three largest proven impact sites are at Sudbury, Ontario, at Vredefort, in South Africa (63) and at Popigay, in Siberia (64). All three are about 100 km in diameter (Sudbury has subsequently been flattened to an ellipse). The first two are Precambrian in age, whereas Popigay has been dated stratigraphically as Late Cretaceous to Quaternary and by K/Ar as 28.8 M.Y. No major extinction occurs at this time, but details of the radiometric dating were not given so its reliability is not clear. The Puchezh-Katunki crater on the upper Volga River (65) is 80 km in

diameter and dated stratigraphically as Early Triassic to Middle Jurassic. No radiometric age was reported. The fifth large crater is at Manicouagan, Quebec (66). It is 65 km in diameter, and has given a Rb-Sr isochron age of 214 ± 5 m.y. (61) In the most recent radiometric time scale (68) the Triassic-Jurassic boundary is at 212 m.y., a major correction from the previous value of about 195-200 m.y. (69) The Triassic-Jurassic boundary is sometimes listed as a major extinction event, although the evidence is somewhat ambiguous. It may be a coincidence that the age of the Manicouagan crater is indistinguishable from that of the Triassic-Jurassic extinctions, but the possibility of a causal relationship should not be ignored. We note however, that 65 km is considerably smaller than the predicted crater of 175 km diameter; we would have expected this 65 km crater to be too small, and events of this magnitude too frequent, to account for major extinctions. We also note that the Triassic-Jurassic boundary falls in the stratigraphic interval to which the somewhat larger Puchezh-Katunki crater is confined. It seems unlikely that the Manicouagan event would produce an extinction if the Puchezh-Katunki event did not. Nevertheless, this topic is worth further investigation.

SUMMARY

The anomalously high concentrations of iridium that we have found in Italian and Danish limestone sections at exactly the level of the Cretaceous-Tertiary boundary strongly suggest that a sudden influx of extraterrestrial material occurred at the time of this great extinction. Several authors have proposed that a nearby supernova could

have produced the Cretaceous-Tertiary extinctions, but we report two observations and probability arguments that this was not the case.

In seeking to explain the paleontological record of the extinction and our observations on iridium, we have developed a new hypothesis which seems to account for most of the available information. The Earth should be struck by an Apollo-type asteroid with a diameter of 10 km about once every 100 M.Y., roughly the interval between major paleontological extinctions. Such an impact event would produce the iridium anomaly we see in the stratigraphic record. It would also produce an impact crater about 175 km in diameter. Pulverized rock debris from the crater would be injected as dust into the stratosphere and rapidly distributed over the whole globe. The quantity of dust in the stratosphere would be sufficient to cut out most of the light normally reaching the surface, and the resulting suppression of photosynthesis would be expected to produce very nearly the pattern of extinctions that is observed in the paleontological record. When the dust settled to earth in the next few years, it would deposit a layer of clay similar to the clay layer in which we have found the iridium anomaly in Italy and Denmark. In support of this suggestion, we note that the gross chemical composition of the clay layer is substantially different from that of the clay mixed with the limestone above and below the layer. We therefore suggest that impact of a 10-km asteroid may have caused the Cretaceous-Tertiary extinctions.

We wish to stress that this is only a hypothesis. We make no claim to have proven that an asteroid impact caused the extinctions at the end of the Cretaceous. Nevertheless, this hypothesis does explain

much of what is known about the extinctions including our new observations, and we know of no other hypothesis which does so. In the process of this work we made predictions based on this hypothesis (including the prediction of an iridium excess at other locations, and the chemical difference between the clay layer and the adjacent layers) which were subsequently verified by measurement. Fortunately, the asteroid impact hypothesis does make additional predictions, including the prediction that the Cretaceous-Tertiary extinctions were all simultaneous and the prediction that there will be iridium excesses at other extinctions. Additional confirmation of the hypothesis depends on the verification of these predictions.

ACKNOWLEDGMENTS

It will be obvious to anyone reading this paper that we have benefited enormously from conversations and correspondence with many friends and colleagues throughout the scientific community. We would particularly like to acknowledge the help we have received from James R. Arnold, Andrew Buffington, I. S. E. Carmichael, Garniss Curtis, Philippe Eberhard, Earl K. Hyde, Christopher McKee, Maynard C. Michel (who was responsible for the mass spectrometric measurements), John Neil, Bernard M. Oliver, Charles Orth, Barrie Pardoe, Andrew M. Sessler, and Eugene Shoemaker. One of us (W.A.) thanks the U. S. National Science Foundation for support, the other three authors thank the U. S. Department of Energy for support, and one of us (L.W.A.) thanks NASA for support. The XRF measurements of trace elements Fe and Ti by Robert D. Giauque and the XRF measurements of major elements

by Steve Flexser and Marian Sturz were most appreciated. We appreciate the assistance of Dan Jackson and Cecilia Nguyen in the sample preparation procedures. We are grateful to Tek Lim and the staff of the Berkeley Research Reactor for many neutron irradiations used in this work. We also appreciate the efforts of Gordon Pefley and the staff of the Livermore Pool Type Reactor for the irradiations used for the Ir isotopic ratio measurements.

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Table I

Abundance of acid-insoluble residues in the Danish section

Sample[a]	Abundance[b] of acid-insoluble residues (percent)
SK (+2.7m)	3.27
(+1.2m)	1.08
(+0.7m)	.836
Boundary	44.5
SK (-0.5m)	.654
(-2.2m)	.621
(-5.4m)	.774

(a) Numerical values are the distance in meters from the boundary layer.

(b) The boundary layer has a much higher proportion of clay than the pelagic limestones above and below.

Table II

Abundance of Trace Elements in Danish Boundary Layer
(expressed in ppm)

Abun. in wh. rk.[a] / Abund. of Residue		Abun. in[a] residue	Abun. in wh. rk. / abun. of residue		Abun. in residue
Enhanced Elements[b]			Depleted Elements		
V	391 ± 27	330 ± 31	Mn	102.0 ± 1.3	21.3 ± 0.5
Cr	371 ± 13	358 ± 9	Rb	27 ± 7	35 ± 4
Co	141.6 ± 1.8	57.2 ± 0.7	Y[c]	79 ± 6	6.3 ± 1.8
Ni	1137 ± 31	479 ± 14	Zr[c]	144 ± 11	125 ± 6
Cu[c]	167 ± 14	93 ± 6	Nb[c]	8 ± 4	6.1 ± 1.8
Zn[c]	1027 ± 49	378 ± 18	Cs	1.87 ± .19	1.51 ± .14
As	96 ± 8	68 ± 4	La	61.1 ± 1.6	6.8 ± .4
Se[d]	46.5 ± 0.6	12.1 ± .3	Ce	57.0 ± 1.2	9.7 ± .6
Mo	29.0 ± 2.5	20.3 ± 1.4	Nd	63.4 ± 2.7	5.4 ± .6
Ag[d]	2.6 ± .9	3.5 ± .7	Sm	11.93 ± .08	.781 ± .008
In[d]	.245 ± .022	.086 ± .019	Eu	2.76 ± .11	.121 ± .010
Sb	8.0 ± .4	6.7 ± .4	Tb	1.84 ± .04	.148 ± .014
Ba	1175 ± 16	747 ± 11	Dy	11.24 ± .12	.908 ± .033
Ir[d]	.0643 ± .0029	.0416 ± .0018	Yb	5.02 ± .09	.56 ± .05
Pb[c]	64 ± 14	28 ± 7	Lu	.553 ± .031	.083 ± .004
			Hf	4.34 ± .16	3.88 ± .07
			Ta	.508 ± .011	.500 ± .005
			Th	7.1 ± .4	1.28 ± .06
			U	8.63 ± .09	.918 ± .024

Table II (continued)

Other Elements[b]		
Sc	20.74 ± .16	14.30 ± .14
Ga[c]	30 ± 6	19.8 ± 3.0
Sr[c]	1465 ± 72	48.1 ± 2.4
Au	<.12	.027 ± .007

(a) The first column minus the second column is the amount of an element which dissolved in the acid or was lost in the firing. Abun. = abundance, wh. rk. = whole rock, abun. of residue = 44.5 percent.

(b) V, Ag and In are at least 20 percent and all other "enhanced elements" are at least a factor of 3 more abundant in the boundary residue than in the other residues. All "depleted elements" are at least 20 percent less abundant in the boundary residue than in the other residues. "Other elements" do not show a consistent pattern of boundary residue abundances relative to the others.

(c) Measured by hard X-ray fluorescence (39)

(d) Fluxmonitors were used in the NAA measurements of these elements. The indicated errors are applicable for comparing the two entries for a given element, but calibration uncertainies of possibly 10-20 percent must be considered when the values are used for other purposes.

Table III

Whole-rock Composition of The Gubbio and Danish Boundary Layers (percent)

Formula or name	Gubbio	Denmark
	Elements[a]	
SiO_2	27.7 ± .6	29.0 ± .6
Al_2O_3	12.19 ± .15	8.01 ± .17
FeO[c]	4.53 ± .05	4.35 ± .04
MgO	1.10 ± .07	3.07 ± .10
CaO	22.6 ± .4	23.1 ± .4
Na_2O	.1806 ± .0036	.0888 ± .0018
K_2O	2.46 ± .20	.38 ± .04
TiO_2	.521 ± .022	.324 ± .016
$S^=$	Not detected	~1.1
PO_4	Not detected	.92 ± .09
CO_2	17.7 ± .3	18.4 ± .3
Traces elements	~.2	~.3
Sum	89.2 ± .8	90.3 ± 1.0
Difference[e]	10.8 ± .8	9.7 ± 1.0

Minerals in the Danish Boundary Layer[b]

	Measured	Normalized
Calcite	~90 percent	41.5 percent (norm)
Quartz	5-7 percent	~3 percent
Iron Pyrites	~5 percent	~2 percent
Illite	2-3 percent	~1 percent

Table III (continued)

(a) Si, Ca, Mg, S, P and Gubbio Ti were measured by soft XRF (38). Some S may be lost in this sample preparation procedure. The Denmark Ti was measured by hard XRF (39). All other measurements were by NAA.

(b) Mineral analyses were made by Mark Ghiorso and I.S.E. Carmichael of the U. C. Berkeley Department of Geology and Geophysics utilizing X-ray diffraction.

(c) Total Fe expressed as FeO.

(d) CO_2 abundance was calculated from the Ca abundance by assuming all Ca was present as the carbonate.

(e) The difference is mainly water and organic material.

FIGURE CAPTIONS

Fig. 1 Stratigraphic section at Guggio (6,7). (a) Meter levels. (b) Systems. (c) Stages. (d) Magnetic polarity zones (black is normal, white is reversed polarity; leters give Gubbio polarity zonation, numbers are equivalent marine magnetic anomalies). (e) Lithology. (f) Samples used in first neutron activation analysis study (samples I, J, L are from equivalent positions in the Contessa section, 2 km to the northwest). (g) Formation names.

Fig. 2 Abundance variations of 28 elements in 12 samples from two Gubbio sections.

Fig. 3 Iridium abundances per unit weight of 2N HNO_3 acid-insoluble residues from Italian limestones near the T/C boundary. Error bars on abundances are the standard deviations in counting radioactivity. Errors bars on stratigraphic position indicate the total uncertainities in position with respect to the boundary center. The dashed line above the boundary is an "eyeball fit" exponential with a half-height of 4.6 cm. The dashed line below the boundary is a best fit exponential (two points) with a half-height of 0.43 cm. |——●——| is the mean and standard deviation of Ir abundances in 4 large samples of boundary clay from different locations.

Fig. 4 Typical gamma-ray spectrum used to determine Ir abundances in nitric acid insoluble residues without further chemistry. Note that the entire spectrum rests on a background of 118,000 counts. Detector volume = 128cc. Length of count = 980 minutes. Count began 39.8 days after the end of the irradiation.

Fig. 5 Stratigraphic section at Højerup Church, Stevns Klint, Denmark. (a) Lithology (C: Cerithium limestone; F: Fish clay). (b) Stages. (c) Samples analyzed in this study. (d) Meter levels.

Fig. 6 Major element abundances in acid-insoluble fractions from Danish rocks near the T/C boundary. The cross-hatched areas for the Cretaceous and Tertiary values each represent root-mean-square deviations for three samples. (Only two measurements of Mg and Si were included in the Cretaceous values). For the boundary layer sample the cross-hatched areas are the standard deviations associated with counting errors. Si and Mg measurements were made by XRF (38) and all others were made by NAA.

Fig. 7 Selected trace element abundances in ppm of Danish acid-insoluble residues. First bar is the mean value (the root-mean-square deviation (RMSD) is shown by the cross-hatched areas), for the given element in the three Cretaceous residues. Second bar is the abundance (counting error is shown by solid areas) for the given element in the boundary layer residue. Third bar is the mean value and RMSD for the given element in the three Tertiary residues. Measurements are by NAA except those of Zn which were measured by XRF (39). Significant amounts of Ni, Zn, Co, Ir and Th in all samples dissolved in the 2N HNO_3. Very little Cs, Ta or Hf in any of the samples dissolved in the acid.

Fig. 8 Some of the element abundances measured in acid-insoluble residues of Cretaceous, T/C boundary layer and Tertiary rocks near Gubbio, Italy. Abundances are in ppm except for the Ti value which is in percent. The data include all samples from that area measured within 19 meters of the boundary. There were four samples from each of the three layers, and the cross-hatched areas are the standard deviations. The abundance patterns for samples from ~27 km north of Gubbio are similar to those shown.

Fig. 9 Comparison of observed elemental abundance patterns in the Gubbio Section samples with average patterns expected for crustal material.

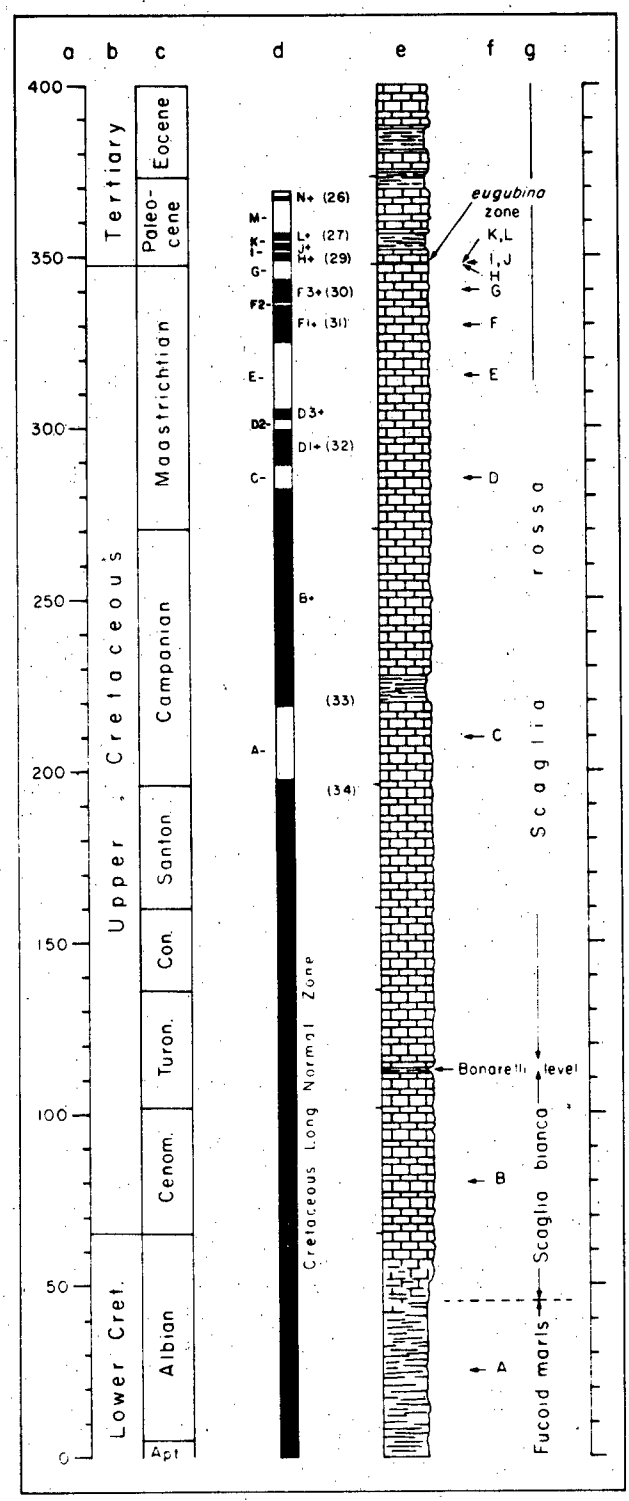
Fig. 10 Comparison of observed elemental abundance patterns in the Gubbio Section samples with the patterns expected for carbonaceous chondrites.

Fig. 11 Gamma-ray spectrum of Pu fraction from acid-insoluble residue of irradiated T/C boundary layer from Gubbio which had been salted with ^{244}Pu and ^{238}Pu containing relatively small amounts of ^{239}Pu, ^{240}Pu and ^{242}Pu.

Fig. 12 Gamma-ray spectrum of Pu fraction from acid-insoluble residue of irradiated boundary-layer clay from Gubbio which had been spiked with ^{238}Pu and relatively smaller amounts of ^{239}Pu, ^{240}Pu and ^{242}Pu.

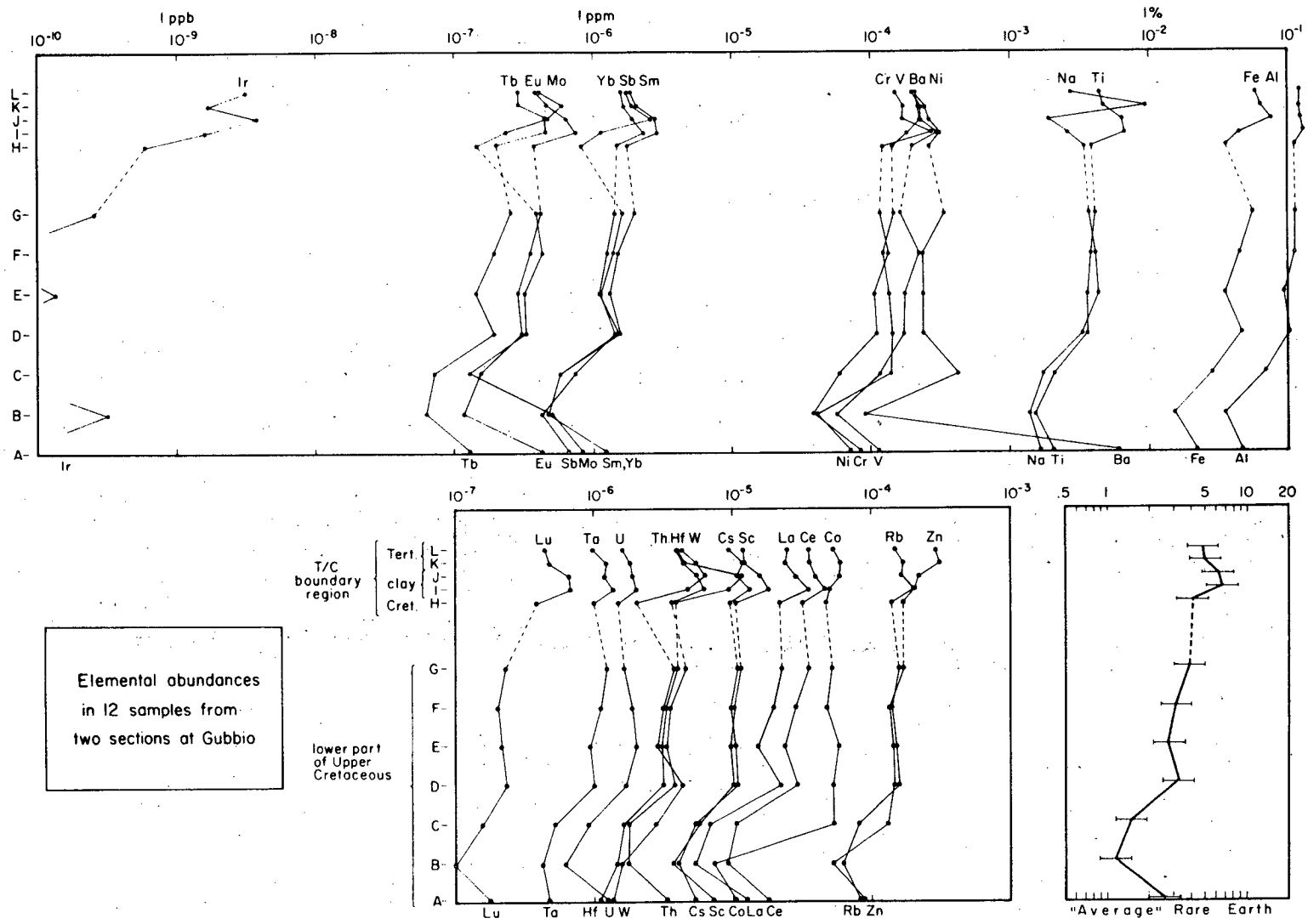
Fig. 13 Upper graph: Gamma-ray spectrum of irradiated acid-insoluble residue from T/C boundary at Gubbio without pre-irradiation or post-irradiation chemistry. Lower graph: Same as above with chemistry before and after irradiation. Counting periods, decay periods and chemical yields are different for the two spectra.

Plate 1. Photomicrographs of (a) the basal bed of the Tertiary, showing globigerina eugubina, and (b) the top bed of the Cretaceous, in which the largest foraminifer is globotruncana contusa. Both sections are from the Bottaccione section at Gubbio, they are shown at the same scale, and the bar in (a) is 1 mm long.



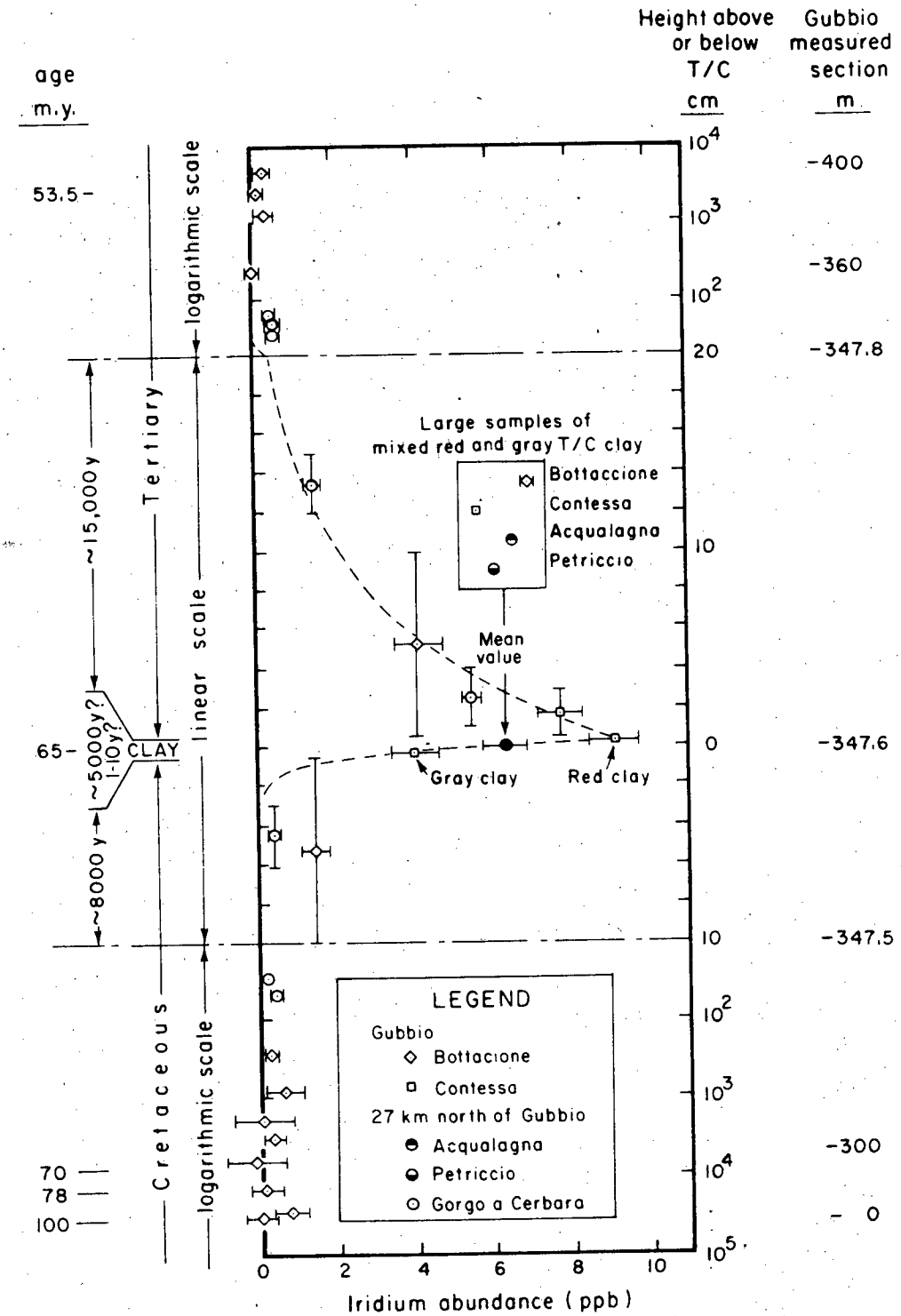
XBL 7911-12894

Fig. 1



XBL 795-9861

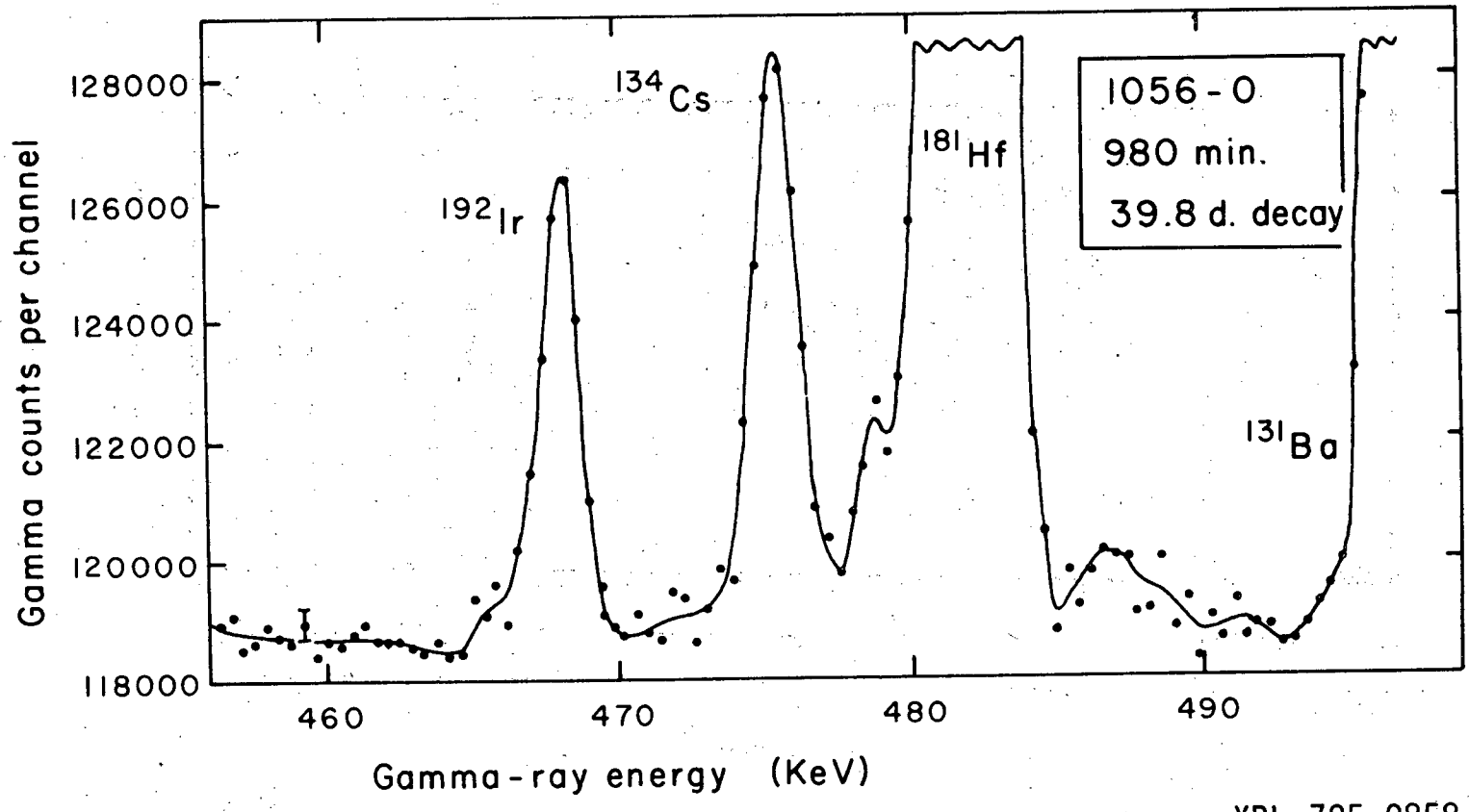
Fig. 2



XBL 7911-13250

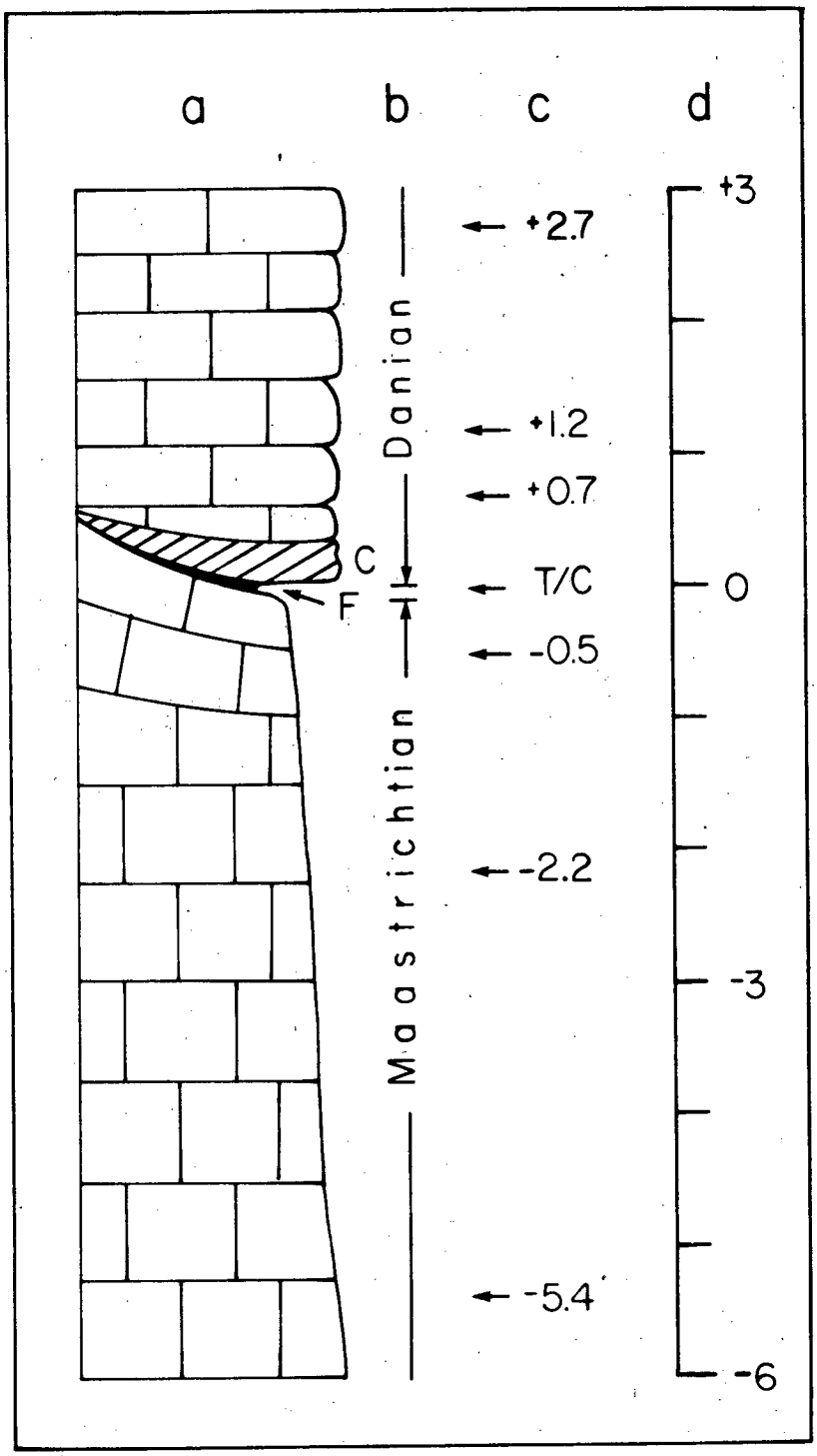
Fig. 3

^{192}Ir gamma-ray peak used in Neutron Activation Analysis of acid-insoluble residues



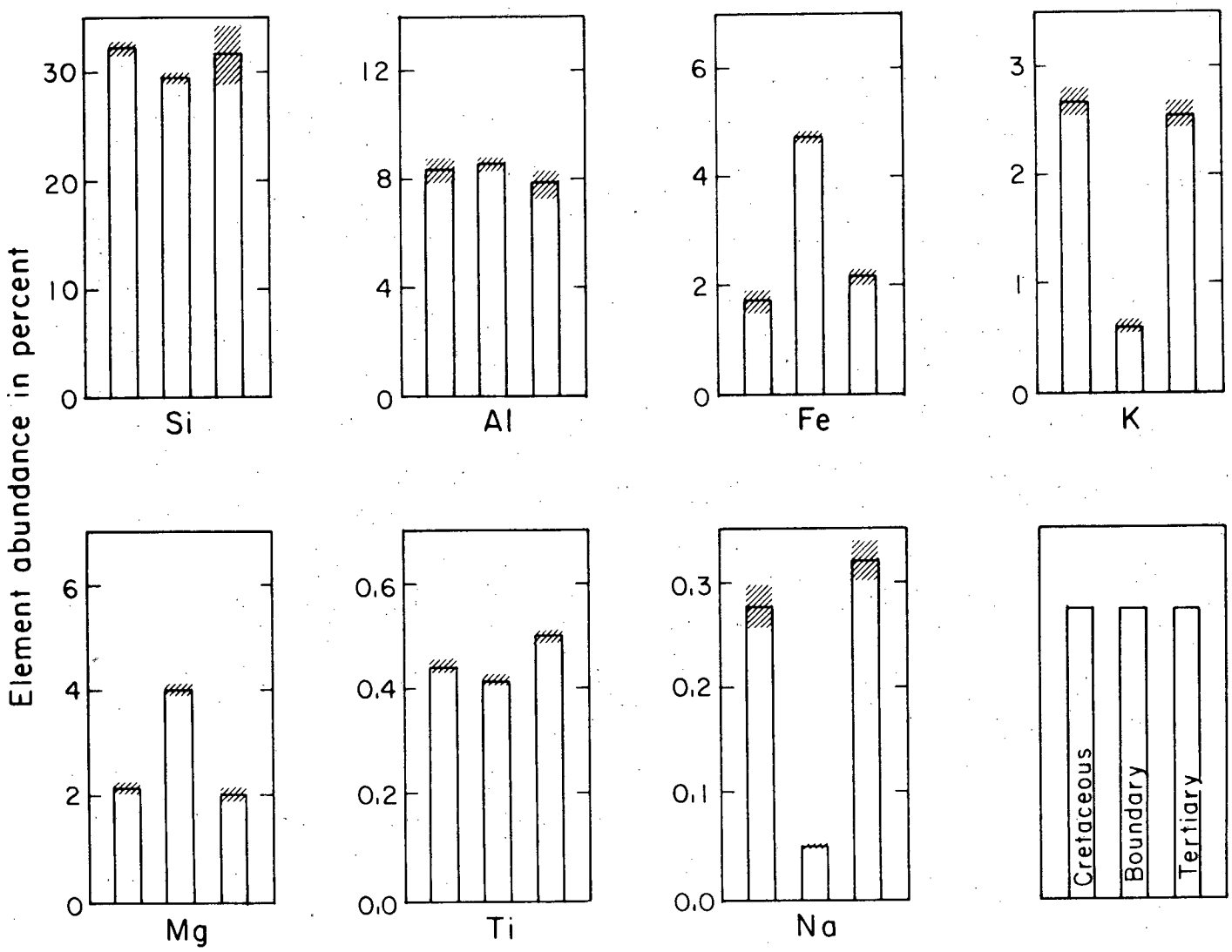
XBL 795-9858

Fig. 4



XBL 7911-12895

Fig. 5



XBL 7911-13248

Fig. 6

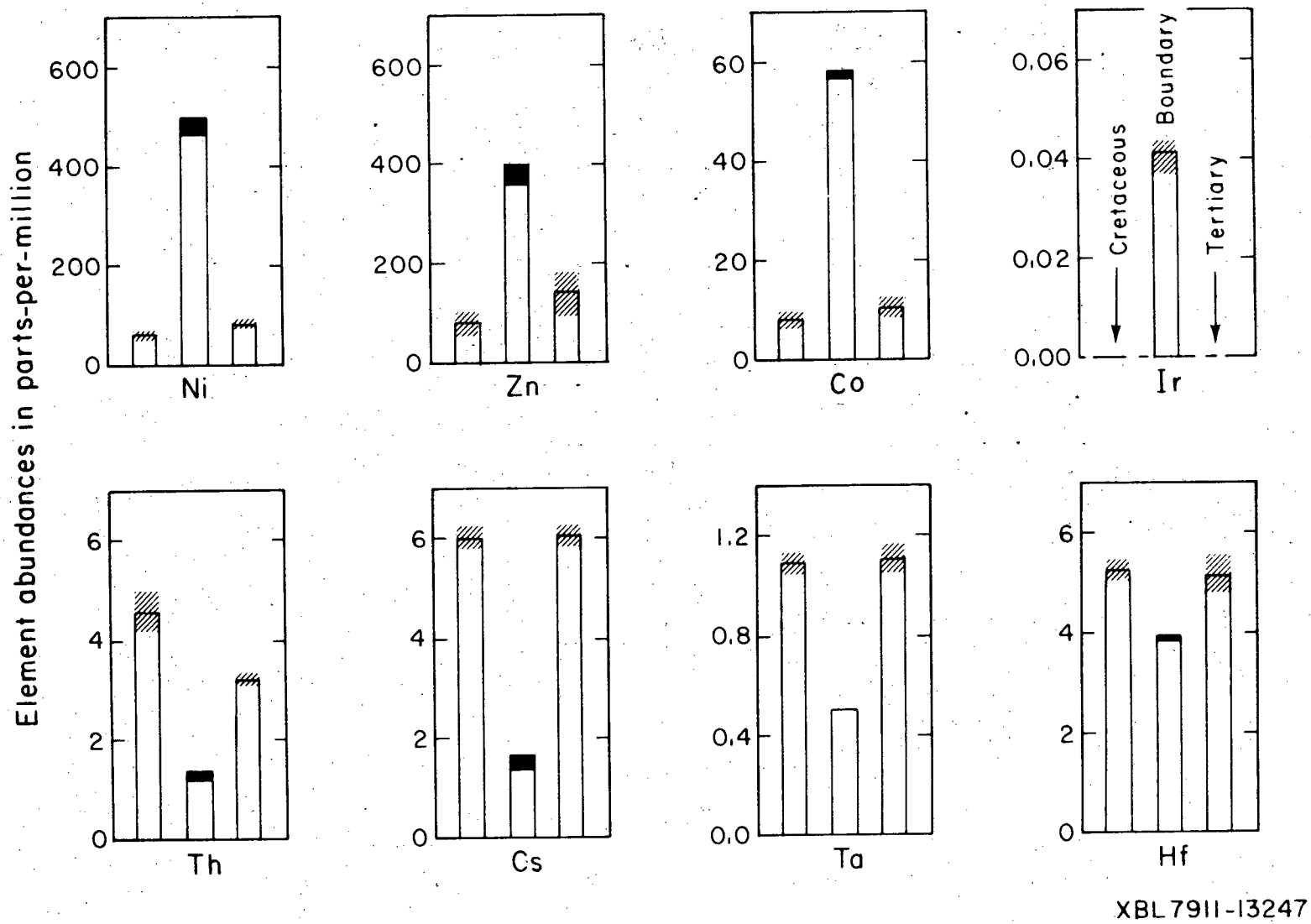

Element abundances in parts-per-million
Ni
Zn
Co
Ir
Th
Cs
Ta
Hf
Cretaceous
Boundary
Tertiary
XBL 7911-13247

Fig. 7

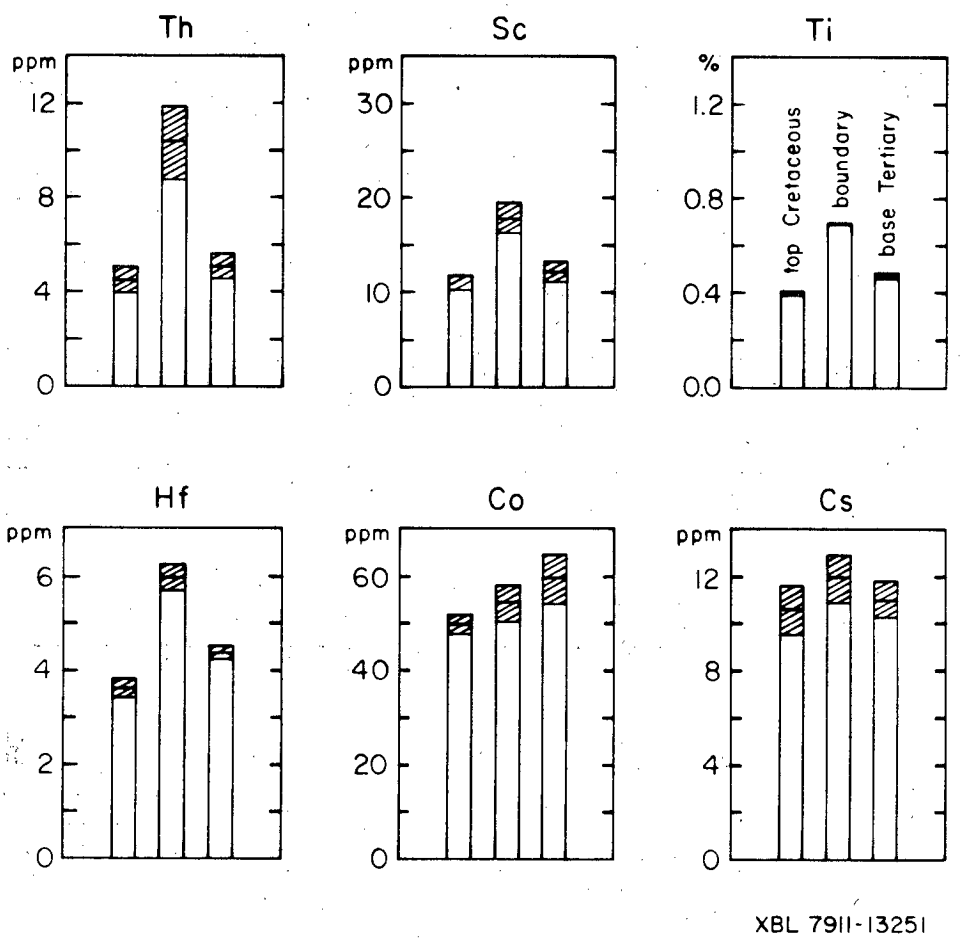
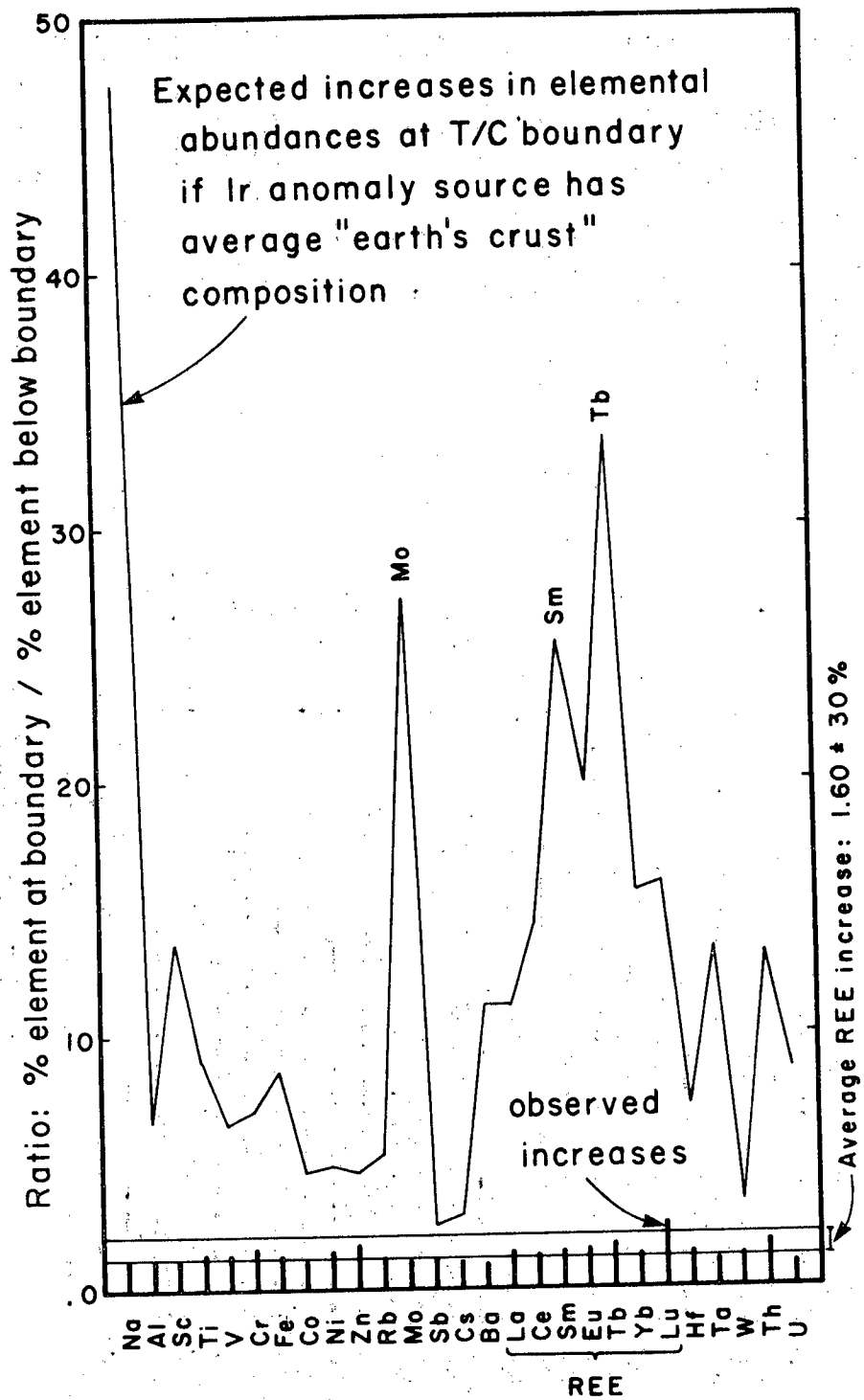
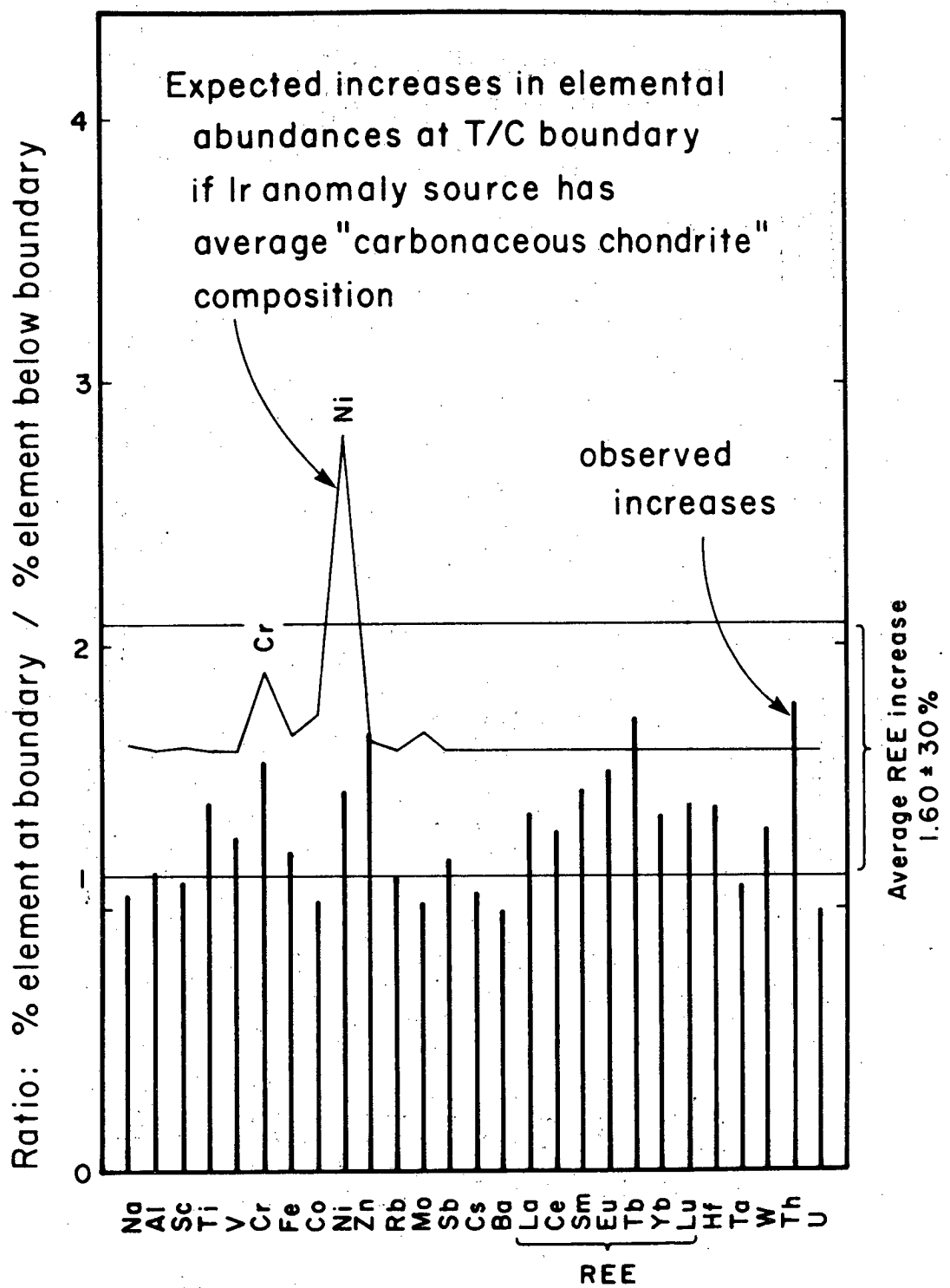

Th
ppm
12
8
4
0
Sc
ppm
30
20
10
0
Ti
%
1.2
0.8
0.4
0.0
top Cretaceous
boundary
base Tertiary
Hf
ppm
6
4
2
0
Co
ppm
60
40
20
0
Cs
ppm
12
8
4
0
XBL 7911-13251

Fig. 8



XBL 795-9855

Fig. 9



XBL 795-9854

Fig. 10

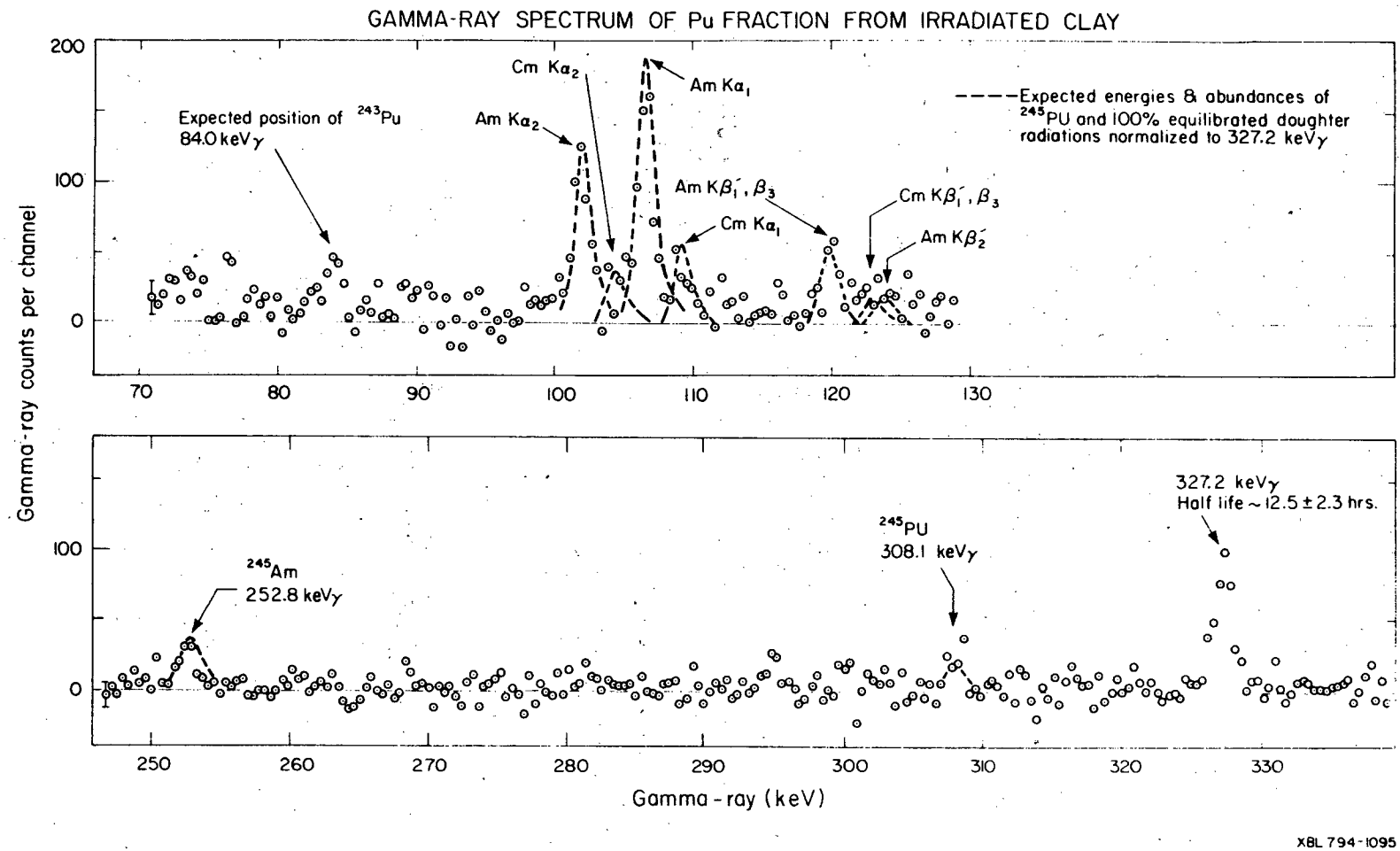
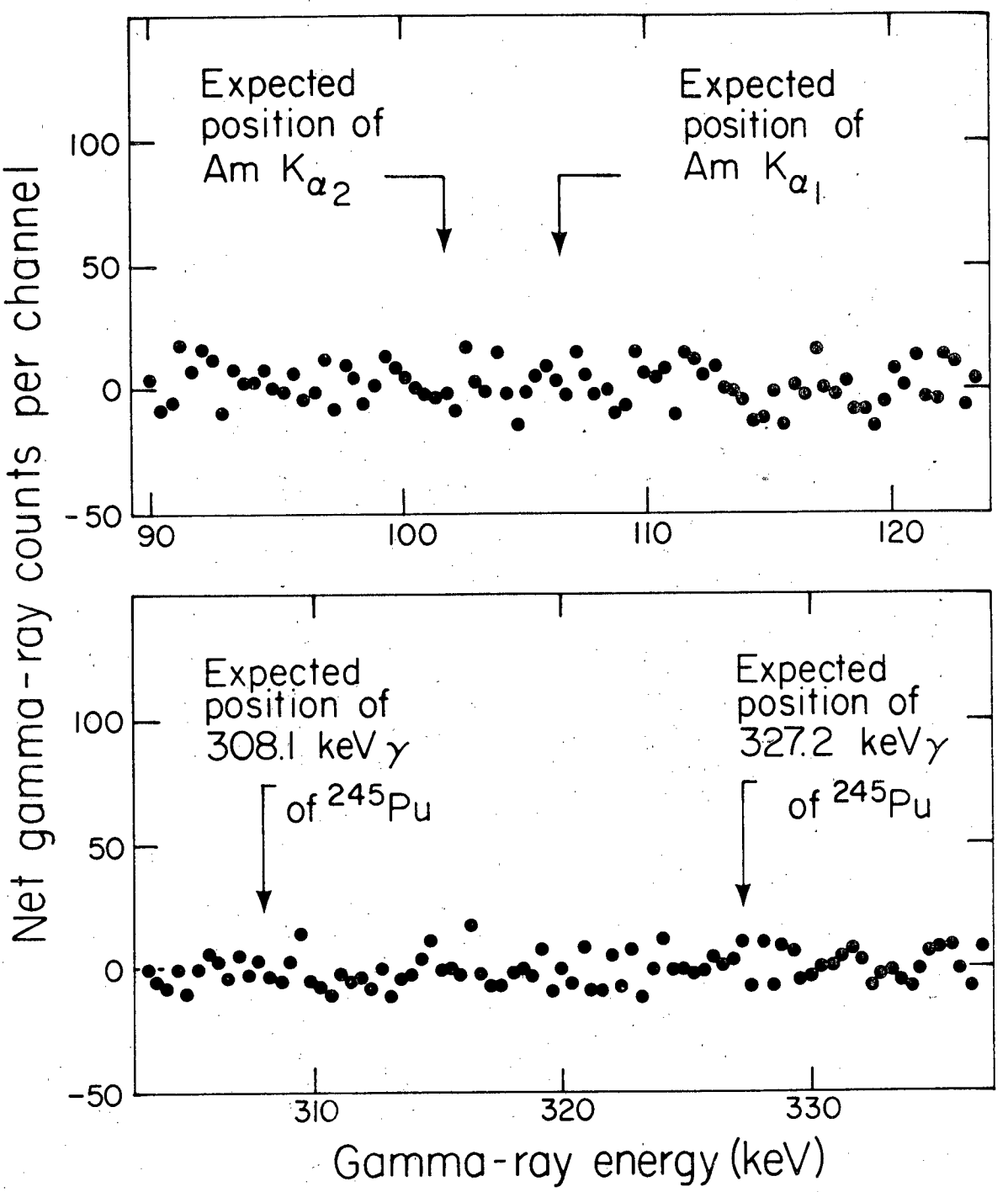

GAMMA-RAY SPECTRUM OF Pu FRACTION FROM IRRADIATED CLAY
Gamma-ray counts per channel
Expected position of ^{243}Pu 84.0 keVγ
Am Kα_2
Cm Kα_2
Am Kα_1
Am Kβ_1', β_3
Cm Kα_1
Cm Kβ_1', β_3
Am Kβ_2'
Expected energies & abundances of ^{245}PU and 100% equilibrated doughter radiations normalized to 327.2 keVγ
^{245}Am 252.8 keVγ
^{245}PU 308.1 keVγ
327.2 keVγ Half life ~ 12.5 ± 2.3 hrs.
Gamma-ray (keV)
XBL 794-1095

Fig. 11



XBL 798-2651

Fig. 12

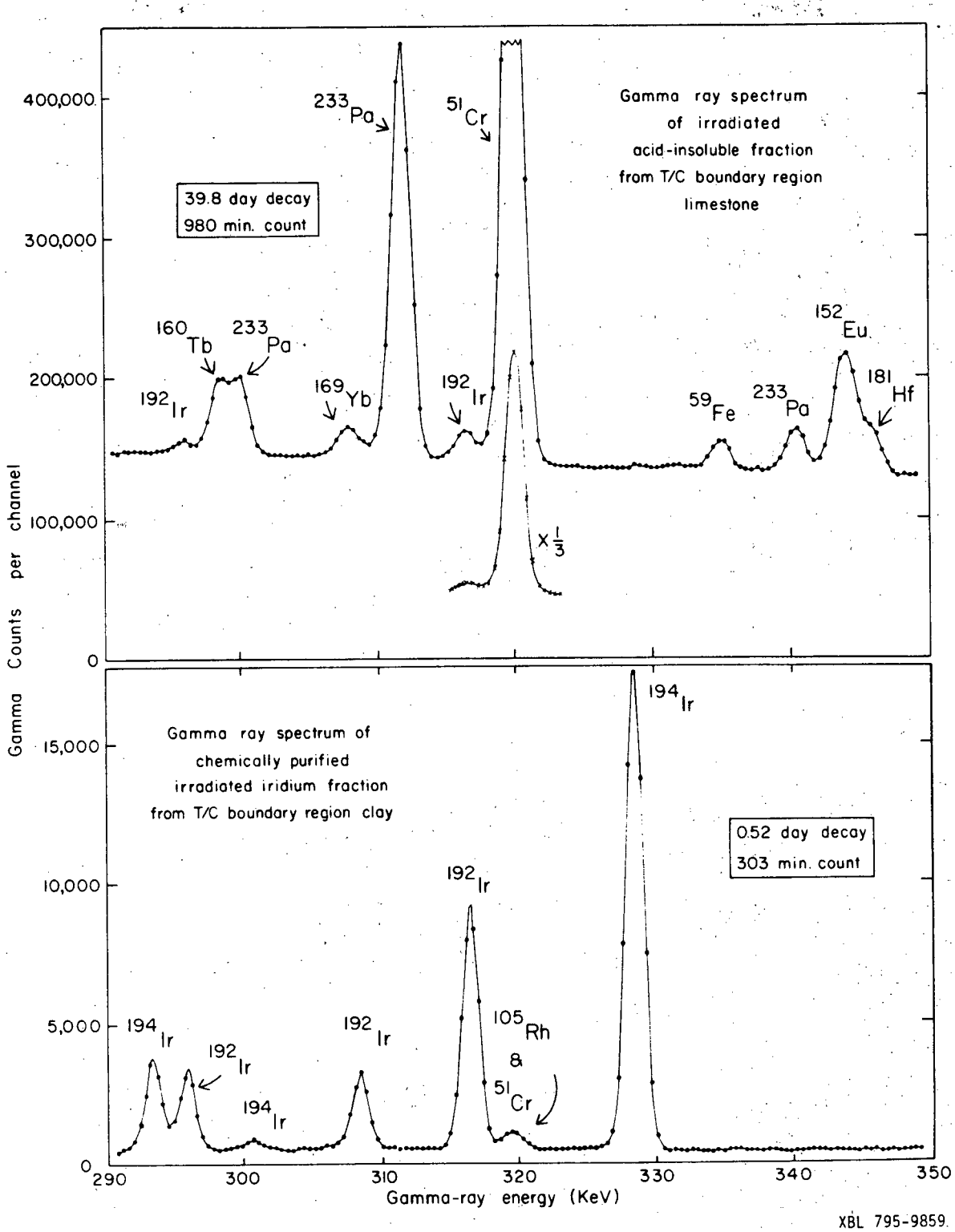
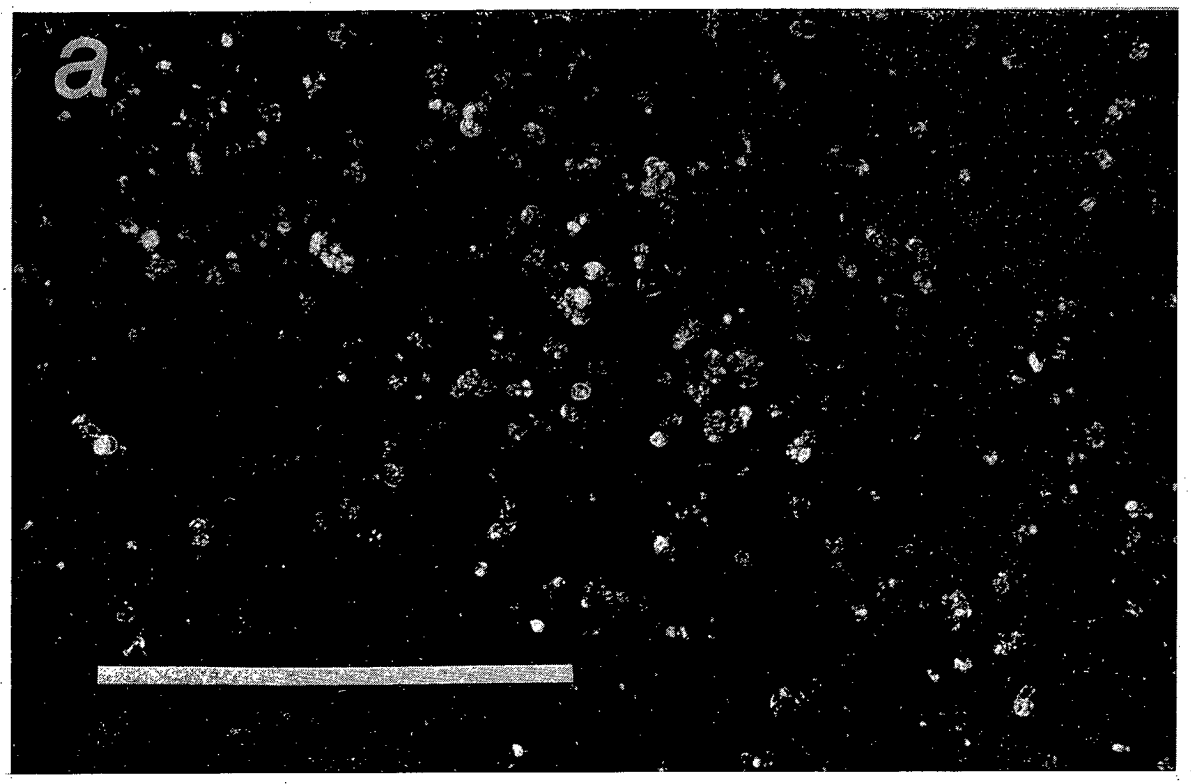

Gamma ray spectrum
of irradiated
acid-insoluble fraction
from T/C boundary region
limestone
39.8 day decay
980 min. count
233Pa
51Cr
160Tb
233Pa
192Ir
169Yb
192Ir
59Fe
233Pa
152Eu
181Hf
x 1/3
400,000
300,000
200,000
100,000
0
Gamma Counts per channel
Gamma ray spectrum of
chemically purified
irradiated iridium fraction
from T/C boundary region clay
0.52 day decay
303 min. count
194Ir
192Ir
192Ir
105Rh
&
51Cr
194Ir
192Ir
194Ir
15,000
10,000
5,000
0
290
300
310
320
330
340
350
Gamma-ray energy (KeV)
XBL 795-9859

Fig. 13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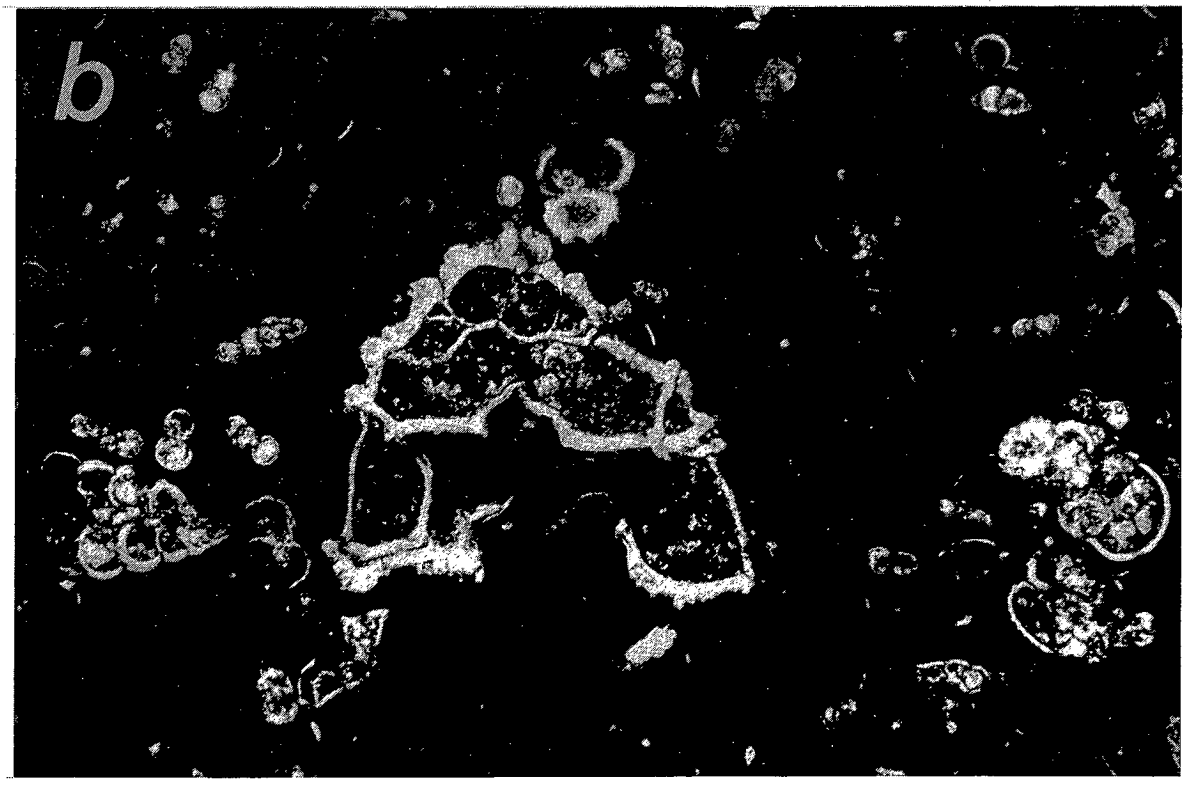
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UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720