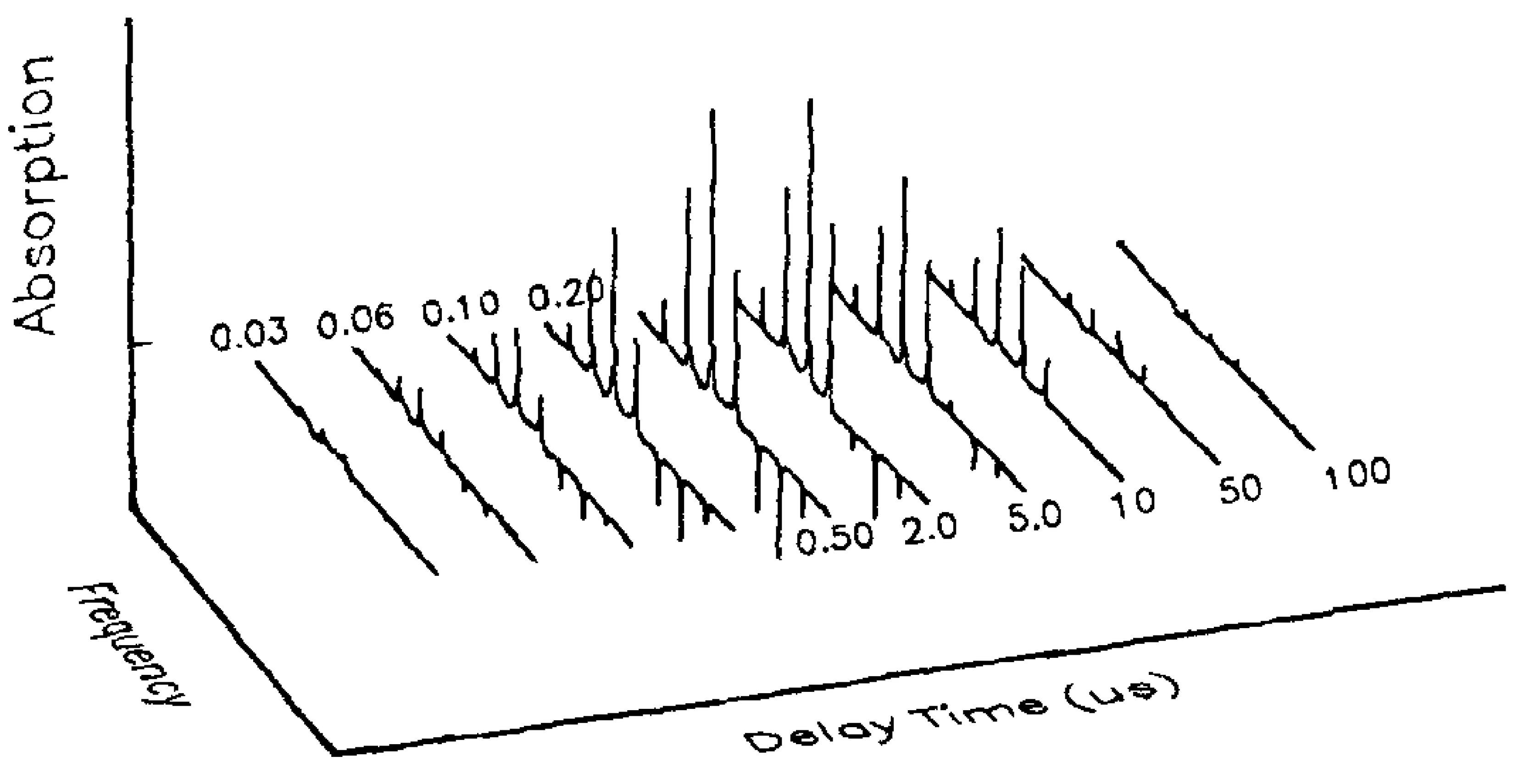

Figure 3. Time development of the FT-EPR spectra from the (perdeuterated) acetone ketyl radical formed by a pulsed laser induced hydrogen abstraction reaction in a solution of acetone (perdeuterated) in 2-propanol (perdeuterated)[8].

been shown that FT EPR is particularly suited for the study of the formation and decay of free radicals produced photochemically. The method provides the ultimate in sensitivity, spectral resolution, and time resolution. Examples of the application of FT EPR in studies of photochemical reactions are given in Figures 1–3. Figure 1 gives the spectra of the duroquinone anion radical formed by photoinduced electron transfer from zinc tetra-phenylporphyrin (ZnTPP) at various times after the excitation of ZnTTP[6]. Noteworthy is the appearance of emission as well as absorption peaks due to CIDEP. The time evolution of the amplitudes of the hyperfine lines in the spectrum of the benzoquinone anion generated by electron transfer from ZnTPP is given in Figure 2. A complete analysis gives detailed information on chemical rate constants as well as rate constants governing electron spin dynamics[7]. Finally, Figure 3 shows spectra from the acetone ketyl radical formed in a photochemical reaction of acetone with 2-propanol[8].

In conclusion it can be stated that the range of applications of EPR has been considerably broadened in recent years as a result of the increase in sophistication of instrumentation, measurement techniques, and data analysis. Particularly in the areas of biology and biochemistry these developments have led to a rapid growth in the number of research groups utilizing EPR.

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Collisions with Earth over geologic times and their consequences to the terrestrial environment

N. Bhandari

Collision of comets and asteroids appears to be a forceful mechanism as an alternative to volcanism for creating stress on life on the earth but a general theory of mass extinctions over geologic ages is difficult to formulate from the meagre observations available so far. Geochemical results in the Indian subcontinent at Cretaceous–Tertiary boundary are presented and the relative importance of Deccan volcanism and asteroidal impact hypothesis is discussed.

To kill the evil doers, wielding a sword
Like ferocious comet Keshava appears in the form of Kalki.
Victory to Thee! Oh Lord Hari

Jayadev in *Gita Govinda*
– circa 12th century AD

Examination of lunar craters, photographs of planetary and satellite surfaces taken by space missions and laboratory studies of lunar samples and meteorites have revealed the importance of interplanetary collisions as a prime process in the formation of planets and in carving out their surfaces as we see them today. In the early stages of the formation of the solar system, once

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the grains condensed from the cooling gas of the solar nebula, the process of collision took over. The growth of grain aggregates to planetesimals was initially controlled by non-disruptive collisional accumulation processes till gravitational forces became effective. The imprints of these initial planet-forming processes are still visible on the scarred faces of Mercury, the Moon and the ancient martian crust. Formation of the moon in a major terminal collision of Proto-moon with the Earth is the most favoured hypothesis[1,2]. In a highly truncated fashion, becoming rare with their dwindling population in the interplanetary space, the process is still going on and the remnants of these events can be found on the earth.

Much has been learnt from the study of crater density on the pre-Marè and post-Marè surfaces of the Moon and by dating of major Marè-forming events. The initial steep decline of the collision frequency 4.5–4.2 billion years ago, major collisional events which gave rise to Marè in the time bracket of 4.0 to 3.1 billion years (see eg Taylor[3]) and subsequent decrease in the population of interplanetary projectiles are now well established[4].

There are many candidates for major collisions with Earth which include Earth-crossing asteroids (Apollo and Amor asteroids), and long- and short-period comets[5,6]. Evidence of more recent interplanetary collisions in the last few million years comes from discrete groupings in cosmic-ray exposures ages of certain types of meteorites such as H4 chondrites. By analogy and proximity, such events as seen on the Moon and in the asteroidal belt, must have occurred on Earth too. The lunar data suggest that a crater (diameter $>D$ m) is formed with the frequency $N(>D) \propto D^{-2}$ and the same is found to be true for the surviving large craters (>22.6 km) on Earth[7]. The vestiges of many of these craters on Earth are rapidly obliterated because of dynamic surface processes due to continental motion, their associated geological processes, weathering, erosion, etc. Identification of impact structures, therefore, is difficult but the geochemical approach enables us to distinguish them from other terrestrial features because some elemental and isotopic compositions of extraterrestrial matter and Earth's crust are different. Although Earth originally formed out of meteorites, subsequent melting and separation of the core have taken iron and associated elements like iridium and osmium to the interior, leaving behind a crust depleted in siderophiles and enriched in lithophiles. Table 1 lists abundances of some of the distinguishing elements and isotopes which are crucial to this problem. Platinum group elements (PGE) like iridium and osmium are considered to be good geochemical markers of chondritic matter, although terrestrial processes of their enrichment are also known. Achondrites, on the other hand, have concentrations of PGE not much higher than the crust[8], but they will still have distinguishable isotopic ratio of $^{187}Os/^{186}Os$.

Grieve[7] has compiled the available information on terrestrial impact structures. Based on associated shock metamorphism and chemical anomalies, more than a hundred such features formed during the past 250 million years have been identified and using the geochemical approach the chemical characteristics of more than a dozen projectiles have been determined. India had its small share of extraterrestrial impacts in the Lonar crater in Maharashtra[9] and possibly also Ramgarh crater in Rajasthan, although geochemical markers in either case have not yet been demonstrated.

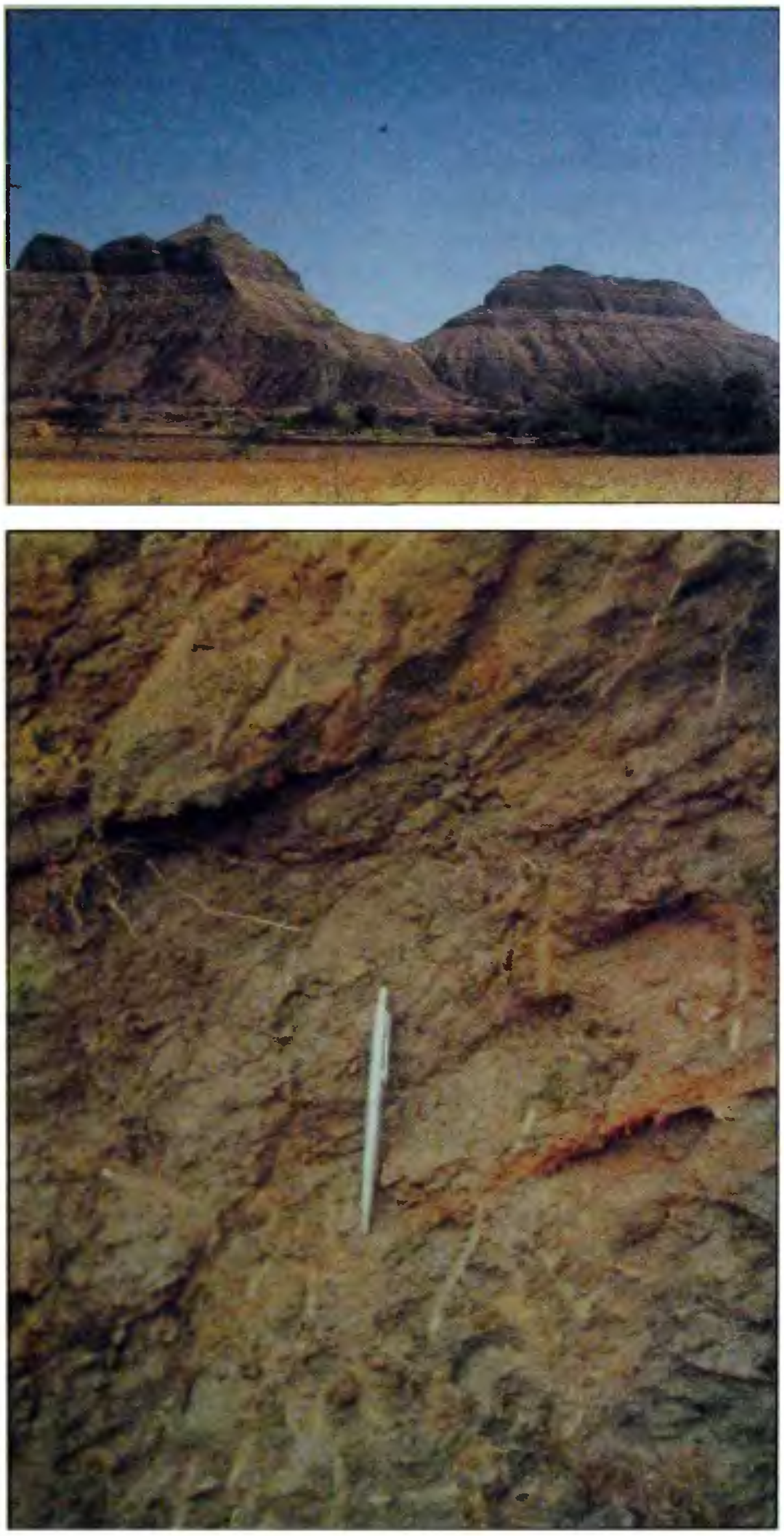
Earth is a unique planet in the sense that it harbours life and life is very fragile, critically dependent on environment and climate. In the past 600 million years several minor and five major events of mass mortality have occurred (Figure 1). The small craters do not

Table 1. Distinguishing elemental and isotopic signatures of Earth's crust and meteorites

	Continental crust (CC)	Oceanic crust (OC)	Primitive mantle (PM)	C1 chondrites (C1C) (ref 46)	Enhancement Factor C_1C/CC	Eucrites (ref. 8, 47)
Na (%)	2.3	2.08	0.25	0.5	0.217	0.349
Mg (%)	3.2	4.64	21.20	9.89	3.09	3.96
Al (%)	8.41	8.47	1.93	0.868	0.10	6.64
Fe (%)	7.07	8.16	6.22	19.04	2.69	14.56
Ni (ppm)	105	135	2000	11000	104.7	13
La (ppm)	16	3.7	0.551	0.2347	0.0146	3.26
Sm (ppm)	3.5	3.3	0.347	0.1471	0.042	2.03
Re (ppb)	0.5	0.9	0.25	36.5	73	0.0008–0.0097
Ir (ppb)	0.1	0.002	3.2	481	4810	0.0022–0.028
Os (ppb)	0.5	<0.004	3.8	486	972	0.003–0.018
U (ppm)	0.91	0.01	0.018	0.0081	0.0089	0.0082–0.104

Isotopic ratios (ref. 19)

	Crust	Meteorites
$^{187}Os/^{186}Os$	~10	~1
$^{187}Re/^{186}Os$	~400	~3.2



Top: Typical Deccan flow sequence. *Bottom*: KT boundary section from Um Sohryngkew River section in Meghalaya. The pen points to the 1.5 cm brown limonitic layer in which high iridium and osmium concentrations have been found.

affect the environment, albeit locally, but the consequences of large impacts can be catastrophic, since the kinetic energy of impact exceeds the energy involved in most terrestrial processes by many orders of magnitude (Table 2). There is still some debate about the partition of impact energy in various terrestrial processes but the impact is instantaneous and the shear magnitude of the power indicates that the consequences can be drastic[10]. Of the various agencies listed in Table 2, the most

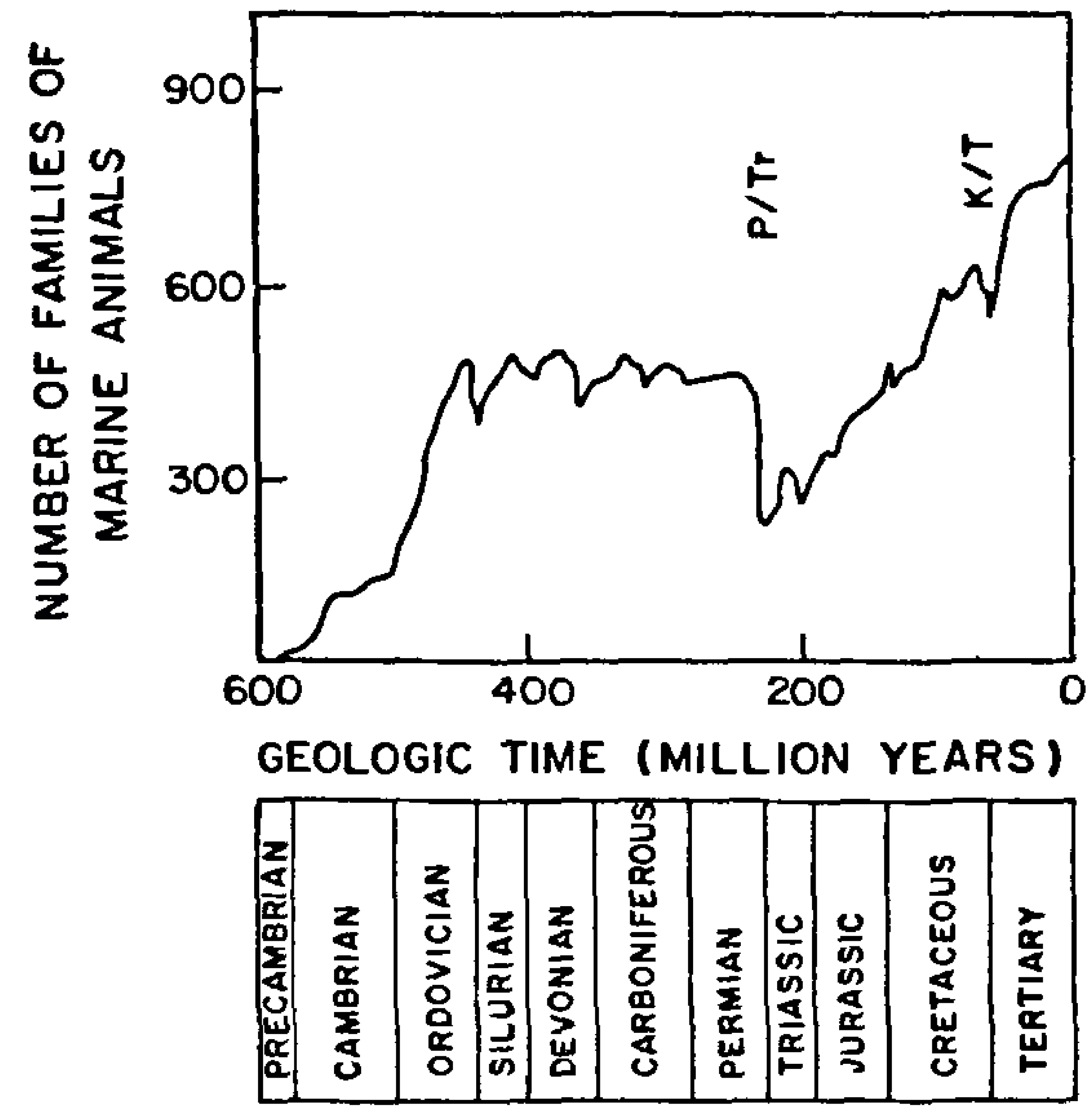


Figure 1. Variation in the number of families of marine vertebrates and invertebrates in the past 600 million years showing the five major and several minor phases of severe stress on life, (after Raup and Sepkoski[56],); Permo-Triassic and Cretaceous-Tertiary events of mass extinction are marked.

efficient transfer of energy to the atmosphere in an intense pulse occurs in a cometary impact.

Study of some continental and many deep-sea sediments, those lifted up by tectonic processes or collected from the ocean bed, have revealed at least three horizons which contain significantly high iridium compared to the adjacent material. These occurred at a time when Earth was going from one epoch to the other, indicative of major changes in the marine environment, i.e. Cretaceous to Tertiary about 66 Ma (ref. 11). Eocene to Oligocene about 34 Ma (ref. 12) and near-Pliocene to Pleistocene transition, about 2.3 Ma (ref. 13). There are other boundaries which show minor iridium enrichment (Table 3). Of these, the K/T transition has been the most dramatic as a number of phenomena (Table 4) associated with it, in addition to the presence of anomalously high concentration of iridium, have been observed such as presence of high temperature spherules[14], shocked quartz[15], soot due to forest fires[16], anomalies in isotopic composition of oxygen and carbon (see e.g ref. 17), possible change in isotopic composition of $^{87}Sr/^{86}Sr$ (ref. 18), non-lithospheric $^{187}Os/^{186}Os$ (ref. 19), and most importantly,

Table 2. Energy considerations

Asteroidal impact (10 km)	10^{31} ergs
Cometary impact (10^{18} g)	10^{31} ergs
Deccan volcanism	10^{33} ergs
Tambora 1815 volcano	8×10^{26} ergs
Krakatoa 1883 volcano	10^{25} ergs
Solar energy received annually	3.5×10^{31} ergs
Annual output of the internal energy of Earth	$\sim 10^{27}$ ergs

Table 3. Summary of some observations at boundary horizons

Boundary	Age (Ma)	Mortality	Iridium (ng/g)	$\delta^{13}C$	$\delta^{18}O$	Other features [Reference]
Near Pliocene–Pleistocene	2.3	Insignificant	One minor peak (0.11–4.7)	—	—	— [ref 48]
Eocene–Oligocene	34.5	Four sudden minor events	Iridium peak (0.10–4.1)	—	—	Microtektites [ref 12]
Cretaceous–Tertiary (K–T)	66	Severe	One sharp peak (5–100 ppb)* some minor peaks?	Sudden decrease	Sudden decrease	see Table 4 [refs 11, 30, 36]
Triassic–Jurassic (Tr–J)	208	Major	Small peak (0.4 ppb)	Decrease	Decrease	Regression [ref 10]
Permian–Triassic (P–Tr)	245	Sudden and severe	(0.003–0.09)	Sharp decrease	—	— [ref 49, 50]
Lower Mississippian	330	—	(0.02–0.56)			[ref 51]
Devonian–Carboniferous (D–C)	360		Four minor iridium peaks (0.02–0.31)	—	—	Sudden fall in sea level [ref 52]
Frasnian–Famennian (F–F)	367	Sudden and significant	Small iridium peaks (0.02–0.08)	Decrease	Decrease	— [ref 53]
Late Ordovician	440?	Sudden	Weak iridium anomaly (0.03–0.05)	—	—	Regression [ref 54]
Precambrian–Cambrian (P€–€)	~600	—	(0.002–0.011)	Decrease	Decrease	— [ref 55]

*In addition to the main peak, some smaller peaks have been found (0.08 to 0.3 ng g^{-1}) which may be due to variation in carbonate content or perturbations of the main peak[37]

Table 4. Events at Cretaceous–Tertiary transition

1. Extinction of a large number of land and marine organisms (~50% of genera and ~15% of families). Only next to P/Tr extinction in severity, gradual or stepwise decline superimposed by a sudden and short event of extinction at KTB
2. Recession of sea causing stress on marine life
3. Iridium-rich layer in continental and marine deposits (global), sharp and sudden increase [$>10^2$ times more iridium than present in the crustal material] Asteroid or cometary impact?
4. Deccan volcanism $\geq 10^6$ km^3, 63–70 Ma with an intense pulse of short (~0.5 Ma) duration at KTB as suggested by the palaeomagnetic data
5. Forest fires $\delta^{13}C$ anomaly in soot
6. A severe temperature fluctuation a cold wave? Superimposed on a slow global cooling, followed by a severe heat pulse based on $\delta^{18}O$
7. Sanidine and basaltic spherules formed from high temperature melt by quick cooling
8. Severely shocked quartz, crystal deformation most likely to have been produced by an impact
9. Acid-rain HNO_3 $^{87}Sr/^{86}Sr$ isotope excursion, H_2SO_4 etc
10. Near-disappearance of pollens and plants
11. Amino acids (α-amino isobutyric acid) probably of cometary origin, tens of centimetres above and below the KTB but absent at KTB at Stevns Klint, Denmark[57]

extinction of a large fraction of marine and non-marine biota[20]. The extinction of dinosaurs is also associated with this event. It is significant that at the same time, large-scale volcanic activity occurred in central India which gave rise to the vast Deccan plateau, containing more than a million cubic kilometres of lava. The Deccan-activity seems to have occurred over an extended period of time[21] and its timing is not precisely determined but there are some indications that major pulse of activity occurred during the magnetic reversal 29R, during a brief period of a million years or less[22]. It must be noted here that Earth looked quite different at that time from what we see now. A large part of the continents was under water, sea level was at the highest level ever recorded, temperatures were warm and equitable, and the configuration of continents was quite different. India, for example, was far in the south, around 20° S passing over the Reunion Island hot spot, Atlantic was just opening up and carbonate was the main sedimentary type depositing at that time.

What is the connection between these various phenomena? Are they related through a common cause, or, are some of them independent? Cosmochemists and geochemists believe that impact is the cause, be it asteroidal or cometary, and there have been attempts to find correlated periodicity in crater formation and mass extinction[23] to support this viewpoint. Many geologists and palaeontologists, on the other hand, favour terrestrial hypothesis and suggest the Deccan volcanism to be the cause (see e.g ref. 22). As either of these events, impact or volcanism, can give rise to nearly the same scenario through a series of physicochemical processes, the debate has remained inconclusive. In either case, the changes are triggered by ejection of dust, accompanied by release of greenhouse gases, acid rains and blanketing of sunlight, resulting in cessation of photosynthesis and extinction of fauna and flora. The presence of shocked quartz and spherules favour the impact hypothesis. The main difference between the two scenarios is that impact is instantaneous whereas volcanism is sustained over extended period of time and can maintain continuous stress on life.

As an attempt to resolve this debate, we thought the best place to look for is in the vicinity of Deccan, because its effect will be most pronounced in its neighbourhood. We have, therefore, analysed marine and continental sediments from India which deposited near or at the Cretaceous–Tertiary boundary (KTB)[24]. The east coast of India has abundant exposures of upper Cretaceous sediments, most notable amongst them being at Pondicherry, Thiruchirapalli, Meghalaya and some section may be exposed in Zanskar[25]. The K/T sequence have unconformity at most sites except in the Um Sohryngkew River section of Meghalaya[26]. The Um Sohryngkew River section has the added advantage, other than its proximity to the Deccan, that it is a shallow marine section and has high deposition rate estimated to be about 40 mm/ka (ref. 27). This may enable us to determine elemental profiles with fine-time resolution. As a result of studies carried out by the Punjab University group, an intertrappean bed extending west to east from Saurashtra to Nagpur and to Hyderabad in the south has been identified as the uppermost horizon containing dinosaurian fossils[28,29]. The extinction of dinosaurs is usually considered to have occurred at KTB, and, therefore, we have also examined these intertrappean sediments. The location of various sites is shown in Figure 2.

A suite of elements, including siderophiles (Fe, Co, Ni, Cr, Ir, Os), lithophiles (Na, K, Ca, Sc) and rare earths (La, Ce, Nd, Sm, Eu, Gd, Tb, Yb and Lu), have been measured in addition to Sb, Hf, Th, etc. using neutron-activation-analysis technique, followed by radiochemical separation in the case of iridium and osmium, in collaboration with P. N. Shukla, J. Pandey and M. Gupta (refs. 24, 26, 30, 31). The depth profiles of some crucial elements are shown in Figures 3 and 4 for Um Sohryngkew River and Takli sections. The following conclusions have been drawn from the distribution pattern of these elements and their depth profiles based on this work.

(i) The Um Sohryngkew River section in Meghalaya contains a strong and narrow peak of iridium (maximum iridium concentration = 12.1 ng g^{-1}), superimposed on a broad and mild band (Ir = 0.2 ng g^{-1}). The iridium is enriched by a factor of about 10 in the broad band (of about 10–20 Ka duration) and by almost a factor of about 500 in the sharp peak above the typical Cretaceous shales (Ir $\cong$ 0.02 ng g^{-1}).

(ii) Osmium/iridium ratio in the peak (0.36) and the broad band (1.5 ± 0.3) are different, suggesting two distinct sources. Both these values are different from ~ 1, characteristic of most meteorite types. It is possible that the osmium/iridium ratio altered by diagenesis due to differential remobilization, as discussed by Kyte *et al.*[32] and Tredoux *et al.*[33], but it is unlikely that such a process will result in the profile observed in the Um Sohryngkew River section.

(iii) The level of iridium anomaly in Meghalaya section is similar to that found elsewhere on the globe, showing no special enhancement in the vicinity of the Deccan, thus ruling out significant contribution from the Deccan volcanism.

(iv) Iridium in the Deccan sediments 50 pg g^{-1} to 100 pg g^{-1} is too small (Figure 4) to give rise to the anomalous peak of iridium at the KTB, but similar in concentration to the broad band observed just above and below the KTB layer in Um Sohryngkew River section (Figure 3).

(v) As noted earlier[24], the iridium peak is accompanied

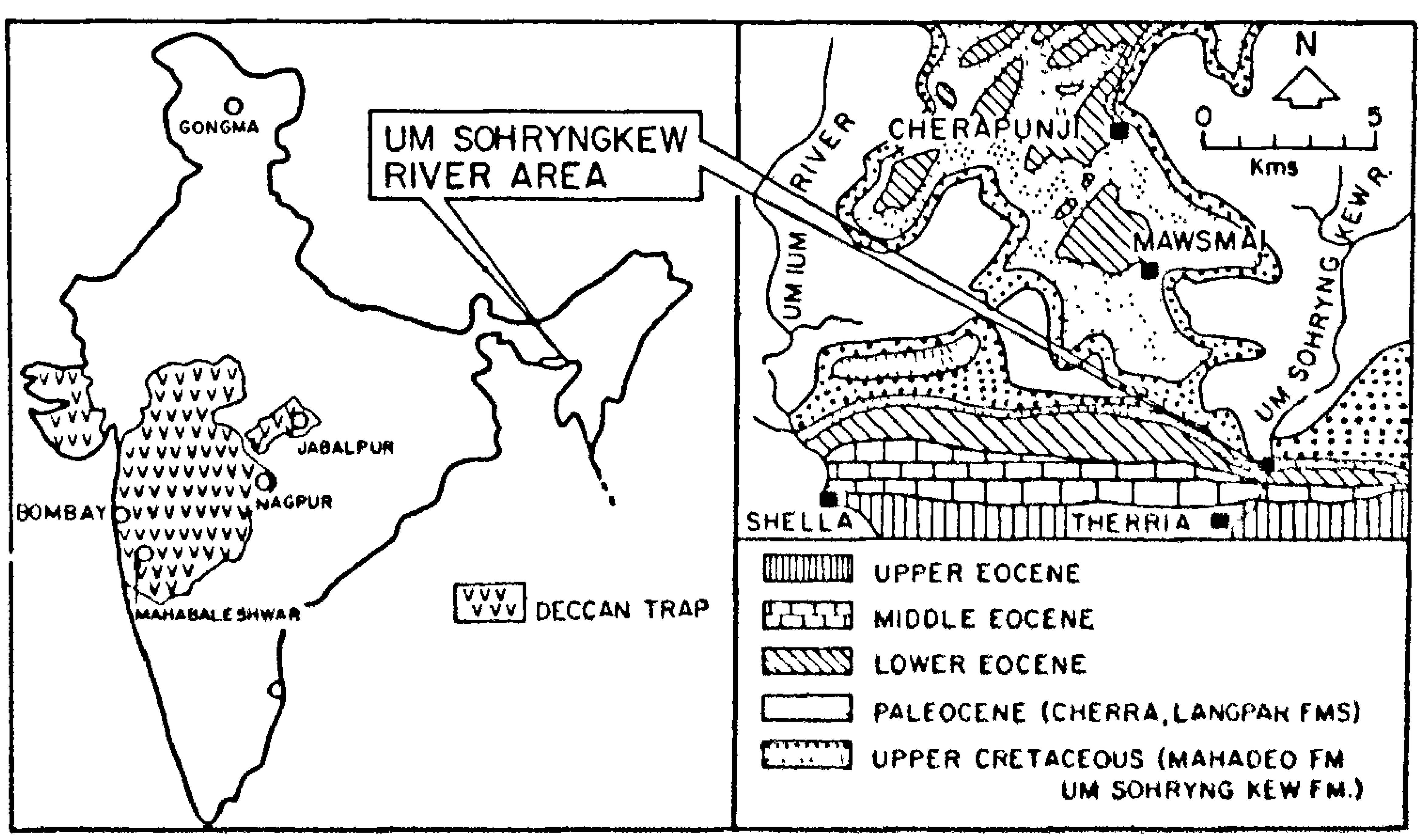


Figure 2. Map showing Cretaceous Tertiary boundary sites of the Um Sohryngkew River section, Meghalaya and the Deccan trap region, India.

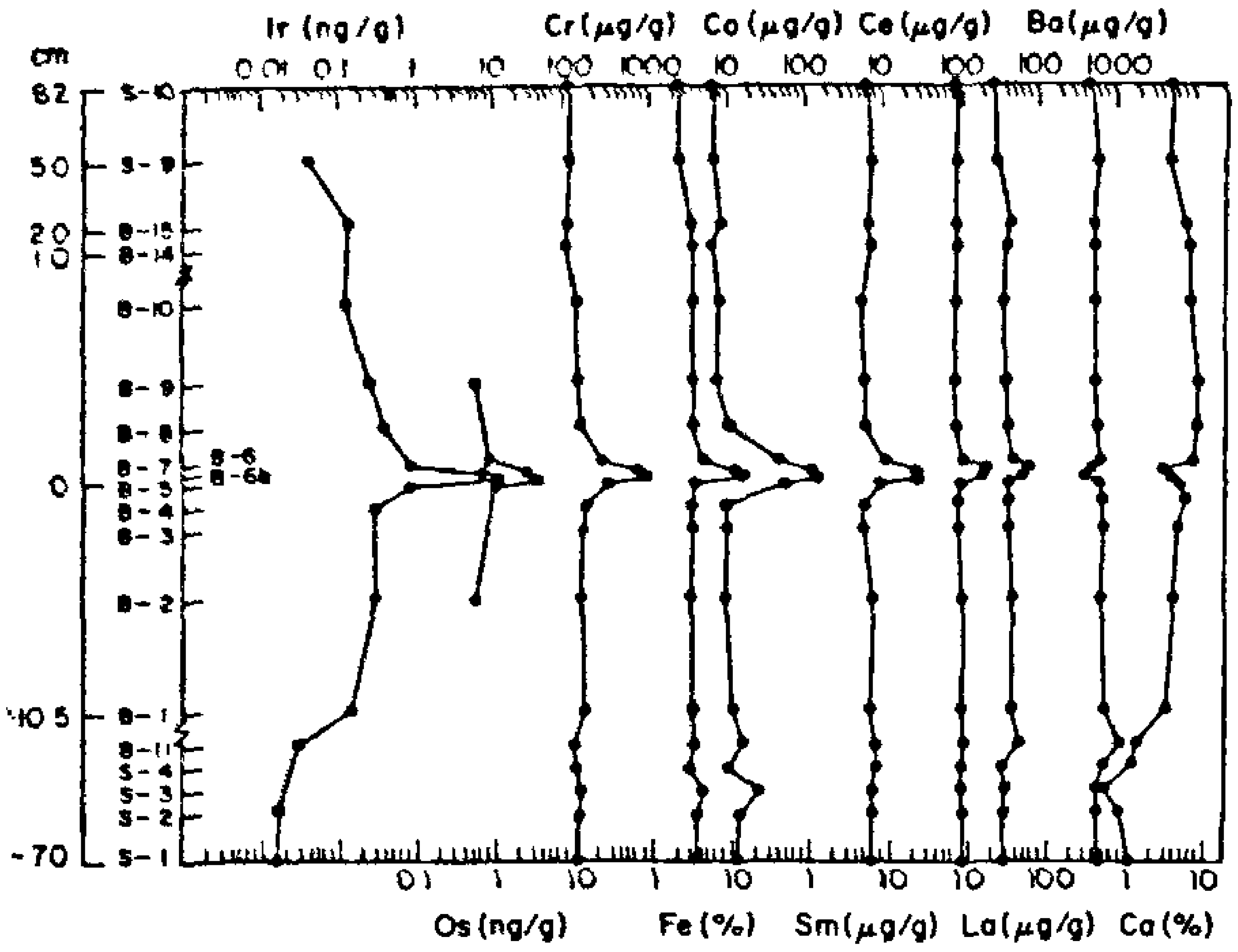


Figure 3. Depth profiles of Ir, Os, Cr, Fe, Co, Sm, La, Ba and Ca across the KTB in Um Sohryngkew River section (after Bhandari *et al.*[29])

by a simultaneous enrichment of Fe, Co, Ni, Cr and also REE. The siderophile enrichment has been observed at KTB at all the sites around the globe but REE enrichment has been seen only at a few places[33]. In fact at Stevns Klint[34,35] there is a depletion of REE where the iridium peak occurs.

(vi) In the intertrappeans at Nagpur, the concentration of REE, Cr and Ir is above their levels in the basalt flows and determines the limit to which volcanism can contribute to their deposition in sediments (Figure 4). Their concentrations, in any case, are too low to explain their anomalous peak enhancement at KTB[31].

From these observations, it appears that the Deccan volcanism is inadequate to explain the peak concentration of iridium found in the marine sections at KTB. The integrated deposition of iridium between the two flows at Takli, amounts to 20 ng cm^{-2} and falls much short of the integrated deposition in the broad band and peak of iridium observed at the Um Sohryngkew section which is 70 ng cm^{-2}. Although, there are many such flows and intertrappeans but, unless there are some which are highly enriched in iridium, they cannot account for the highest values of iridium observed at Stevns Klint which is around 185 ng g^{-1} (carbonate-free basis) and the observed large integrated deposition in marine sections. The enrichment of iridium and other siderophiles may, therefore, well be due to cometary or asteroidal impact. But could the broad- and mild-band observed in Meghalaya be due to the Deccan? The osmium/iridium ratio in the band, different from the value in the peak, suggests a different source. And its deposition is not as instantaneous as impact hypothesis would envisage. The crucial parameter would be to measure osmium/iridium ratio in the Deccan sediments and to determine if it is identical with the value of 1.5 seen in the broad-band. These measurements are currently in progress in our laboratory.

Once the impact hypothesis is accepted as a cause of

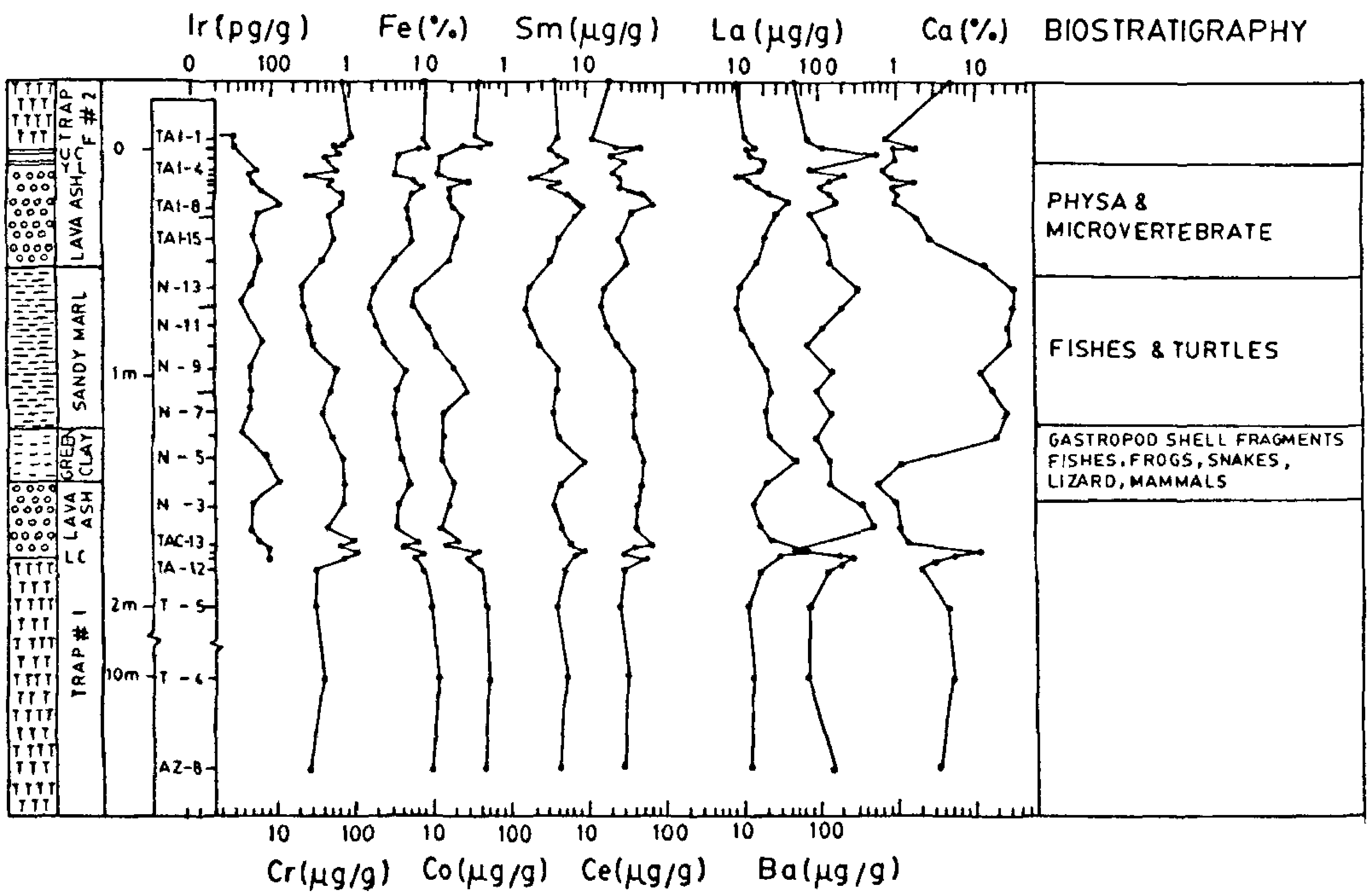


Figure 4. Depth profiles of Ir, Cr, Fe, Co, Sm, Ce, La, Ba and Ca across the intertrappeans of the Takli section, Nagpur (after Bhandari *et al.*[30]).

the iridium anomaly at KTB, it is desirable to go into the question whether it was asteroidal or cometary. An asteroidal collision is a single-isolated event which should give rise to a sharp and sudden iridium anomaly, whereas a cometary impact will be preceded and followed by a high flux of cometary debris which usually populates the cometary orbit. The iridium peak will be superimposed on extended, slightly higher than background level of iridium in such a case. The observed profile of iridium in meghalaya, and some other sites is compatible with such expectations. The distribution of trace elements in sediments is, unfortunately modified by bioturbation, mixing and diagenesis and therefore, by itself, it cannot provide any compelling evidence. Other distinguishing features of the two would be in size and frequency of the craters. Asteroidal impact would require a 200-km impact to account for the observed iridium, whereas cometary collisions, most favourably believed to occur due to gravitational perturbations of a nearby star, may result in a large number of small impacts closely spaced in time, resulting in multiple iridium peaks. No evidence of multiple impacts has been found in deep-sea sediments[36], but multiple-iridium layers in Gubbio sections have been reported[37]. Whether there is a close grouping in age of some terrestrial impact craters is also doubtful, because of poor precision in dating. The 32-km diameter Manson crater, Iowa, is a possible candidate because its age is indistinguishable from KTB[38]. If comets bring interstellar material from the outer fringes of the solar system which may, at least partly, be freshly synthesized in stellar nuclear processes, they can be distinguished by the peculiar isotopic characteristics from the asteroidal material which is basically solar system material akin to meteorites. There are a number of such distinguishing isotopes but their measurements with the required precision are difficult[39].

Consequences on Earth

It is important to assess if a general theory of extinction by impact is tenable on the basis of the evidence available so far, or, at least, if an impact can bring about significant changes in terrestrial environment, giving rise to transition from one epoch to another[40]. Table 3 summarizes the available observations at some of the boundaries. It is clear that there is no significant enrichment of PGE at most of the boundaries, including the Permian-Triasic boundary[41] where the most severe extinction has occurred. Many of these horizons are characterized by decrease in $\delta^{13}C$, indicator of a major biomass change and also of $\delta^{18}O$ which indicates change in the atmospheric temperature. Tektites or spheroids, indicative of an impact have been found at some sites. Impact of an achondritic bolide, depleted in PGE, can be invoked to explain absence of PGE enhancement. Impact of a bolide can thus offer a plausible explanation of all the observed phenomena at the boundaries listed in Table 3.

The sequence of events following incidence of a bolide has been modelled and discussed much in the literature. A heat wave through the atmosphere, resulting in combination of N_2 and O_2 to give rise to nitrogen oxides which eventually results in HNO_3, precedes the impact[42]. Impact on carbonate-rich sediments which existed in the upper Cretaceous would result in release of large amounts of CO_2, increasing CO_2 concentration in the atmosphere by a factor of 2 to 10 (ref. 43) O'Keefe and Ahrens[43] have also shown that in a shallow marine impact, additional dessolution of CO_2 from the oceanic photic zone could slowly release more CO_2 which would increase global temperatures by 2 to 10 K for 10^4 to 10^5 years by greenhouse effect. Blanketing of sunlight by dust and smoke thrown aloft would screen light and inhibit photosynthesis. Thus there are several forceful processes which can bring about severe stress on life, resulting in mass mortality.

Influx of cometary or meteorite material containing prebiotic organic matter which served as source of precursors of life is sometimes invoked to explain quick development of primitive life on Earth[44]. Some compounds such as amino acids of extraterrestrial origin have been found in meteorites[45]. If this be true, it appears as a clever design by nature that the same process which gave rise to life on Earth can cause its extinction.

Acknowledgements. Some of the work reviewed here was done in collaboration with Drs P. N. Shukla, M. Gupta and J. Pandey, who have also provided useful suggestions in preparing this article.

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