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Palaeogeography, Palaeoclimatology, Palaeoecology 178 (2002) 1–17

PALAEO

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A possible causal relationship between extinction of dinosaurs and K/T iridium enrichment in the Nanxiong Basin, South China: evidence from dinosaur eggshells

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Received 10 November 2000; accepted 27 August 2001

Abstract

Multiple distinct iridium anomalies have been observed in dinosaur eggshells in the K/T boundary sections of the Nanxiong Basin, Guangdong Province, South China. The eggshells collected at and near the fossil–pollen-defined K/T boundary interval show iridium increases of about 19 and 28 times, respectively, above the background level. The enrichment of other trace elements in the eggshells occurs mostly at and near the interval. The distribution of Ir-bearing levels proves the existence of at least six Ir-delivering events from the latest Cretaceous into the earliest Paleocene. The enrichment of Ir and other trace elements in eggshells may have been caused by the assimilation of these elements into the dinosaur body through food, and then into the eggs laid by them.

Two types of pathological development, i.e. variation in eggshell thickness and eggshell microstructure, have been observed from the basin. It seems that they occur frequently during the K/T transition. The physiological mechanisms producing pathologic dinosaur eggshells are evaluated in the light of homologous phenomena occurring in living birds. On this basis, it is concluded that the formation of pathologic dinosaur eggshells was caused by anomalous trace element concentrations. The extinction of the dinosaurs in the Nanxiong Basin did not occur instantaneously, but spread out within 250 ka with major extinction beginning at the boundary interval. The cause may have been environmental poisoning and adverse changes in climatic conditions as indicated by the geochemical analyses of the dinosaur eggshells. A repeating short- and long-term geochemically induced environmental stress adversely affected the reproductive process and contributed to the extinction of the dinosaurs. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: K/T boundary; Ir anomaly; trace element; dinosaur eggshell; dinosaur extinction

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1. Introduction

The extinction of dinosaurs at the end of Cretaceous is considered to be one of the most dramati-

ic events in the Earth's history. The potential significance of the event was enhanced by the suggestion of Alvarez et al., (1980) that the dinosaur extinction could be attributed to the impact of an extraterrestrial body on earth as indicated by iridium enrichment at the K/T boundary. It has sparked an immense amount of interdisciplinary research on this problem and expanded into one of the truly great debates in the history of science. Many arguments have focused on two questions: What caused the extinction of dinosaurs, terrestrial or extraterrestrial in origin, and how long did the event last, gradual or catastrophic? After two decades, however, the intense debate appears to be far from over although the impact hypothesis has gained precedence. Neither the proponents of extraterrestrial nor terrestrial hypotheses have been able to achieve a consensus within the scientific community (Courtilot, 1999; Keller and Stinnesbeck, 1996; Stinnesbeck and Keller, 1996). A major obstacle to determining the cause of the dinosaur extinction is that virtually all the available information on the dinosaur fossils at and near the K/T boundary is relatively rare and is therefore indirect or speculative.

The best record we can use so far to discuss this issue is from the Western Interior of North America (Archibald, 1996). Another area that holds promise for clues to dinosaur extinction is the fluviolacustrine section in the Nanxiong Basin of Guangdong Province, South China. A record of dinosaur egg fauna spanning the Cretaceous–Paleocene boundary seems to provide some direct evidence for interpreting the dinosaur extinction (Zhao, 1978, 1994; Zhao et al., 1991; Zhao et al., 1993; Erben et al., 1995). In 1998, we reported the observation on three Ir anomalies in the dinosaur eggshells collected at and near the pollen-defined K/T boundary interval from the CGY–CGD Section of the Pingling Formation, located south of Datang Village at the northeast margin of the Nanxiong Basin (Zhao et al., 1998). Therefore, the possibility of another approach to this problem lay in a search for the anomalies in fossil vertebrates in the K/T sections.

Since iridium enrichment has not been observed in any vertebrate fossils and all Ir data were only reported from the geological boundary clay or

sediments in previous investigations, our observation potentially has important implications for the interpretations of the dinosaur extinction during the K/T transition time. It allows us to link directly events marked by the K/T iridium enrichment with the dinosaur extinction. Success in locating Ir anomalies in the dinosaur eggshell samples from the CGY–CGD Section led us to search in another continuous stratigraphic sequence of dinosaur eggshells (the CGT–CGF Section) from this basin (Fig. 1). In addition, we also include some supplementary observations made on the eggshells from the CGY–CGD Section of the Pingling Formation and the CGH Section of the underlying strata, the Yuanpu Formation. Detailed examination of eggshell samples, in addition to the original CGY–CGD Section, has provided us with data regarding the distribution of elemental abundance pattern contained in the dinosaur eggshells across the boundary interval. All the eggshell samples used in this study were identified as *Macroolithus yaotunensis*. This species was chosen because it is common and widely distributed in this basin. The original eggshell structure is well preserved (Zhao, 1993, 1994).

We now report elemental abundance patterns and paleobiological observations of the dinosaur eggshells over a large area with common paleoenvironmental setting in the Nanxiong Basin. Interpretation of the data provides clues to the event

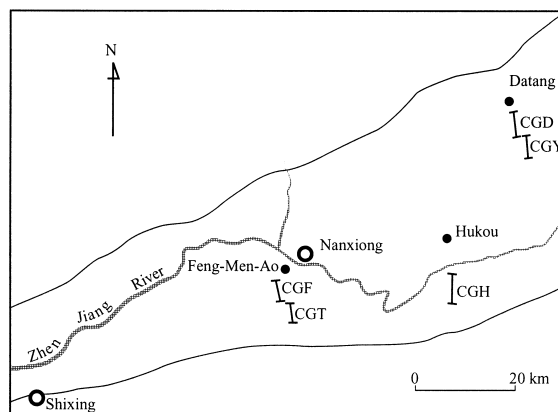


Fig. 1. Datang and Feng-Men-Ao areas in the Nanxiong Basin. Both have exposed K/T boundary sections that contain dinosaur egg and other vertebrate fossils.

marked by the K/T iridium enrichment with the dinosaur extinction.

2. Geological setting of dinosaur eggs in the Nanxiong Basin

The Nanxiong Basin is a small intermontane basin situated in northern Guangdong Province, South China. The basin is elongated with its axis oriented northeast–southwest. The topography within the basin is a hilly terrain, with rolling hills less than 50 m high. The Upper Cretaceous to Tertiary sediments are well-developed, and attain a maximum thickness of over 7000 m. The Yuanpu, Pingling, and Shanghu formations in ascending order are exposed over a large area of the basin (Fig. 2), and are Late Cretaceous and early and middle Paleocene in age (Zhou et al., 1977; Zhao et al., 1991; Erben et al., 1995). These units are predominantly fluviolacustrine or lacustrine in origin and were deposited in rivers, streams, and lake in the basin. Many well-preserved nests of dinosaur eggs and eggshell fragments were discovered in fluvial sediments of two formations, the upper part of the Yuanpu Formation and throughout the Pingling Formation (Young, 1965; Zhao et al., 1991; Erben et al., 1995).

Stratigraphically, the fossil eggs appear in the upper part of the Yuanpu Formation, and in the Pingling Formation. According to the field observation, complete eggs in nest and eggshells fragments occur with an appreciable frequency in sedimentary sequences of the Pingling Formation, from which more than 30 clutches of eggs have been collected by the IVPP and other institutions from 1961 to 1996. In the CGY-CGD Section, autochthonous eggshell fragments and occasional complete eggs occur through the sequence of the Pingling Formation. In many cases they were found in heaps (Fig. 3A), and may represent clutches destroyed in situ or nearby. In the CGT-CGF Section, definitely autochthonous eggshells are also found in the Pingling Formation. The two clutches of eggs, a clutch with 20 eggs (V. 2784) and the other with nine eggs (V. 2785), described by Young (1965) and by Zhao (1975),

may come from about 220 to 230 m of CGT and 80–90 m of CGF, respectively. About 1 km to the east of Feng-Men-Ao Village, a clutch of 18 eggs (Fig. 3B) has been discovered just below the boundary between the Pingling and Shanghu formations. Obviously, the complete clutches are preserved in situ, and show no trace of transport. Excellent preservation of the clutch geometry indicates that the egg laying and sediment formation were nearly synchronous. The eggshell fragments are lying in or around nesting sites. This case strongly suggests that the sediments with abundant eggshell fragments were clearly not re-worked.

The position of the K/T boundary in the studied area has not been defined precisely. A basalt intercalated within mudstone beds in the upper part of the Yuanpu Formation has given a K/Ar dating of 67.2 Ma (Zhao et al., 1991). Undoubtedly, the overlying strata, including the top of Yuanpu Formation and the whole Pingling Formation, were laid down after that time, indicating Maastrichtian in age. In previous works the K/T boundary was placed at the contact between the Pingling Formation and Shanghu Formation based on the level marked by the disappearance of the dinosaur eggshells (Fig. 2). The boundary profile, the CGY-CGD Section located south of Datang Village about 30 km to the northeast of the Nanxiong county seat (Fig. 1), has been studied in detail by biostratigraphy, magnetostratigraphy, sedimentology and geochemistry, and presently serves as an informal stratotype section for the K/T boundary. Sloan (1987) reported a member of the Asian lizard *Polyglyphanodontinae* from the upper 80 m of “the Nanxiong Formation” (should be ‘the CGD Section of the Pingling Formation’). It can be compared with “the previously most specialized form known, *Polyglyphanodon* from the latest Lancian North Horn Formation of North America, just under the Puercan Wagonroad fauna”. In view of conformity between the Pingling and Shanghu formations, he argued that the upper part of the Pingling Formation could be early Paleocene (Puercan).

Erben et al. (1995) defined the palynological K/T boundary in the upper part of the Pingling

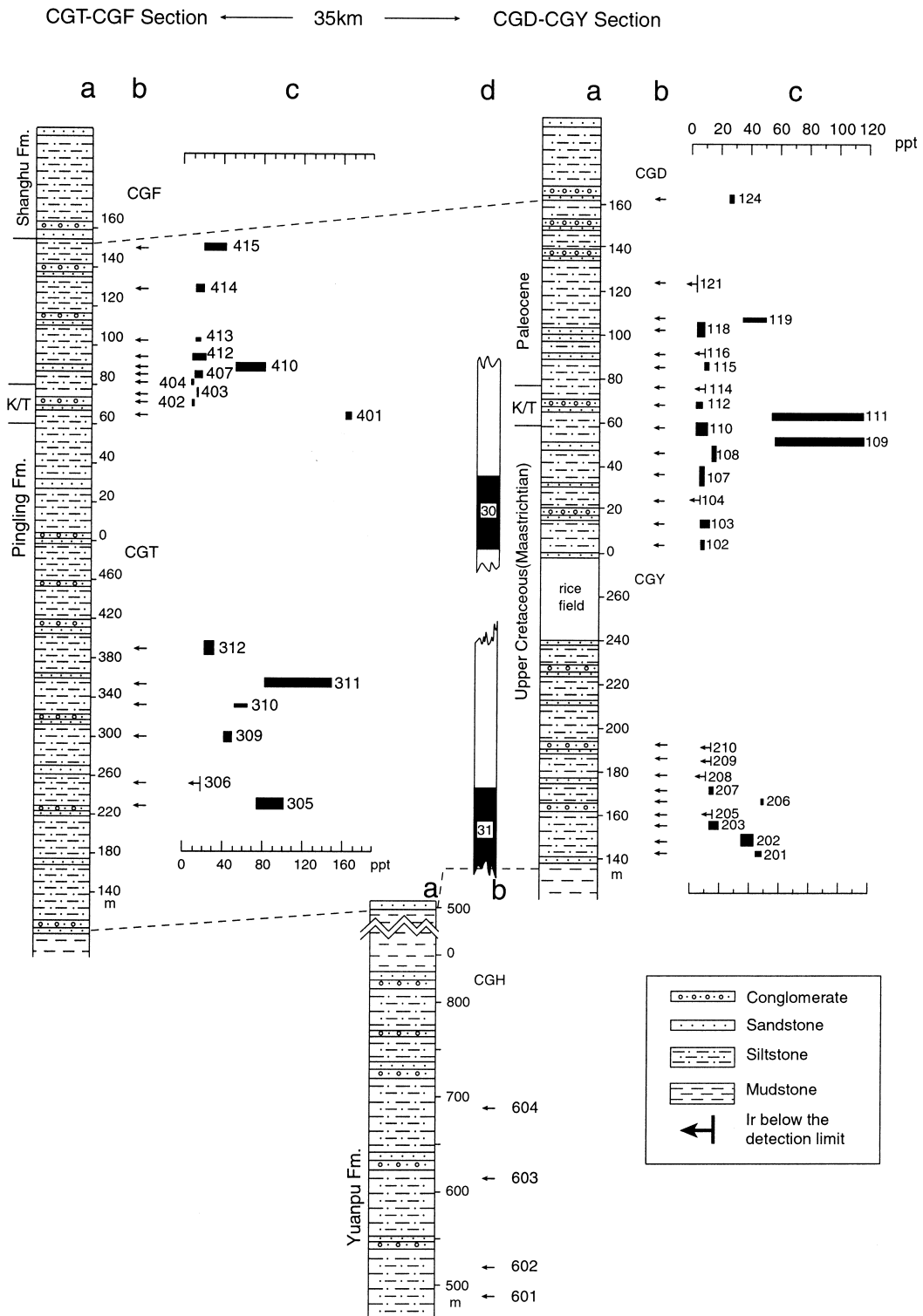


Fig. 2. Stratigraphy of the studied K/T sections in the Nanxiong Basin. (a) Lithology and meter levels. (b) Stratigraphic positions of the eggshell samples used in RNAA. (c) Ir abundance of the eggshells (ppt, 10^{-12} g/g). (d) Magnetic chrons.

Formation, which occurs in a stratigraphic interval of approximately 20 m, but no sharp line can be drawn. This corresponds to a time span of about 50 000 yr, assuming a sedimentation rate of 40 cm/ka (Erben et al., 1995). They documented that a significant floral change occurred in a stratigraphic interval from 57 to 78 m in the CGD Section of the Pingling Formation. The horizons below 57 m contain the typical Cretaceous palynomorphs *Cyathidites*, *Lygodiosporites*, *Cicatricosisporites*, *Gabonisporis vigourouseii*, *Chomotrilletes almegrensis* and *Schizaeoisporites cretaciis*. At the level of 60 m the first Tertiary pollen appears, and above 78 m yields Tertiary forms represented by *Pinuspollenites ruginosa*, *Abiespollenites*, *Abietineaepollenites*, *Piceapollenites*, *Triatriopollenites bituitus*, *Paranyssapollenites striatus*, *Juglanspollenites verus*. Thus, the K/T boundary is located at 57–78 m in the CGD Section. The preliminary magnetostratigraphy shows

that this boundary interval falls within 29R (Zhao et al., 1991; Erben et al., 1995). It is interesting that three iridium anomalies (Zhao et al., 1998) and multiple positive oxygen-isotope excursions (Zhao and Yan, 2000) were first observed in dinosaur eggshell samples collected at and near the pollen-defined K/T boundary interval. It is considered, therefore, that in the Nanxiong Basin it is acceptable to take for the K/T boundary as defined by fossil–pollen changes.

3. Iridium and other trace element patterns in dinosaur eggshells in Nanxiong Basin

3.1. The CGY-CGD Section

Our first experiment involved radiochemical neutron activation analysis (RNAA) (Kong and Chai, 1989) of 20 eggshell samples from this sec-

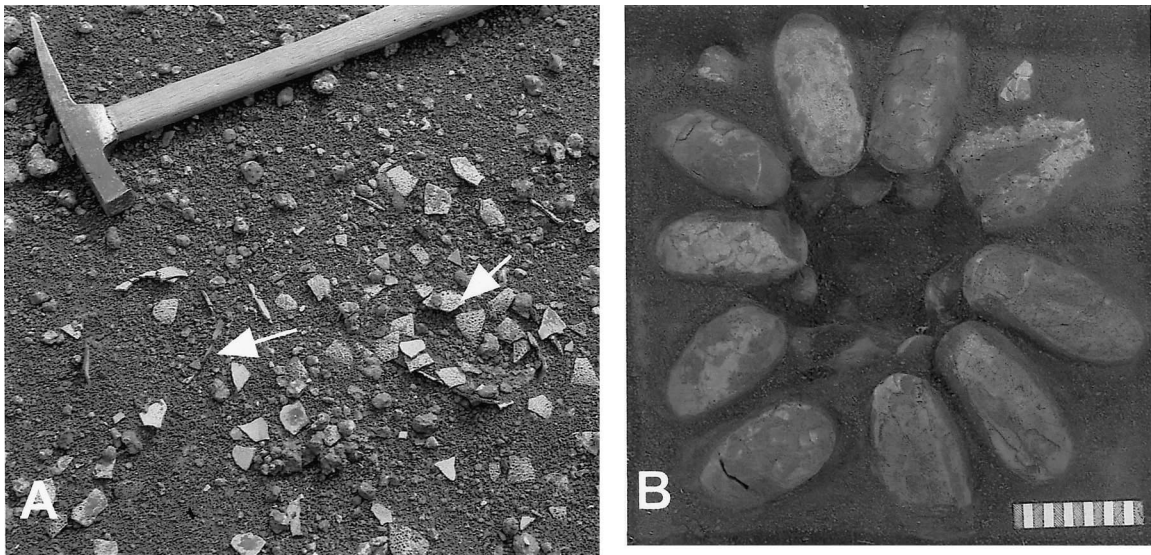


Fig. 3. Eggshell fragments and a complete clutch of eggs discovered in the Pingling Formation. (A) Eggshell fragments and two crushed eggs (arrows) of *Macrooolithus yaotunensis*, preserved in siltstone at the level of 82–87 m in the CGD Section. (B) A clutch of 18 eggs of *Macrooolithus yaotunensis* discovered below the contact between the Pingling and Shanghu formations, about 1 km to the east of Feng-Men-Ao Village.

tion (four eggshell samples from 162 to 182 m of the CGY plus 16 eggshell samples from 3 m up to 161 m of CGD, the contact between the Pingling Formation and the Shanghu Formation). This was supplemented by five samples collected at and near the palynological K/T boundary interval (one from CGD 119, three from CGD 111, one eggshell sample and one dinosaur bone fragment from CGD 109), and seven eggshell samples collected at five levels (CGY 207, 206, 203, 202 and 201) from 171 m down to 141 m of the CGY. Stratigraphic positions of these samples are shown in Fig. 2.

The results of Ir analyses on the eggshell samples from the CGY-CGD Section are shown in Table 1 and on the upper right side of Fig. 2. They give a clear picture of the trend of iridium concentrations as a function of stratigraphic level. Three distinct Ir anomalies occur in three levels (CGD 109, 111 and 119) at and near the palynological K/T boundary interval. Another three, in three levels (CGY 201, 202 and 206) at about 160

Table 1
Ir values in the eggshell samples from the CGY-CGD Section^a (ppt, 10⁻¹²g/g)

Sample	Depth (m)	Ir (ppt)
CGD 124	158–161	28.8 ± 3.7
CGD 119	109–110	
a		51.0 ± 8.8
b		35.6 ± 1.6
CGD 111	60–63 (boundary base)	
a		61.9 ± 13.3
b		118.0 ± 2.3
c		81.5 ± 2.4
d		48.6 ± 6.1
CGD 109	52–55	
a		49.5 ± 5.0
b		117.6 ± 2.1
c ^b		84.5 ± 6.5
CGY 206	168	50.8 ± 0.5
CGY 202	144–150	
a		45.5 ± 5.0
b		36.2 ± 1.1
CGY 201	141–144	
a		45.6 ± 7.2
b		50.8 ± 9.5

^a Ir values from all other levels are below 23.9 ppt and are not given here.

^b Ir value is from the sample of a broken bone (dinosaur?).

m below the boundary interval. Iridium abundance shows maximum at and near the palynological K/T boundary interval analyzed with varying strength, 118 ppt in CGD 111 and 117.6 ppt in CGD 109, and is approximately 19 times the background level (< 6 ppt). In between these six Ir peaks, and below and above the interval, no enhancements of Ir (detection limit is about < 6 ppt) were observed.

To test the possibility that iridium might be concentrated in eggshells from the Yuanpu Formation, we subsequently analyzed four eggshell samples collected at four levels (CGH 601, 602, 603 and 604) from 496 m up to 680 m of the CGH Section (see the lower part of Fig. 2). The Ir content is below the detection limit in CGH 601 and 602, and is 19.0 ± 1.9 ppt in CGH 603, and 17.3 ± 3.0 ppt in CGH 604, respectively, showing no evidence of iridium enhancement.

Twenty-four eggshell samples, belonging to *Macroolithus yaotunensis*, from the stratigraphic sequence of the CGY-CGD Section were used for analysis of other trace elements with the atomic absorption spectrometry. Although the elemental abundance data for the CGD have been earlier reported (Zhao et al., 1991), these and the unpublished data of the CGY are included and plotted in Fig. 4 because they provide a scale of reference for comparison of elemental distributions with other K/T boundary sections. The elements such as Ni, Co, Zn, Pb, K, Cu, Mn, Sr and V, show similar patterns of abundance variations, and seven of them reach a maximum level at and near the boundary interval.

3.2. The CGT-CGF Section

The CGT-CGF Section is located south of the Feng-Men-Ao village about 6 km to the southwest of the Nanxiong county seat (Fig. 1). Lithologically, the section was described in detail (Erben et al., 1995), and is similar to the CGY-CGD Section (Fig. 2). But the Pingling Formation here attains a thickness of about 600 m.

Twenty-two eggshell samples, belonging to *Macroolithus yaotunensis*, collected at 16 levels were taken from this section (10 from CGT and 12 from CGF), and parallelly analyzed at the Lab-

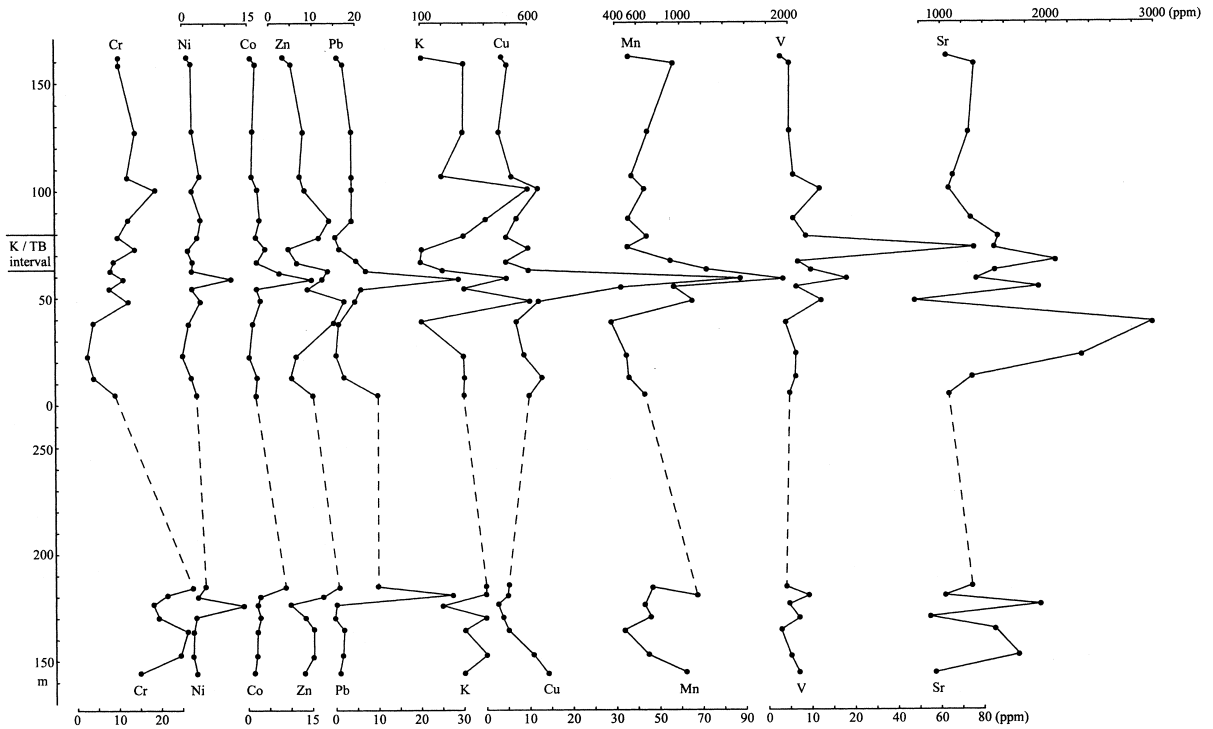


Fig. 4. Abundance variations of 10 trace elements contained in 24 eggshell samples, *Macroolithus yaotunensis*, from the CGY-CGD Section.

oratory of Tokyo Metropolitan University (TMU), Japan (Uchino et al., 1996), and at the Laboratory of Nuclear Analytical Techniques (LNAT) in Beijing, respectively. Two portions from each eggshell sample were irradiated with thermal neutrons. One portion was determined by RNAA for iridium abundance. The second portion was measured by instrumental neutron activation analysis (INAA) for other trace elements. The Ir results obtained in TMU and LNAT are in agreement within analytical error, which confirm the reliability of the data.

The iridium data from this section are presented here (Table 2 and the upper left side of Fig. 2). The Ir enrichment pattern observed in this section is similar to that of the CGY-CGD Section discussed above. Two Ir abundance anomalies occur in two levels (CGF 401 and 410) at and near the K/T boundary interval. Another four, in four levels (CGT 311, 310, 309 and

305) at about 200 m below the boundary interval. The Ca concentrations of the eggshells used in this investigation are similar to those of CGY-CGD Section (Zhao et al. 1998), varying from 34 to 39% (Fig. 5). The abundance rates of Ir/Ca in these levels are consistently high, indicating that the anomalous Ir concentrations still exist in these samples. The highest Ir abundance occurs just at the boundary interval (CGF 401), and reaches 168 ppt, being approximately 28 times higher than the background level.

The distribution of some trace elements in this section is shown in Fig. 5. It seems that the patterns of the elemental abundance variations are similar to those of the CGY-CGD Section discussed above. They also reach a maximum level at and near the palynological K/T boundary interval, excepting Co and Se whose maximum levels occur at about 200 m below the boundary interval.

Table 2
Ir values in the eggshell samples from the CGT-CGF Section^{a,b} (ppt, 10⁻¹²g/g)

Sample	Depth (m)	Ir (ppt)
CGF 415	150–153	
a		41.2 ± 1.0
b		20.1 ± 4.0
CGF 410	87–91	
a		80.6 ± 0.4
b		47.6 ± 14.9
CGF 401	63–66 (Boundary interval)	168.0 ± 0.3
CGT 311	349–350	
a		86.2 ± 6.5
b		156.3 ± 2.2
CGT 310	332–333	
a		74.3 ± 14.0
b		51.3 ± 7.6
CGT 309	300–310	
a		49.9 ± 1.9
b		40.6 ± 6.0
CGT 305	226–234	
a		78.5 ± 15.1
b		105.0 ± 5.1

^a Ir values from all other levels are below 30.2 ppt, and are not given here.

^b Ir abundance of these samples was determined at TUM and LNAT, respectively.

3.3. The processes leading to the enrichment of the Ir and other trace elements in the dinosaur eggshells

As has been shown above, multiple iridium anomalies have been observed in the two studied sections across the K/T boundary, respectively. Iridium enrichment, which occurred just at and near the palynological K/T boundary interval shows a maximum at the boundary interval in the two sections analyzed with varying strength, 118 ppt at the CGY-CGD Section, and 168 ppt at the CGT-CGF Section. The variation in the anomalous concentrations of other trace elements

in the eggshells from the two studied sections also reveals the similar tendency (Figs. 4 and 5), showing that they reach mostly a maximum level at and near the boundary interval. This coincidence is unlikely to be the result of diagenetic processes. The eggshell samples analyzed in this report have the same identical form, e.g. *Macroolithus yaotunensis*, and were collected from the same stratigraphic unit, the Pingling Formation. They were selected by micromorphological methods and have well-preserved microstructure with primary calcitic growth and little or no recrystallization. This observation also lends support to our assumption that diagenesis did not play a significant role in changing the elemental concentrations. Abundant Ir and other trace elements in the dinosaur eggshells should reflect changes of their original mineral composition before fossilization began. On the other hand, paleobiological studies of dinosaur eggshells reveal that anomalous variation in eggshell thickness (Fig. 6) and eggshell microstructure (Fig. 7) occur rather frequently in this stratigraphic unit. We observed the frequency of pathological eggshell belonging to the common species, *Macroolithus yaotunensis*, from the two studied sections of the Pingling Formation (see below). The frequency of the pathological eggshells in this species at and near the boundary interval increases to about 74% of the observed eggshells in the CGY-CGD Section (Table 3), and about 56% in the CGT-CGF Section (Table 4). This indicates that there is a relationship between high trace element concentrations and abnormal structure of the eggshell. The high levels of these elements in the eggshells may be responsible for producing pathological ones, as discussed later.

It is reasonable to assume that the enrichment of Ir and other trace elements in the eggshells may have been caused by the assimilation of these elements into the dinosaur body through food, per-

Table 3
Frequency of pathological eggshells in *Macroolithus yaotunensis* from successive horizons of CGY-CGD Section

Sampling interval (m)	Numbers of eggshell samples	Numbers of pathologic eggshells	Frequency (%)
CGD 110–161	59	21	36
CGD52–110	82	61	74
CGD 0–52	40	9	22
CGY 144–186	30	8	27

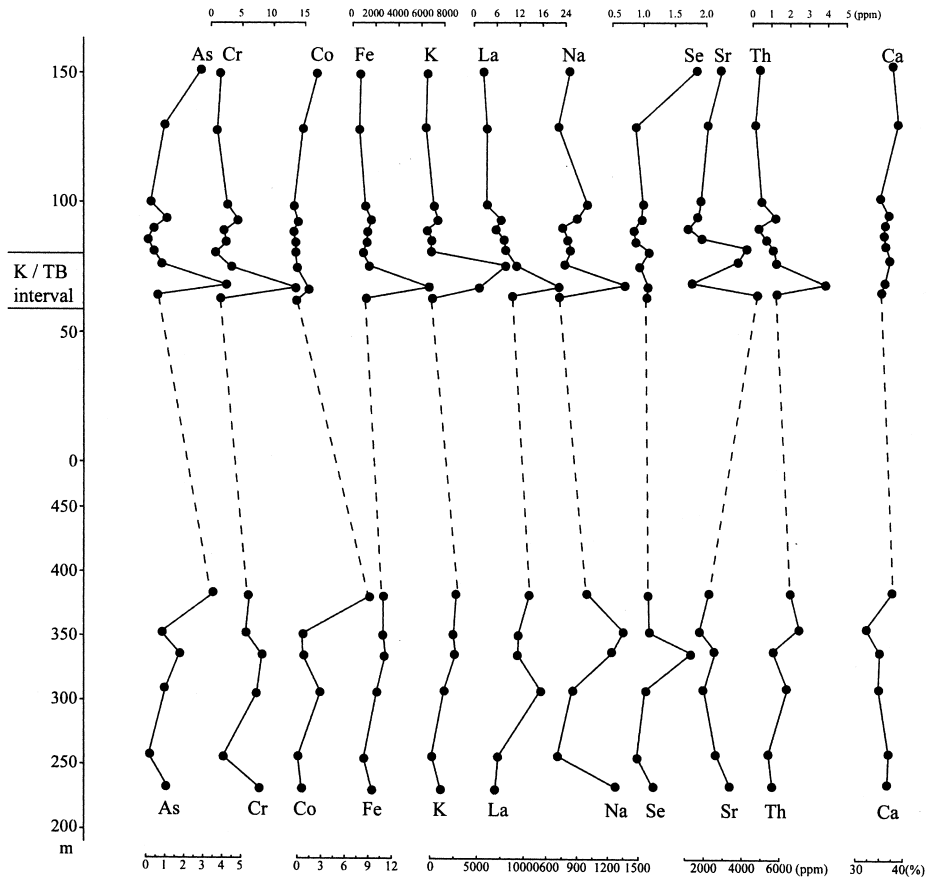


Fig. 5. Abundance variations of trace elements and a major element, Ca, contained in the eggshell samples belonging to *Macroolithus yaotunensis* from the CGT-CGF Section.

haps including drinking water and/or air, and then into the eggs laid by them. This assumption was confirmed by a feeding experiment with an Ir-tracer, which our research group has performed (Yang et al., 2001). Preliminary results indicate that Ir is deposited in the contents of egg and the eggshell. Several parts per ten thousands of Ir fed a flock of hens are deposited in the eggshells. According to the experimental data of Ro-

manoff and Romanoff (1949) and Mueller and Leach (1974), the radioactive isotope of Sr and P given to the hen can be deposited in the eggshell, and less than 0.6% of the radioactive P injected into a hen can be deposited in the eggshell. REE absorption by mammals usually is less than one per mil after feeding of them (Guo et al., 1988). This shows that trace elements are absorbed in the digestive tract in minute quantity

Table 4
Frequency of pathological eggshells in *Macroolithus yaotunensis* from successive horizons of CGT-CGF Section

Sampling interval (m)	Numbers of eggshell sample	Numbers of pathological eggshell	Frequency (%)
CGF 126–153	30	11	37
CGF63–100	50	28	56
CGT 250–390	30	8	27
CGT 111–234	30	9	30

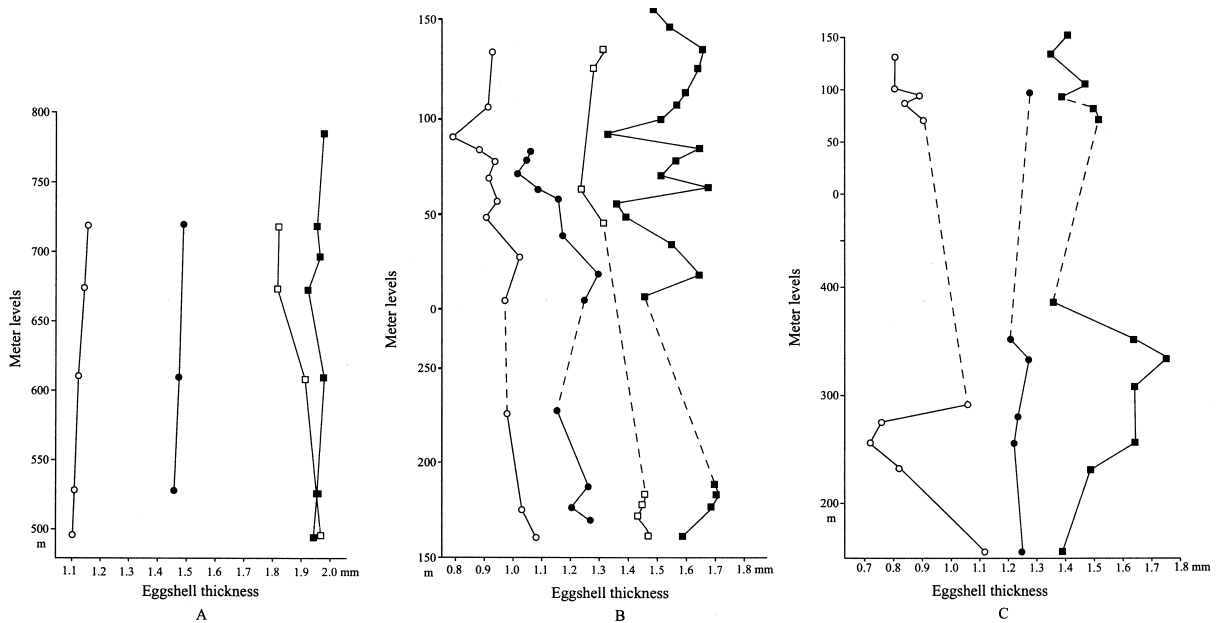


Fig. 6. Diagram showing variations in average values of eggshell thickness of the four species from the successive horizons of the three studied sections ○ *Elongatoolithus elongatus*; ● *Elongatoolithus andrewsi*; □ *Macroolithus rugustus*; ■ *Macroolithus yaotunensis*. (A) CGH Section; (B) CGY-CGD Section; (C) CGT-CGF Section. The mean eggshell thickness of *Macroolithus rugustus* is not presented in this section because of insufficient samples for statistical evaluation.

only. It seems logical to estimate, therefore, that in the case of dinosaurs the intake of Ir and other trace elements through food could be 100–1000 times higher than the amount deposited in the eggshell. If this is so, the Ir concentration in the geological environment during the transition time in the Nanxiong Basin must reach ppb level, such as that documented for various K/T boundary over the earth (Alvarez et al., 1980; Alvarez and Asaro, 1990). Therefore, the most reasonable explanation is that the anomalous concentrations of Ir and other trace elements contained in the eggshells were introduced from an external source, most probably from fall-out aerosols of either extraterrestrial or volcanic origin, and were not derived from crustal rocks on the Earth's surface. The high concentrations were taken into the dinosaurian body through food, and then into eggs laid by them, as mentioned above. This process would affect the normal physiological function of dinosaur body, and cause pathologic variation in eggshell thickness and microstructural alteration of the eggshell.

The distribution of Ir anomalies argues for the existence of at least six Ir-delivering events from the latest Cretaceous into the earliest Paleocene. This means that there may have been great environmental change marked by these Ir-bearing horizons. Recently, general trends in the carbon and oxygen isotopic stratigraphy were established based on a series of dinosaur eggshell samples in this basin (Zhao and Yan, 2000). The oxygen isotope ratios show multiple positive excursions during the K/T transition time, indicating the existence of unusual climatic conditions. The trend toward more negative carbon isotopic values of the eggshells is regarded as an indirect consequence of a lasting increase in atmospheric CO₂ concentration taking place at that time. The above-mentioned various evidences imply that in the Nanxiong Basin the K/T event was not marked by an instantaneous geochemical environmental change, but stretched out over a considerable time. Such a period apparently favors a volcanic source, and seems to be comparable to the duration of the Deccan volcanism in India (Offi-

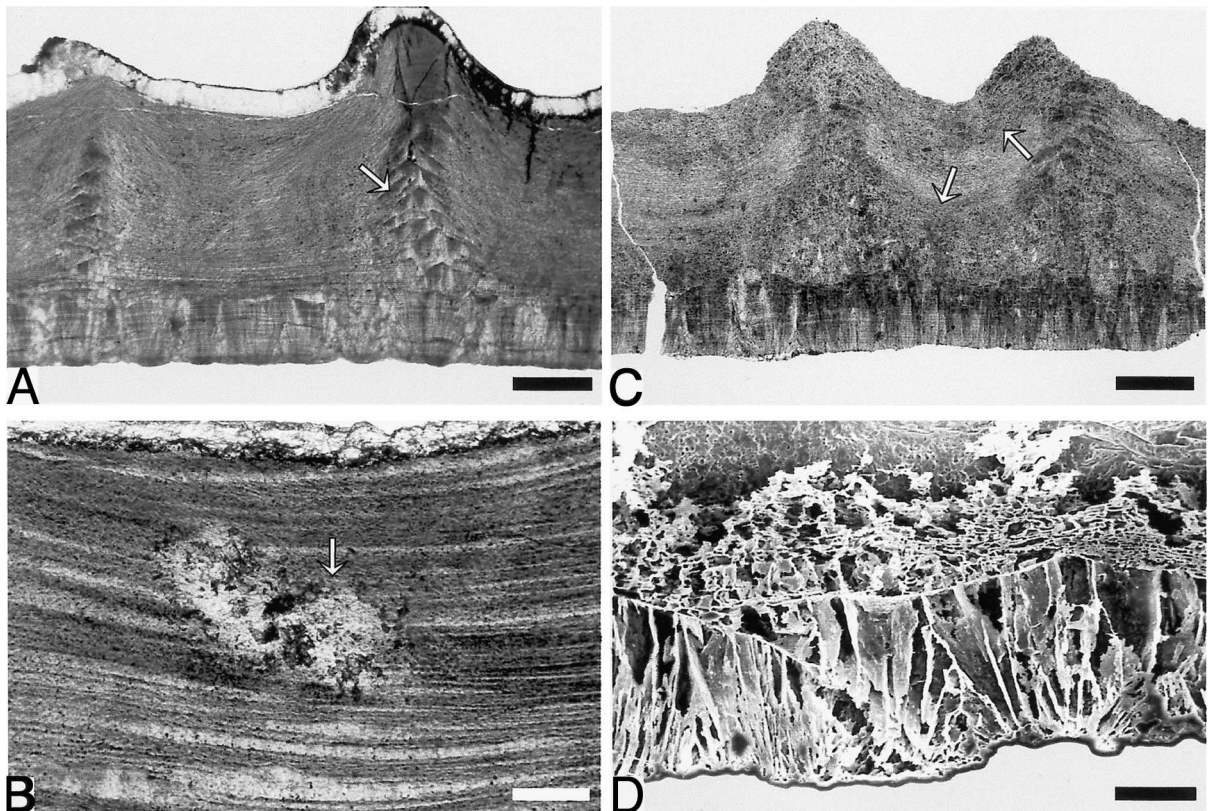


Fig. 7. Radial section of pathologic eggshells from the studied K/T sections. (A) *Macroolithus rugustus*, (CGD 108). Note multi-layered cone in the columnar layer (arrow). Bar = 300µm. (B) *Elongatoolithus elongatus*, (CGF 138). Note an irregular space in the columnar layer (arrow). Bar = 200µm. (C) *Macroolithus yaotunensis*, (CGD 119). The eggshell with poor quality biocrystallization; note two zones of disorderly arranged crystallines in the columnar layer (arrow). Bar = 300µm. (D) *Macroolithus yaotunensis*, (CGD 112). Note the cone layer consisting of defective formation of cones and organic matter preserved in the lower part of the columnar layer. Bar = 100µm.

cer et al., 1987; Courtillot et al., 1988), but incompatible with a sudden asteroid impact. It seems extremely improbable that one would need to invoke successive impacts of perhaps six extraterrestrial bodies of necessary mass over a period of a million years to account for the iridium peaks now recognized in the Nanxiong Basin.

K/T iridium was the basis for the impact theory (Alvarez et al., 1980). However, multiple iridium spikes have been found in some K/T boundary sections (Ganapathy et al., 1981; Donovan et al., 1988; Graup and Spettel, 1989; Bhandari et al., 1995, 1996). Nearly all deep-sea K/T boundary sequences presented by MacLeod and Keller (1991a,b) have intervals of non-deposition from

the latest Cretaceous through the earliest Tertiary. They concluded that single-peak Ir anomaly, and apparently instantaneous mass extinctions of marine plankton, which are routinely reported from these K/T boundary sequences, may be artifacts of a temporally incomplete deep-sea stratigraphic record. Bajpai and Prasad (2000) reported that the extinction of dinosaurs in the Indian subcontinent occurred after the deposition of Ir layers at Anjar, and that these Ir anomalies may significantly predate the K/T boundary. Therefore, the discovery of multiple Ir abundance in dinosaur eggshells in the Nanxiong Basin further demonstrates that the geochemical environmental changes and the dinosaur extinction may have

been a rather long-duration process and not an instantaneous event, lasting a few years or centuries as is often portrayed.

4. Pathologic dinosaur eggshells from the Pingling Formation

It is remarkable that among the different species of dinosaur eggshells found in the Nanxiong Basin, many of them exhibit abnormalities. Two types of pathological development were sketchily discussed in previous reports (Zhao, 1978, 1994; Zhao et al., 1991). One is an anomalous variation in eggshell thickness, and the other affects shell structure. But it is not well documented that this concerns the structural characteristics of the abnormal eggshells and the stratigraphic occurrence.

4.1. Abnormal changes in eggshell thickness

The variation in eggshell thickness has been studied in the CGH Section of the Yuanpu Formation and in the CGY-CGD and the CGT-CGF sections of the Pingling Formation. Among 12 species known, however, only the eggshell samples of four species: *Macroolithus yaotunensis*, *Macroolithus rugustus*, *Elongatoolithus andrewsi* and *Elongatoolithus elongatus* are numerous enough for statistically significant evaluations. Measurements of the shell thickness involve about 8258 eggshell fragments of the four species from the stratigraphic sequences of the three sections mentioned above.

In the CGH Section of the Yuanpu Formation the eggshells of each species are relatively thicker. The range of variation in the eggshell thickness of each species has been obtained. *Macroolithus yaotunensis* is between 1.70 mm and 2.52 mm, *Macroolithus rugustus*, between 1.66 mm and 2.38 mm, *Elongatoolithus andrewsi*, between 1.36 mm and 1.58 mm, and *Elongatoolithus elongatus*, between 0.9 mm and 1.26 mm. Obviously, each species has a small variation in the eggshell thickness.

In both the CGY-CGD and the CGT-CGF sections of the Pingling Formation the eggshells of each species are more variable in thickness. *Macroolithus yaotunensis* ranges from 0.9 mm to 2.38

mm, *Macroolithus rugustus*, from 0.66 mm to 2.48 mm, *Elongatoolithus andrewsi*, from 0.64 mm to 1.50 mm, and *Elongatoolithus elongatus*, from 0.5 mm to 1.20 mm.

The data of the eggshell thickness are plotted on a scale in order to show the frequency of mean shell thickness in each of the four species from successive level of the three sections, respectively (Fig. 6). In the CGH Section of the Yuanpu Formation the average values of shell thickness in each species at various levels show almost no change, and represent the normal range of predominating variation (Fig. 6A). In both the CGY-CGD and the CGT-CGF sections of the Pingling Formation, however, the average values of shell thickness in each species distinctly vary (Fig. 6B,C).

4.2. Histopathological pattern of the eggshells

The use of modern analytical methods (PLM, SEM, elemental analysis) can help in recognizing and identifying pathological eggshells. Based on the microphotograph of the radial thin sections of eggshells, various histopathological patterns are recognized.

Eggshells with a bi- or multi-layered cone: Zhao described and discussed this local abnormal structure of the eggshell belonging to *Macroolithus yaotunensis* (Zhao, 1994: fig. 12.15). A secondary cone layer or a multi-layered cone is formed between the original cone layer and the columnar layer. Fig. 7A shows the pathological eggshell with multi-layered cones in the columnar layer in the *Macroolithus rugustus*. The overlapping secondary layered cones vary more or less in their structure, and are arranged in a single column. This pathological development is different from the teratological cases called 'ovum in ovo' in living birds (Romanoff and Romanoff, 1949) and 'multi-layered eggshells' in living and fossil turtles and dinosaurs (Erben, 1972; Erben et al., 1979; Ewert et al., 1984; Mohabey, 1984; Hirsch, 1989; Hirsch et al., 1989).

The eggshell with a bi- or multi-layered cone was apparently not caused by the antiperistaltic movements of the oviduct or by the retention of eggs beyond the time of normal oviposition. In-

stead, the formation of this pathological eggshell may have been correlated with changes in the protein profiles of the organic matrix in the calcareous eggshell (see below).

Eggshells with irregular space: this abnormal structure of the eggshell from *Elongatoolithus elongatus* is shown in Fig. 7B. An irregular space exists in the columnar layer. It may be due to formation of hollow 'globular inclusion' during the eggshell calcification (see below).

Eggshells containing biocrystals with amorphous structures and irregular orientations: Under normal conditions, biocrystallite calcite develops outwards from the interior surface of the shell (Sharp and Silyn-Roberts, 1984; Silyn-Roberts and Sharp, 1985) and these crystallites have well-defined angles and a vertical orientation (Heyn, 1963; Beeking, 1975). Fig. 7C displays the eggshell with poor quality biocrystallization from *Macroolithus yaotunensis*. It shows two zones of disorderly arranged crystallines in the columnar layer of the eggshell. Another case shows the contour of the layered cone and the junction between the cunei (Fig. 7D). The whitish network in lower part of the columnar layer represents organic matter preserved. This indicated that the synthesis of the organic matter and calcium carbonate deposition could be affected during the formation of the eggshell in the oviduct.

The above-described pathological dinosaur eggshells are not frequent in the Yuanpu Formation. In a random sample of 30 eggshell fragments belonging to the *Macroolithus yaotunensis* from 605 to 790 m of the CGH Section, we found only two eggshell fragments with abnormal microstructure, and the frequency is approximately 7% of the observed eggshells. However, they occur most frequently in the Pingling Formation, especially at and near the palynological K/T boundary interval.

In the CGY-CGD Section (Table 3), the frequency of pathological eggshells of this species accounts for about 27% of the observed eggshells from 144 to 186 m of the CGY Section, and about 22% from 0 to 52 m of the CGD Section. From 52 up to 110 m of the CGD Section (i.e. at and near the palynological K/T boundary interval) their frequency increases to about 74%, and

from above 110 m to 161 m marked by the disappearance of the dinosaur eggshells it is about 36% of the eggshells.

In the CGT-CGF Section (Table 4), the frequency of the pathological eggshells in this species is similar to that of the CGY-CGD Section, mentioned above. It is represented at 111–234 m of CGT Section by 30% of the observed eggshells and at 250–390 m by 27%. However, at 63–100 m of the CGF Section (i.e. at and near the K/T boundary interval) the frequency of the pathological eggshells increases to 56% and at 126–153 m marked by last disappearance of the dinosaur eggshells it decreases to 37%.

4.3. Physiological mechanisms producing pathological eggshells

As has been shown above, both pathological tendencies occur frequently in different species of the dinosaur eggshells in both the CGY-CGD and the CGT-CGF sections of the Pingling Formation. Since the normal eggs in the CGH Section of Yuanpu Formation are stratigraphically older, it is evident that the last dinosaur populations represented by these different species of eggshells, i.e. the ones from the Pingling Formation, laid abnormal eggs. This seems to have coincided with the distribution of anomalous concentrations of Ir and other trace elements in this formation.

Pathological eggshells in living animals are caused by several environmental factors: i.e. pollution by certain insecticides, pesticides and heavy metals or trace elements (Mueller and Leach, 1974; Erben, 1972; Erben et al., 1979; Gonzalez and Hiroldo, 1988), and stress due to temperature and humidity (El-Boushy et al., 1968). The mechanism of this process has been identified in the case of abnormally thin eggshells in many birds of prey formed under the influence of organochlorine pollutants (Bitman et al., 1969; Cooke, 1973, 1979; Peakall et al., 1973; Blus, 1982). The eggshells of the Spanish Imperial Eagle [*Aquila (heliaca) adalberti*] collected from 1972 to 1984 in Spain not only contain small amounts of heavy metals (Hg, Pb, Cd, Zn, Cu) and organochlorine contaminants, but also have a significant decrease in shell thickness in conjunction with poor quality

biocrystallization (Gonzalez and Hiroldo, 1988). Investigation on the eggshells of the raptor, *Falco tinnunculus*, indicated that a correlation exists between the shell thickness and high level of lead in the contents (Grandjean, 1976). On the other hand, deficiency or excess of some trace elements, such as Mn, Zn, Sr, etc., may greatly affect the synthesis of the eggshell matrix and calcium carbonate deposition, thus causing the formation of the abnormal eggshell (Mueller and Leach, 1974). It has been observed that the notable changes in the eggshell structure were the presence of hollow 'globular inclusion', the disorder in the amino acid ratios of the organic shell matrix (Erben and Krampitz, 1971), and malformation of the mammillary knobs (McFarland et al., 1971).

Although the overall mechanism of eggshell formation is not fully understood on a molecular basis, it is quite clear that the organic matrix plays a key role during eggshell calcification. There is some evidence that the protein profiles in an avian eggshell can be influenced by environmental toxins, such as Cd, Pb, Hg, Be, etc., and changes in the protein profile may in turn lead to a pathological formation of eggshell (Krampitz, 1982; Krampitz and Witt, 1979).

In view of the comparable bird and dinosaur eggshell structure (Zhao, 1994) and the similar amino acid composition of the organic shell matrix (Zhao et al., 1993), it is inconceivable that the mechanisms and processes of the eggshell formation in the dinosaurs should have been different from those observed in extant birds. Under these circumstances, it is justified to assume that the abnormal changes in the eggshell thickness and the microstructure alteration of the dinosaur eggshells in the study were brought about by the same mechanisms as summarized above. It seems that the formation of pathologic dinosaur eggshells was caused by the presence of trace elements.

In both types of pathologic eggshell, the consequences for the embryo are lethal. These eggshells became so brittle that they failed to protect the developing embryos.

It is also well known that trace elemental composition of the egg is important in its effect on normal embryonic development. The hatchability

of eggs may be greatly impaired by a deficiency of some elements, or by an excess of toxic elements transmitted to the egg from a bird's diet. The presence of selenium in the egg causes abnormal embryonic development and thus considerably reduces hatchability (Romanoff and Romanoff, 1949). It is possible, therefore, that excess of trace elements transmitted to egg from the dinosaur's food causes abnormal embryonic development and considerably reduces hatchability.

5. Dinosaur egg fauna and sequential nature of K/T boundary extinction

The dinosaur eggshells found in the Nanxiong Basin have been classified into over 12 species allocated to 4 families and 8 genera (Zhao, 1975, 1994; Zhao et al., 1991; Zhao et al., 1999). At the present stage of our knowledge they are considered as an egg fauna. It appears that this egg fauna is dominated in a number of specimens by two genera, *Macroolithus* and *Elongatoolithus*.

As shown above, the CGY-CGD Section represents a transitional zone covering approximately 20 m. The level of marked palynological changes within this zone is used as the K/T chronostratigraphic boundary in this report. This indicates that the dinosaur eggshells and dinosaur remains from the upper part of the Pingling Formation, corresponding to the horizons above 78 m, are early Paleocene rather than late Cretaceous in age. Therefore, the earlier statements (Zhao, 1978, 1994; Zhao et al., 1991; Zhao et al., 1993) on the extinction of the dinosaur population in the Nanxiong Basin require significant revision.

We have re-examined the dinosaur eggshell fauna at both sections (i.e. the CGY-CGD and the CGT-CGF sections), and the range chart is reproduced in Fig. 8. We note that of 11 species of the dinosaur eggshell in the Upper Cretaceous at the CGY-CGD Section, only six rare species, identified as *Aphelolithus shuinanensis*, *Stromatoolithus pinglingensis*, *Prismatoolithus hukouensis*, *Ovalolithus* cf. *O. laminadermus*, *Ovalolithus* cf. *O. chinkangkouensis*, and *Nanshiugoolithus chuetiensis*, became extinct exactly at the K/T bound-

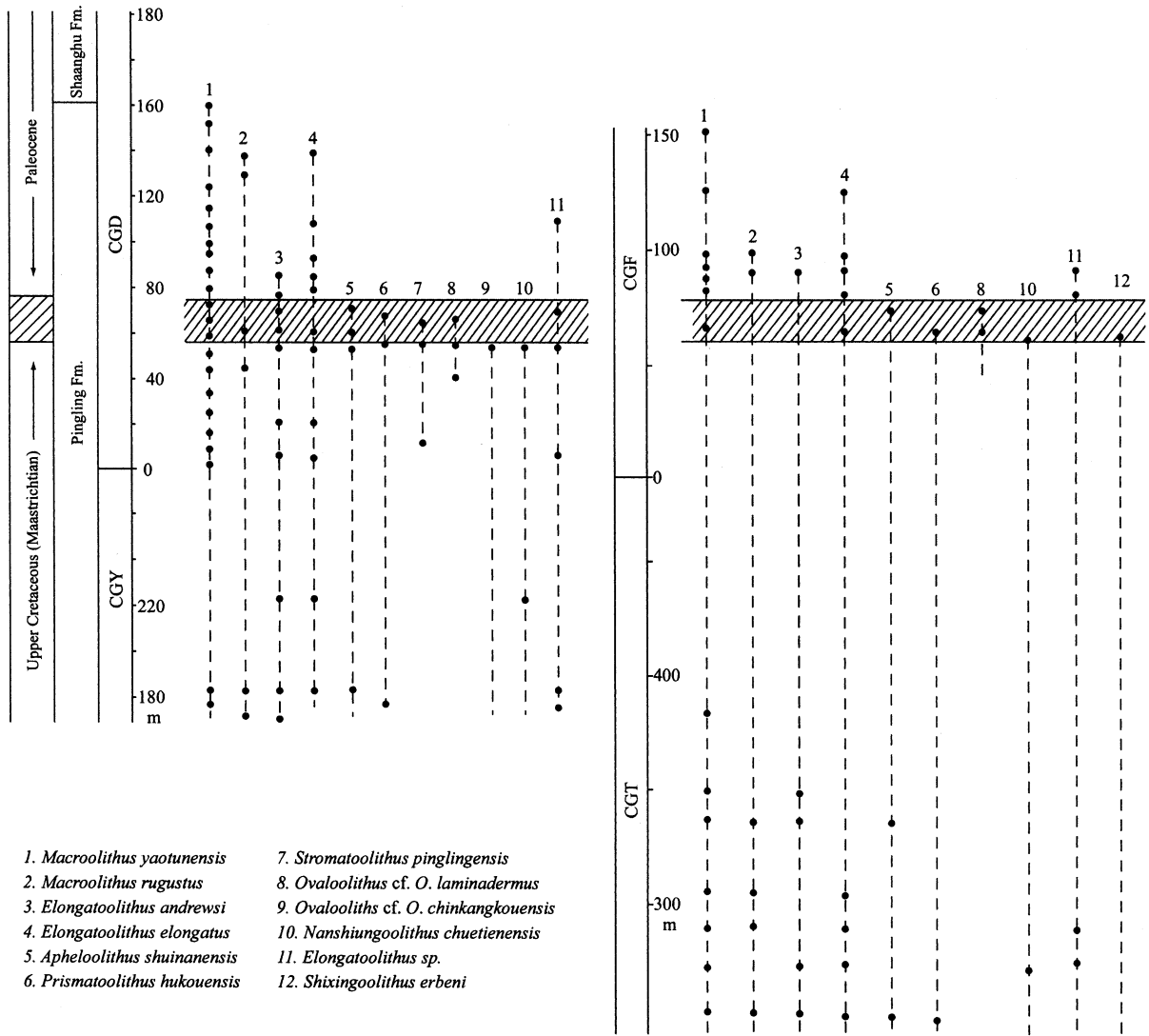


Fig. 8. Diagram showing stratigraphic occurrence of different species of dinosaur eggshell in the studied K/T sections.

ary interval. The remaining five species overstepped the boundary interval and survived into the early Paleocene, indicating one species disappearing after another over an interval of about 100 m. Only one species, *Macroolithus yaotunensis*, ranged up to the contact between the Pingling Formation and the Shanghu Formation.

The stratigraphic record of dinosaur eggshells in the CGT-CGF Section is quite similar to that of the CGY-CGD Section mentioned above. Of the 10 species of the dinosaur eggshells found

in the Upper Cretaceous, only five rare species, identified as *Apheloolithus shuinanensis*, *Nanhsiungoolithus chuetienensis*, *Ovaloolithus* cf. *O. Laminadermus*, *Prismatoolithus hukouensis*, and *Shixingoolithus erbeni* disappeared at the K/T boundary interval. The remaining five species also survived into the early Paleocene. The last species, *Macroolithus yaotunensis*, ranged up to the contact between the Pingling Formation and the Shanghu Formation, where it became extinct. The graphic correlation plot based on last appearances of spe-

cies at both sections indicates that not all species became extinct simultaneously, but spread out within 100 m of sediment with major extinction beginning at the boundary interval. This corresponds to a time span of about 250 ka, assuming the mean deposition rate of 40 cm/ka, as mentioned above.

Acknowledgements

The funding for this research was supported by the National Natural Science Foundation of China (NNSFC, Contract no. 19935020), Chinese Academy of Sciences (CAS, Contract no. 21039751) and the Laboratory of Nuclear Analytical Techniques, CAS. Zhao is particularly indebted to Ms. Y.X. Duan, who made the thin sections of eggshells and the measurements of eggshell thickness used in this study. The authors would like to thank Drs. A.R. Ashraf, G. Keller, K. Carpenter and J.D. Archibald for many helpful comments and suggestions on early drafts of the manuscript. We are also indebted to Drs. A. Hallam and M. Vianey-Liaud for critical comments and helpful suggestions on this manuscript.

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