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Discussion

# Reply to comments by Sanjay K. Mukhopadhyay, Sucharita Pal, J. P. Shrivastava on the paper by Sial et al. (2016) Mercury enrichments and Hg isotopes in Cretaceous–Paleogene boundary successions: Links to volcanism and palaeoenvironmental impacts. Cretaceous Research 66, 60–81

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ABSTRACT

One key to address the issue of the location of the K/Pg boundary at the Um Sohryngkew sections to look at $C_{carb}$, $C_{org}$, Hg/TOC, Mo/Al chemostratigraphies, alongside Hg isotopes, in previous studied K/Pg sections in the Meghalaya area, and compare them with same type of data reported in Sial et al. (2016). The possibility of diachronic deposition cannot be totally discarded, and the K/Pg boundary could have been recorded in the Langpar Formation at one site and in the contact between the Mahadeo and Langpar formations in another. A tectonic-induced doubling of the sequence is also possible as the Meghalaya area, within the Shillong plateau, has been linked to a number of kinematic changes, including pop-up tectonics, since the Cretaceous.



We thank Mukhopadhyay and colleagues (Mukhopadhyay et al., 2017) for their keen interest on our chemostratigraphic and isotopic studies on some of well-known sections across the Cretaceous–Paleogene boundary (K/Pg). We are also grateful for the opportunity to clarify some points of our original manuscript (Sial et al., 2016) and discuss issues raised in their Comments.

Volcanic activities of Large Igneous Provinces (LIPs) are coeval with four of the major mass extinctions during the Phanerozoic (e.g., Percival et al., 2015 and references therein). In several recent studies, mercury (Hg) has been regarded as a potential indicator of massive volcanism, allowing for a new insight into the relationship between LIP activity, abrupt environmental changes and mass extinctions (e.g., Sial et al., 2010, 2013, 2014, 2016; Nascimento-Silva et al., 2011, 2013; Sanei et al., 2012; Adatte et al., 2015; Grasby et al., 2015, 2017; Percival et al., 2015; Font et al., 2016; Jones et al., 2016; Thibodeau et al., 2016; Charbonnier et al., 2017; Thibodeau

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and Bergquist, 2017). It has also been speculated that some of the observed Hg enrichments across some chronostratigraphical boundaries may represent true Hg loading to the environment, recorded by an increase in the Hg/TOC ratio (e.g., Grasby et al., 2013; Percival et al., 2015; references therein).

Following this line of reasoning, we have sought for Hg/TOC spikes across the K/Pg boundary in six sections, three of which regarded as continuous ones as demonstrated by planktic foraminiferal biostratigraphy: (a) Højerup, Stevns Klint, Denmark; (b) Bottaccione, Gubbio, Italy, both distal in relation to the Deccan volcanic center, and (c) the proximal Um Sohryngkew section, Meghalaya, India (Sial et al., 2016). The presence of three Hg/TOC spikes across the K/Pg boundary in the latter three sections, possibly associated to the Deccan phase-2 volcanism, became evident. Furthermore, we have analyzed Hg isotopes in these Hg spikes to test a possible geogenic source for Hg.

The major objection raised by Mukhopadhyay et al. (2017) is the possibility of an incorrect location of the Um Sohryngkew K/Pg boundary, in our study, placed in the contact between the Mahadeo and Langpar formations. We have carefully taken into account the pertinent geological literature regarding all of the K/Pg sections discussed in our study. The Meghalaya area was no exception and we have consulted, among others, the results of biostratigraphical and geochemical studies published by Mukhopadhyay and colleagues in the last ten years (Mukhopadhyay, 2008, 2012a, 2012b; Srivastava et al., 2013; Pal et al., 2015a, 2015b, 2015c).

Neither Mukhopadhyay (2008) nor Gertsch et al. (2011) have provided a precise location of their studied Um Sohryngkew K/Pg sections. Therefore, we could not locate them for our chemostratigraphic and isotopic study in which samples were collected from a section located at 25°11′ 18″N and 91°45′ 54″ W (Fig. 1). The location of the K/Pg boundary at the Um Sohryngkew section, based on foraminiferal and calcareous nannofossil biostratigraphy and geochemical investigations, has remained contentious for a long time (e.g., Bhandari et al., 1987, 1994; Lahiri et al., 1988; Pandey, 1990; Garg and Jain, 1995; Jauhri and Agarwal, 2001; Keller, 2001; Garg et al., 2006; Mukhopadhyay, 2008, 2012a, 2012b; Tewari et al., 2010a, 2010b; Gertsch et al., 2011; Srivastava et al., 2013; Pal et al., 2015a, 2015b, 2015c and references therein).

Below, we have addressed the raised issues in the Comments, in an itemized way:

1. Apparently, there are divergent opinions about ascribing particular sedimentary successions to the Mahadeo or Langpar formations in the Meghalaya area and, in consequence, some studies seem to have avoided the use of such formation names (e.g., Gertsch et al., 2011). We have indicated to a possible coincidence between the K/Pg boundary and the contact between the Mahadeo and Langpar formations in the section we studied, but we understand that, perhaps, the use of formation names in fig. 10 and elsewhere in our paper should have been wisely avoided, preventing further conflict outside the scope of the paper.

Sial et al. (2016) have sampled a succession within the Meghalaya area in which the K/Pg boundary location was defined on the basis of the fossil content, including foraminifera, dinoflagellates and ammonoids (e.g., Tewari et al., 2010a). In the chosen section, several fossiliferous beds with gastropods, ammonoids, echinoids and foraminiferal limestone are observed (Pandey, 1990; Tewari et al., 2010a: fig. 7; see also Figs. 2, 3). The biostratigraphic zonation of the Maastrichtian Mahadeo Formation includes benthic foraminifera *Dorothia oxycona* Zone from greenish gray shales followed by the planktic *Globotruncanita stuartiformis* Zone from light to gray shales (Pandey, 1990; Tewari et al., 2010a: fig. 2). These light- to medium-gray Maastrichtian shales are overlain by the Danian Langpar Formation, and the K/Pg boundary lies within this shaly facies. The base of the Danian Langpar Formation is marked

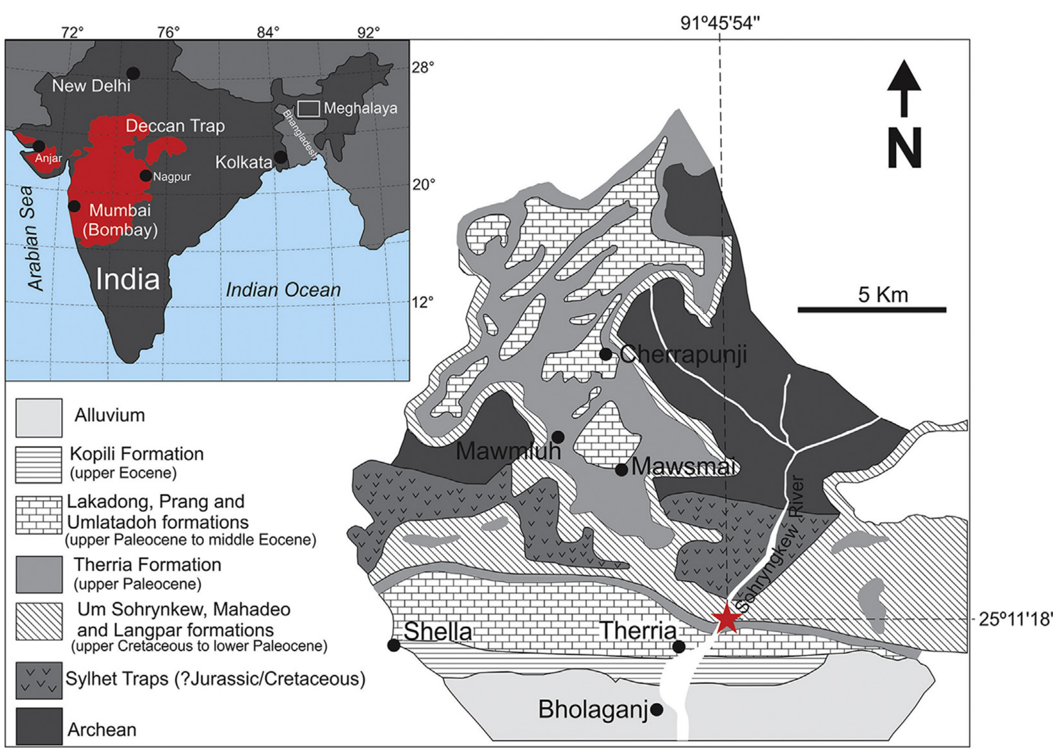


**Fig. 1.** Geological map of South Shillong Plateau, Meghalaya, northeastern India (modified from Tewari et al., 2010a; Sial et al., 2016), showing locality (red star) and geographic coordinates of the sampled section. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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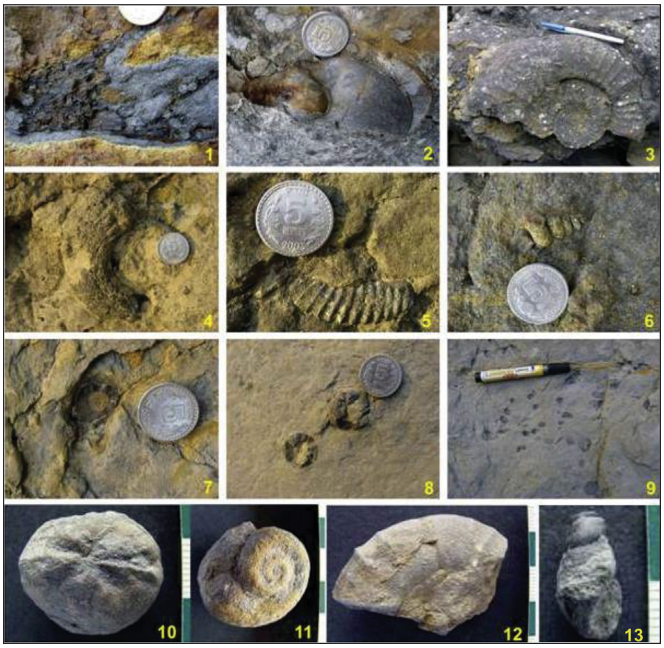

**Fig. 2.** Fossils from the Maastrichtian Mahadeo Formation (1–6, 11–12) and the Danian Langpar Formation (7–10, 13), Um Sohryngkew River section. 1, burned wood in sandstone; 2, nautiloid; 3–5 and 12, large coiled and uncoiled ammonoids; 6, 11 and 13, gastropods; 7–8, cross sections of burrows; 9, holes left by tiny gastropods; 10, echinoid.

by the planktic foraminifera *Parvularugoglobigerina eugubina* Zone followed by the *Parasubbotina pseudobulloides* Zone (Tewari et al., 2010a: fig. 2). The studied samples number KT1 through KT25 were collected from light-gray and dark-gray shales across the K/Pg boundary in this profile aiming at establishing $C_{carb}$, $C_{org}$, TOC, Mo/Al and Hg/TOC chemostratigraphies, besides Hg isotope determination. We are not sure about the exact location of the section in which Mukhopadhyay (2008) recorded the K/Pg boundary in the Langpar Formation.

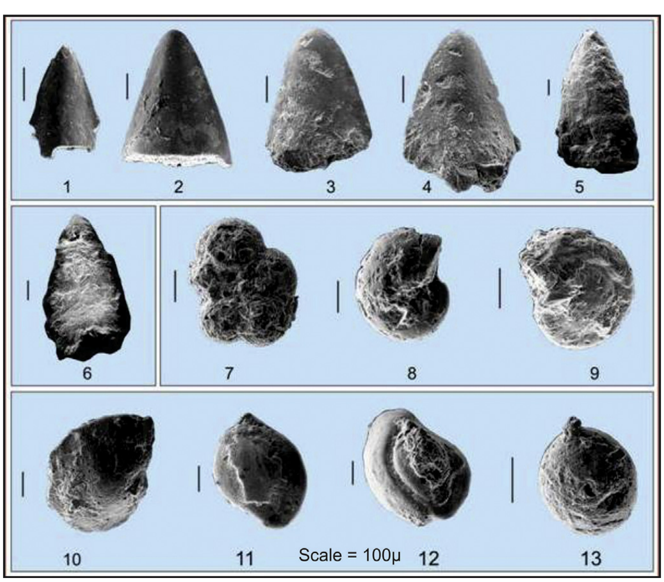


**Fig. 3.** Danian/Paleocene microfossils from the Langpar Formation, 1–6, unidentified fish teeth, possibly representing enchodontids; 7, *Parvularugoglobigerina* (?); 8–9, *Gyroidinoides girardanus* (umbilical and spiral views); 10, *Lenticulina* sp.; 11, *Quinqueloculina* sp.; 12, (*Spiroloculina* sp.; 13, *Lagena* sp. (Tewari et al., 2010a: fig. 7) reproduced with permission of the editor of JIGC, India).

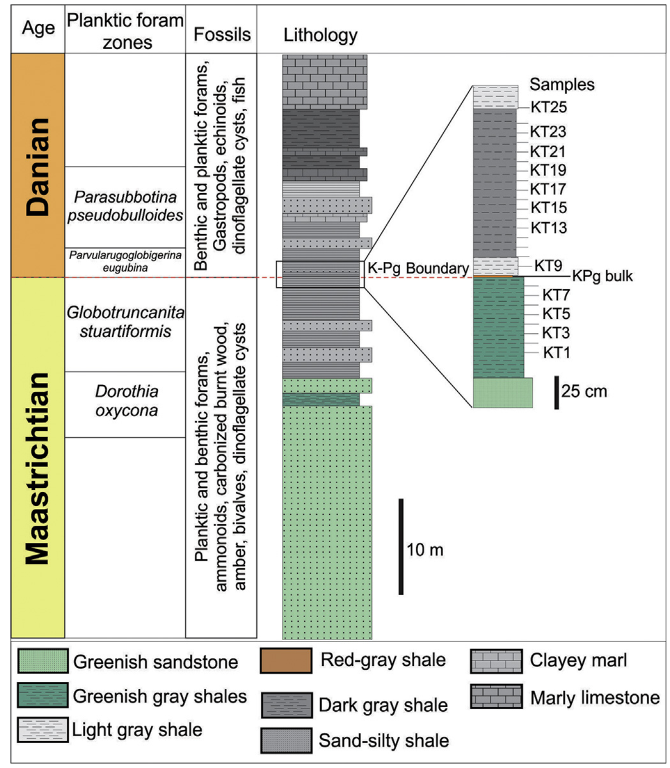


**Fig. 4.** Detailed litholog of the section across the K/Pg boundary in the western flank of the Um Sohryngkew River section, Meghalaya (Pandey, 1990; Tewari et al., 2010a) from where the samples were collected for C and Hg elemental and isotope analyses (Sial et al., 2016). The biozonation is based on micro-, nanno- and megafossils recorded from this section by different authors.

2. The outcrop shown in fig. 5a in Sial et al. (2016) is exposed few meters south of the Therriaghat Gorge, and displays mainly greenish sandstone and thin shale beds overlain by predominantly light-to medium-gray shales and dark shales. The litholog and fossils from this K/Pg section are shown in Fig. 4. Illustrations of fossils from this section are found in Figs. 2 and 3, comprising ammonoids, gastropods, burned fossil wood and echinoids from the Maastrichtian Mahadeo Formation, and microfossils, including foraminifera and fish microremains from the Danian Langpar Formation (Tewari et al., 2010a: fig. 7). The field photo in the Mukhopadhyay et al. (2017) comment displays only a portion of the Langpar Formation at about 250 m south of Therria Gorge. The shale portion shown is highly disturbed, discontinuous and has been tectonically displaced. The location of the K/Pg boundary has been marked by a yellow oblique line in relation to the bedding at this outcrop, on a photo of rather poor resolution that provides little help for any attempt of correlation with the Langpar Formation, represented by continuous and thinly-bedded shale in this region.
3. Multiple iridium enrichment layers, platinum group elements (PGE) and micro-spherules have been identified at the Um Sohryngkew section (Bhandari et al., 1987; Pandey, 1990; Bhandari et al., 1994; Mukhopadhyay, 2008, 2012a, 2012b). The position of the K/Pg boundary has been accordingly placed at different levels between the Maastrichtian Mahadeo Formation and the Danian Langpar Formation. The location of this boundary in Sial et al. (2016) lies within Unit III and lower

portion of Unit IV of Mukhopadhyay (2008). The portion of our section corresponding to Unit III, the lower portion of his Unit IV and reddish brown clay layer at the K/Pg boundary of Mukhopadhyay (2008) have been considered in our study as part of the Mahadeo Formation (sample numbers KT1 through KT8), while samples from light to dark-gray shales (KT9 through KT25, across the K/Pg boundary) have been regarded as part of the Langpar Formation. The original table grid in the table 3 of Sial et al. (2016) was removed during printing, and the name "Langpar Formation" has been displaced upward generating, unintentionally, further confusion. We regret for not having checked on this in due time.

Mukhopadhyay et al. (2017) comment mentioned that the Deccan phase-2 volcanism ranged from 65.4 to 65.2 Ma, based on studies by Srivastava et al. (2015 and references therein). However, it is widely accepted that this phase of volcanism started at 66.288 ± 0.027 Ma (U–Pb zircon dating; Schoene et al., 2015) about 250,000 years before the K/Pg boundary (65.968 ± 0.085 Ma; Renne et al., 2013), encompassing the age interval of the CF2 and CF1 planktic foraminiferal biozones, and ended at 500,000 years after the K/Pg boundary (65.552 ± 0.026 Ma) (Schoene et al., op. cit.). Apparently, the Deccan phase-2 volcanism has been recorded by three Hg/TOC spikes in the Meghalaya section (Sial et al., 2016: fig. 10a). Spike 1 lies about 50 cm below the K/Pg boundary, probably within the CF2 planktic foraminiferal biozone, spike 2 at the K/Pg boundary, and spike 3, about 30 cm above this boundary. Similar behavior has been observed at the Højerup and Bottaccione K/Pg sections in our paper.

4. The litholog showing the sample location is found in Fig. 4. Samples KT1 through KT8 are from the medium-to light-gray shales of the Mahadeo Formation. Samples KT9 through KT 25 are from the light-gray to dark-gray Danian Langpar Formation. The K/Pg boundary sample is from the yellowish brown to reddish brown shale layer (K/Pg bulk) as shown in our stratigraphic column (Sial et al., 2016: fig. 10 and table 3). The foraminiferal assemblage is shown in the litholog across the K/Pg boundary (Fig. 4) and the other fossil assemblage is displayed in Figs. 2 and 3.
5. Fig. 4 here shows the planktic foraminiferal assemblage and litholog. The position of the samples studied within this section has been already discussed in Point 3. As to enrichment due to secondary processes, we have reported and discussed in detail the Mo/Al ratio chemostratigraphy for all of the studied sections. This ratio, among other things, is known to be useful for discerning the original compositions of rocks that were subjected to later diagenetic processes, low-grade metamorphism or weathering (Wilde et al., 2004). Besides, one of the most effective parameter to screen carbonate samples that have not undergone post-depositional alteration is Mn/Sr ratio (e.g., Jacobsen and Kaufman, 1999), which should be below 1.5 (Fölling and Frimmel, 2002). These criteria have been adopted in the screening of unaltered samples in our paper and, therefore, we assume that the reported elemental and isotopic results are rather near-primary ones.
6. C and Hg/TOC chemostratigraphy of well-known K/Pg boundary sections, besides the use of Hg isotopes in screening of Hg source, were the two main goals of our study. With this purpose, we have analyzed samples calibrated in terms of standard foraminiferal biozones from well-known K/Pg sections in Europe (e. g., Højerup and Bottaccione), distal to the Deccan volcanic center. These two sections have yielded three Hg/TOC enrichment spikes, interpreted as true Hg loading to the environment and, likely, representing Deccan phase-2 volcanism fingerprints. Likewise, the Bidart K/Pg section, in France, has also yielded three spikes (Font et al., 2016). The Um Sohryngkew K/Pg section is regarded as a continuous one, based on standard foraminiferal biozones (e.g., Mukhopadhyay, 2008; Gertsch et al., 2011). The twenty-five samples we collected from Um Sohryngkew section also yielded three Hg/TOC spikes in a similar way to the three classical K/Pg sections in Europe. This strongly suggested that we were dealing with a section straddling the K/Pg boundary.

Hg isotopes in samples that recorded the Hg/TOC spikes in our Um Sohryngkew section, as well as those from the three European K/Pg sections above, lie within the volcanic emission field (Sial et al., 2016: fig. 11). The Deccan phase-2 represents 80% of the total Deccan volcanism, in contrast to Deccan phase-1 which represents only 6% (e.g., Chenet et al., 2009). Therefore, it is likely that the Hg/TOC spikes in our Um Sohryngkew section are fingerprints of the Deccan phase-2 volcanism, rather than Deccan phase-1 as proposed in the Comment.

The detailed geochemical investigations on the K/Pg transition by Srivastava et al. (2013) and Pal et al. (2015a, 2015b, 2015c) are, undoubtedly, meaningful. It sounds wise, however, to examine Hg/TOC ratios and Hg isotopes in samples from the K/Pg boundary section studied by Mukhopadhyay (2008) and contrast the results to those found in our study.

7. We appreciate the contribution of Srivastava et al. (2013) on clay mineralogy, TOC and REE patterns across the K/Pg boundary from Um Sohryngkew section and their findings similar to results from K/Pg sites elsewhere. TOC contents for all of K/Pg sections studied by us have been extensively discussed in our paper, and we hold at hands REE analyses for all of them (publication in preparation). We share the importance of clay mineralogy in studies of the K/Pg boundary, but this was outside the scope of our paper and we believe it does not invalidate our conclusions. The coherence between the isotopic results of our Um Sohryngkew section and K/Pg sections in Europe focused in our paper seems to have provided sufficient evidence to confirm that we were dealing with a K/Pg boundary section at Meghalaya.
8. We appreciate the significant contribution of Pal et al. (2015a, 2015b) on organic geochemical investigations across the K/Pg boundary clay layer in the Um Sohryngkew section and its comparison with other similar clay layers from the Mediterranean region. As explained above, the application of this tool was outside the scope of our paper. We agree that the nature of PAH and the enrichment of kaolinite are important aspects to be investigated in K/Pg sections worldwide.
9. The K/Pg section in which Mukhopadhyay (2008) has identified planktic foraminiferal zones (CF2 to P1a) in the Langpar Formation differs, perhaps, from the one mentioned in the Comment here. The photo they have provided shows only disturbed Langpar Formation shale beds, which look highly deformed, discontinuous and tectonically affected. The transition from the Mahadeoto the Langpar Formation is not seen in that section. One of the keys to address this issue is to look at Hg and C chemostratigraphies, alongside Hg isotopes in samples studied by Mukhopadhyay (2008, 2012a, 2012b), comparing them with the equivalent data reported in Sial et al. (2016). If his stratigraphic section is complete and tectonically undisturbed, this might help addressing this issue.

One cannot exclude the possibility of diachronism of the two formations focused in this discussion, something common in many basins, although they are marine sections at short distance from

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each other. In the Meghalaya area, the K/Pg boundary could have been recorded in the Langpar Formation at one site or in the contact between the Mahadeo and Langpar formations in another. Besides, tectonic-induced doubling of the sequence is also possible as the Meghalaya area, inserted in the Shillong plateau, has experienced kinematic changes due to stresses from the Himalayan collision and Indo-Burma subduction zone, and undergone vertical uplift since the Cretaceous (Islam et al., 2011).

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