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Shallow-water hydrothermal venting linked to the Palaeocene–Eocene Thermal Maximum

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This supplementary information file contains background on Integrated stratigraphy - Shipboard biostratigraphy and organic matter stable carbon isotope analyses for Sites U1567 & U1568

Microfossil content of two Holes (U1567A and U1568A) were intensely studied, while samples from the three middle Holes (U1567B, C and U1568B) were used as supporting information (Table S1) and a similar approach was applied for the carbon isotope analyses.

We here focus on the Paleocene-Eocene succession, starting from *ca.* 30 m below sea floor. In general, the same succession is found at all holes and is subdivided primarily on lithological characteristics (Planke et al., 2022). The succession comprises - from top to bottom:

Early Eocene Lithological unit III; this unit is a consolidated mudstone, typically around 15 m thick and recovered at all holes in subtly increasing thickness from the centre of the hydrothermal vent complex (HTVC); thinnest at U1568A, thickest at U1567A. Unit III is bound by erosional unconformities to lithological units above and below.

Primarily based on the presence of the dinoflagellate cyst (dinocyst) *Membranilarnacia glabra* and rare specimens of the genus *Dracodinium* we assign this lithological unit to the late Early Eocene zone E3 of Bujak & Mudge, which spans from *ca.* 50.4 – 47.4 Million years ago (Ma) (Bujak and Mudge, 1994; Mudge and Bujak, 1996). The palynofacies in this unit is dominated by marine amorphous organic matter (AOM). Carbon isotope analyses of organic matter $\delta^{13}C_{org}$ show values around -28‰ in this unit, in line with values expected for Early Eocene marine organic matter (Sluijs and Dickens, 2012).

Latest Paleocene – earliest Eocene Lithological unit IV and V consist of mud, ash and biosilica-rich strata, with laminated intervals and limited bioturbation. Unit IV is progressively more expanded towards the centre of the HTVC, spanning a massive ~67 m in U1568A, but only ~7 m in U1567A. Seismic imaging shows the top of Unit IV is likely to have been progressively more truncated towards the edge of the HTVC. The underlying Unit V is more consistent in thickness, and spans ~10-15 m.
Unit IV and V in U1567 holes have abundant, often pristinely preserved diatoms, mostly in finely laminated facies. The PETM diatom assemblage closely aligns with that described from the Russian Platform (Oreshkina and Radionova, 2014). The diatoms in U1568A, more proximal to the HTVC centre, are mostly diagenetically altered and unidentifiable.

Dinocyst assemblages are marked by typical Paleocene-Eocene taxa, most importantly the presence of *Apectodinium augustum*, a taxon limited to the Paleocene-Eocene Thermal Maximum (PETM) carbon isotope excursion (CIE) (e.g., Sluijs et al., 2006; 2007), and its presence defines subzone P6b of Bujak

& Mudge, 1994. The only productive sample within Unit IV that did not contain *A. augustum*, was taken from the upper part of the interval at U1568A (U1568A-7H-CC). In this particular sample, the assemblage with common *Hystrichosphaeridium tubiferum* is consistent with the earliest Eocene dinocyst zone E1.

A few metres above the lower boundary of Unit V and coincident with the appearance of *A. augustum*, we record a 1.5–6‰ decrease in $\delta^{13}C_{org}$. We thus assign Unit IV and the top part of Unit V to the PETM CIE. Some further indications for the timing relative to the global CIE comes from the uppermost sample taken at U1568A, where the dinocyst zone E1 suggest the later stages of the CIE (recovery) might be preserved at the centre of the HTVC but this cannot be confirmed by $\delta^{13}C_{org}$ measurements.

As the CIE occurs within Unit V, the onset of the PETM does not coincide with the transition from Unit VI to V. This is evidenced by a few samples within the lowermost parts of Unit V at U1567C, U1568B and U1567A that show $\delta^{13}C_{org}$ around -25.5 to -26‰, similar to the Late Paleocene Unit VI and not ^{13}C-depleted as most of the PETM record above. Some of the samples from the lower parts of Unit V and uppermost part of Unit VI, (U1567A-10X-CC, U1567B-11X-CC) contain *Apectodinium*, but not *A. augustum*, confirming a latest Paleocene (pre-PETM) age (upper part of zone P6a) for this interval. The early appearance of the genus *Apectodinium*, is typically seen not more than a few 10s to 100 kyr prior to the PETM at high-latitudes (Sluijs et al., 2007; Sluijs et al., 2011; Frieling et al., 2018). Whether this interval is condensed and missed in other Holes, by low sampling resolution or poor yield, or locally absent from the stratigraphy could not be resolved with the current data.

Furthermore, it remains somewhat uncertain whether the lowermost samples within Unit V represent vent-infill deposited prior to the onset of the global CIE. For example, we cannot exclude the possibility that these samples are influenced by input of older organic matter or that some of the deposition of the unit is related to slumping of Paleocene material down the rim of the crater during the eruption of the HTVC which is consistent with the lowermost seismic unit in the infill being slightly thicker towards the rim (Fig. 2).

The most expanded interval within all U1567 and U1568 Holes appears to be the PETM interval, as identified here by the coeval appearance of the 1.5–6 ‰ negative CIE and *A. augustum*. The palynofacies, that consists almost purely of terrestrial organic matter (OM) suggests that the overall shape of the CIE may have been influenced by a shift in organic matter sourcing. This may occur as Paleogene marine-dominated OM is far more ^{13}C-depleted compared to present-day marine OM (Hayes et al., 1999) and depleted by ~4‰ compared to terrestrial OM in the early Paleogene (Sluijs and Dickens, 2012). Generally, shifts towards more marine-dominated organic matter, as a result of sea level rise and increased marine productivity (Sluijs et al., 2014; Carmichael et al., 2017), are common

(Sluijs and Dickens, 2012) and can amplify the observed CIE in $\delta^{13}C_{org}$. Indeed, most marginal marine PETM successions (e.g., Zachos et al., 2006; Kender et al., 2012; Jones et al., 2019) have a tendency to record greater-amplitude CIEs in $\delta^{13}C_{org}$ compared to the global average. The marine-organic matter dominated samples from U1567B and 1567C are also the most ^{13}C-depleted (Fig. 3), while samples below have more or less equal proportions of marine and terrestrial organic matter. We therefore relate the exceptional amplitude of the CIE (~6‰) to a short-lived increase in marine organic matter loading. In contrast, a very dominant terrestrial organic carbon fraction marks much of the Unit V and IV strata above, implying for those strata the CIE amplitude may have been suppressed compared to the late Paleocene. When compared to previously analysed age-equivalent samples (SI Fig. 1), we find $\delta^{13}C_{org}$ values of -27 to -28‰ during the CIE are in excellent agreement with the terrestrial OM-dominated samples from the PETM CIE body in the Arctic Ocean (Sluijs and Dickens, 2012) and nearby Grane core (Jones et al., 2019).

Duration of infill prior to CIE onset

Sedimentological and stratigraphic evidence clearly suggest infill commenced prior to the CIE, as observed in Hole U1567C. We can provide a pre-CIE infill duration estimate by extrapolating the sediment accumulation rates from the strata above. Assuming the ~100 kyr body of the CIE is complete (a maximum duration), and sediment accumulation of ca. 30 m in U1567C, we derive minimum accumulation rates of 30 cm kyr^{-1}. Extrapolating that number to the ~13 m of strata below the CIE in U1567C gives a maximum infill duration of ~43 kyr. This is likely to be a significant overestimate for several reasons:

In the absence of further constraints being available and the lack of CIE recovery values in Hole U1567C, the minimum sediment accumulation rate estimates used to extrapolate have to be based on the assumption the CIE body is complete, which is unlikely to be the case. Further, the initial infilling of a sea floor depression, as for example shown in the Figge Maar blow-out crater (refer to SI Fig. 1/ED Fig. 1), is likely to be (even) higher than in the PETM-aged strata above, also due to slumping and reworking of soft sea-floor materials near the crater. Indeed, in the U1568A succession, the CIE is close to 80 m thick (Fig. 2) and clear CIE recovery values are not recorded, which shows that sediment supply to this system was exceptionally high. In addition to biogenic and siliciclastic sedimentation, the initial infill and CIE onset interval in U1567C and U1567B are marked by substantial and more frequent ash deposition than strata above and Unit VI below (Figure 2). Lastly, it is intriguing the U1568A record does not appear to preserve any pre-CIE infill strata. This could suggest that sediment could not accumulate in the most vent-proximal areas due to ongoing activity in the hydrothermal system. As fluid flow largely depends on heating by sills in the subsurface, it may be assumed that vigorous activity in these systems does not last much beyond the cooling time (~1 kyr for a 100-m thick sill, e.g. Aarnes et al. 2010), and if the system had a single eruptive phase, this may suggest the duration of pre-CIE infill could in fact be limited to within a few millennia of the CIE onset. We surmise that the biostratigraphic and geochemical evidence strongly supports an infill duration of <<43 kyr, and it is plausible that the vent erupted as little as 1 - 2 millennia prior to the onset of the PETM CIE

Late Paleocene Unit VI is characterized by green bioturbated mudstones and yields typical Late Paleocene dinocysts, such as *Spiniferites rhomboideus* and *Alisocysta margarita* (Vieira and Mahdi, 2019). Unit VI is assigned to Bujak & Mudge zone P5, but in most Holes the zone could not be further subdivided due to relatively low yields and limited processing time often resulting in low sample quality. Marine and terrestrial organics are more or less equally abundant in the palynofacies. $\delta^{13}C_{org}$ shows values around -25.5‰, typical of Late Paleocene mixed organic matter (e.g., Harding et al., 2011; Frieling et al., 2018) (SI Fig. 1).

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