Marine and terrestrial environmental changes in NW Europe preceding carbon release at the Paleocene-Eocene transition

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ABSTRACT

Environmental changes associated with the Paleocene-Eocene Thermal Maximum (PETM, ~56 Ma) have not yet been documented in detail from the North Sea Basin. Located within proximity to the North Atlantic Igneous Province (NAIP), the Kilda Basin, and the northern rain belt (paleolatitude ~54°N) during the PETM, this is a critical region for testing proposed triggers of atmospheric carbon release that may have caused the global negative $\delta^{13}C$ carbon isotope excursion (CIE) in marine and terrestrial environments. The CIE onset is identified from organic matter $\delta^{13}C$ in exceptional detail within a highly expanded sedimentary sequence. Pollen and spore assemblages analysed in the same samples for the first time allow a reconstruction of possible changes to vegetation on the surrounding landmass. Multiproxy palynological, geochemical, and sedimentologic records demonstrate enhanced halocline stratification and terrigenous deposition well before (10^3 yrs) the CIE, interpreted as due to either tectonic uplift possibly from a nearby magmatic intrusion, or increased precipitation and fluvial runoff possibly from an enhanced hydrologic cycle. Stratification and terrigenous deposition increased further at the onset and within the earliest CIE which, coupled with evidence for sea level rise, may be interpreted as an increase in precipitation over NW Europe consistent with an enhanced hydrologic cycle in response to global warming during the PETM. Palynological evidence indicates a flora dominated by pollen from coastal swamp conifers before the CIE was abruptly replaced with a more diverse assemblage of generalist species including pollen similar to modern alder, fern and fungal spores. This may have resulted from flooding of coastal areas due to relative sea level rise, and/or ecological changes forced by climate. A shift towards more diverse angiosperm and pteridophyte vegetation within the early CIE, including pollen similar to modern hickory, documents a long term change to regional vegetation.

Keywords:
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1. Introduction

The PETM was a period of geologically-rapid global warming that punctuated a warming Eocene climate ~55.8 Ma ago (Charles et al., 2011), and saw sea surface temperatures rise by ~5–8°C from background levels (Zachos et al., 2005; Sluijs et al., 2007). It was associated with a substantial injection of δ^{13}C-depleted carbon into the ocean-atmosphere system (see Pagani et al., 2006a) over <20 ka (Cui et al., 2011), causing a negative carbon isotope excursion (CIE) of between –2 and –7‰ in marine and terrestrial sediments (see overview in Schouten et al., 2007) lasting ~170 ka (Röhl et al., 2007), and a prominent dissolution horizon in the deep sea signifying deep ocean acidification (Kennett and Stott 1991; Zachos et al., 2005). The source and rate of released carbon is still under debate (Pagani et al., 2006a; Zeebe et al., 2009; Cui et al., 2011), but may have been linked to the dissociation of marine hydrates containing biogenic methane (δ^{13}C of < –60 ‰) (Dickens et al., 1995), thermogenic methane from marine sediments around the Norwegian Sea (Svensen et al., 2004), or dissolved methane from a silled Kilda Basin between Greenland and Norway (Nisbet et al., 2009).

The PETM may be a good analogue to test modelling studies that suggest current global warming trends may result in an enhanced hydrologic cycle (Seager et al., 2010), whereby increased precipitation in temperate rain belts is coupled with increased evaporation in lower latitudes. Modelling studies of the PETM have further indicated the potential importance of an increased hydrologic cycle (Lunt et al., 2010; Bice and Marotzke 2002), which could have altered ocean circulation causing methane hydrate reservoirs to destabilise, triggering massive carbon release (Bice and Marotzke, 2002). Palynological evidence from Arctic Spitsbergen (Harding et al., 2011) and New Zealand (Crouch et al., 2003a) suggests increased terrestrial runoff occurred at the onset of the CIE which may be related to hydrologic changes, and massive Pyrenees conglomerate deposits have been interpreted as the result of an abrupt increase in extreme precipitation within the early CIE (Schmitz and Pujalte, 2007). In addition hydrogen and carbon isotope measurements of terrestrial plant and aquatic-derived *n*-alkanes from the central Arctic Ocean indicate that the core of the PETM was associated with increased precipitation and hence hydrologic cycle (Pagani et al., 2006b), although the onset of PETM warming was not recovered in the sediment core. Despite numerous additional evidence for changes in terrestrial runoff and potentially hydrology during the PETM (see overview in McInterney and Wing, 2011), there is a lack of clear evidence for hydrologic changes from high resolution sections able to resolve important lead/lag relationships, and therefore there remains a need for studies of hydrologic changes in sensitive locations over the onset of the CIE in order to understand the relationship between precipitation and global carbon release.

Biome changes in response to modern global warming have been observed, but approaches to predict the vulnerability of ecosystems to future changes are still in development (Gonzalez et al., 2010). Vegetation shifts during the rapid warming associated with the PETM may provide a useful analogue to future biome responses. Whilst neotropical vegetation in Central America appears to have responded to warming during the PETM with increased diversity and origination rates (Jaramillo et al., 2010), central North America experienced a rapid migration of plant communities associated with lower precipitation at the onset of the CIE (Wing et al., 2005), and southern England may have experienced a major change in plant composition due to changes in local fire-regime (Collinson et al., 2009). To better understand biome responses to climatic change during the PETM, further high resolution vegetation cover studies are needed, specifically from temperate and boreal forests which may be amongst the most vulnerable ecosystems to global warming (Gonzalez et al., 2010).

In this study we focus on paleoenvironmental signals from a high resolution marine core collected from the central North Sea (Fig. 1), in order to understand changes to precipitation, ocean stratification, productivity and vegetation over the onset of the PETM. This core is located in a critical region proximal to the NAIP, as there are currently no high resolution records of environmental change during the PETM from the North Sea Basin. Furthermore, as overturning of a stratified Kilda Basin is hypothesised as a possible trigger for dissolved methane release to the atmosphere (Nisbet et al., 2009), analysing the stratification history of the nearby North Sea is a possible way to test this hypothesis.

2. Regional setting

During the late Paleocene–early Eocene the North Sea was a restricted marine basin, characterised by siliciclastic sedimentation and high terrigenous input, principally from the Scotland-Faeroe-Shetland landmass (Knox 1998, Fig. S1). Core 22/10a-4 is located in the central part of the basin (Figs. 1, S1), and is therefore disconnected from many marginal marine processes that could mask oceanographic signals (e.g. tidal or storm-induced erosion and slumping). Paleobathymetry estimates in the North Sea during the Paleocene and Eocene are difficult to constrain accurately, as the extant benthic foraminifera present in the Paleogene are found today living between 200 and >1000 m water depth (Gradstein et al., 1992), and are controlled predominantly by substrate and bottom water properties. However using a number of paleoecologic micropaleontology methods together (Gillmore et al., 2001), along with 2D structural restoration (Kjennerud and Sylta 2001), broad agreement was found and central parts of the northern North Sea

appear to have had paleodepths of >0.5 km in the earliest Eocene near 22/10a-4 (Kjennerud and Gillmore, 2003, Fig. S1).

As 22/10a-4 is in the deep (>0.5 km) central part of the basin, it acted as a depocentre and thus exhibits an expanded sedimentary sequence in which we observe no evidence of hiatuses (erosional surfaces, discontinuous bedding/lamination, truncated isotope stratigraphy). Although transported material is evident by the thin sandstones (typically <10 cm) interpreted as turbidites, these have not been sampled in this study. During the late Paleocene, the basin became restricted following a fall in sea-level of ~100 m that resulted from regional uplift associated with the proto-Iceland mantle plume in the North Atlantic (see Knox, 1996). This event is evident in 22/10a-4 as a lithologic change from unbedded to bedded mudstone (the Lista and Sele Formation boundary, Fig. 2). Restriction of the basin also led to the establishment of poorly oxygenated bottom waters, as is evident by a shift in the benthic foraminiferal assemblages towards low diversity low oxygen-tolerant agglutinated species (Knox 1996). The CIE at the Paleocene–Eocene boundary was accompanied by a relative sea level rise, as documented in southeast England (Powell et al., 1996) and Spitsbergen (Harding et al., 2011), due to the thermal expansion of sea water and possible melting of ice caps, although the North Sea basin remained restricted as evidenced by the persistence of low-oxygen facies in 22/10a-4. The North Sea had a widespread freshwater catchment area, and a halocline was in place from the late Paleocene to early Eocene (Zacke et al., 2009). Therefore, surface water salinity changes in the North Sea Basin provide a sensitive gauge for stratification forced by changes in tectonics and the hydrologic cycle.

3. Methods and materials

3.1. Sedimentology

Borehole 22/10a-4 (57°44’8.47’’N. 1°50’26.59’’E.) comprises a continuous core through the Forties Sandstone Member and into the Lista Formation (Fig. 2). The core consists of fine grained shale with interbedded fine to coarse grained sandstone layers interpreted as turbidites, with occasional mm-thick ash layers (Fig. 3). All samples in this study were taken from claystone horizons to avoid sampling substantial quantities of transported material. The section of 22/10a-4 analysed in this study is from 2605 m to 2634 m (core depth), chosen because this part of the core is predominantly in claystone facies and provides a greatly expanded section over the onset of the CIE (Figs. 2, 3). At ~2609 to 2613 m the claystone becomes finely laminated, with alternately pale and dark laminae couplets ranging from 1-25 per mm (Fig. S2). The pale laminae consist of clay and

fine sand, and the dark laminae are rich in organic carbon and pyrite inclusions. Laminae were counted at 26 horizons throughout the core and average ~13 pairs per mm.

3.2. Micropaleontology

A total of 71 palynology samples were prepared at the British Geological Survey using standard preparation procedures (Moore et al., 1991). Samples were demineralized with hydrochloric (HCl) and hydrofluoric (HF) acids, and residual mineral grains removed using heavy liquid (zinc bromide) separation. Elvacite was used to mount slides. The palynomorphs were analysed using a Nikon transmitted light microscope, counting the total number of palynomorphs on a strew slide (Table S1). Each slide was produced from 1/100th of the total material processed, where the initial weight of material was 5 g of dried sediment. Thus, the palynology counts represent the total number of specimens per 0.05 g of dried sediment. Statistical analysis was carried out using the software of Hammer et al. (2005). The %wood/plant tissue was determined by palynological investigation, and is the sum of '%wood plant tissue' and '%various (non-woody) plant tissue' in Table S1. Organic material for $\delta^{13}C_{AOM}$ analysis was collected from the same palynology samples, and the remaining processed material separated into size fractions. The >250 µm fractions, found through light microscope analysis to be dominated (>90%) by amorphous organic matter (AOM), were also analysed for $\delta^{13}C$. Foraminifera samples of between 20 and 60 g of dried sediment were processed by washing through a 63 µm sieve with water. All specimens were counted and converted to foraminifera/g (Table S2). All species exhibited agglutinated (non calcareous) test walls.

3.3. Geochemistry

All analyses were carried out at the NERC Isotope Geosciences Laboratory. C and N analyses (from which we present weight % C/N) were performed on 225 samples by combustion in a Costech ECS4010 Elemental Analyser (EA) calibrated against an Acetanilide standard (Table S3). C/N atomic ratios were calculated by multiplying by 1.167. Replicate analysis of well-mixed samples indicated a precision of ± <0.1. Carbon isotope analysis was carried out on 289 bulk rock samples (Table S4) after removing migrated hydrocarbons (Stephenson et al., 2005). The hydrocarbons were removed by crushing the rock fragments using a ball mill, and the soluble organic matter from all rock samples was extracted using a Soxhlet extractor. The samples were refluxed for 24 h in an azeotropic mixture of dichloromethane and methanol (93:7, v/v). All materials (cellulose Soxhlet thimbles, silica wool, vials) were cleaned with analytical grade organic

solvents prior to use. Any remaining solvent was then removed by evaporation and the dried sediments were transferred to vials. Any calcites (shelly fragments) were removed by placing the samples in 5% HCl overnight before rinsing and drying down. Carbon isotope analysis was also carried out on palynology residues of the >250 µm size fractions dominated by AOM. $^{13}C/^{12}C$ analyses were performed on 35 samples by combustion in a Costech Elemental Analyser (EA) on-line to a VG TripleTrap and Optima dual-inlet mass spectrometer, with $\delta^{13}C$ values calculated to the VPDB scale using a within-run laboratory standards calibrated against NBS-18, NBS-19 and NBS-22. Replicate $^{13}C/^{12}C$ analyses were carried out on the section, and the mean standard deviation on the replicate analyses is 0.4‰.

4. Results and discussion

4.1. Statistical analysis

Correspondence analysis (CA) and statistical diversity analysis was carried out on the palynological dataset (total counts per gram) to confirm assemblage designations (Figs. 4, 5), to identify any disturbance to the core prior to interpretation, and to estimate diversity (Fig. 6). Dinoflagellate cyst assemblages (DA1–DA5) and pollen assemblages (PA1–PA4) were defined by visually comparing changes in the species dominance (Figs. 7, 8), and confirmed by CA (Fig. 4) using the first three axes (describing the highest percentages of variance). Five samples from below the CIE at 2619.60, 2617.35, 2617.44, 2614.73 and 2614.71 m (indicated in Fig. 5) contain *Apectodinium*, in contrast to the other samples below the CIE (Figs 3, 7). Some of these samples (2619.60, 2614.73 and 2614.71 m) also contain negative $\delta^{13}C_{TOC}$, indicative of the CIE (Fig. 3). To test if coincident pollen and spore changes also occur in these samples, we used CA on the pollen and spore data only (Fig. 5a). PA1–PA4 (symbols) plot in clusters, signifying their palynological similarity. The species most associated with an assemblage are clustered with the samples from that assemblage. For example, *Inaperturopollenites hiatus* and bisaccate pollen (highly abundant before the CIE, Fig. 8) are high on axis 1 where the earlier samples from PA1 and PA2 occur, and *Caryapollenites* spp. and fungal spores (abundant after the CIE, Fig. 8) are low on axis 1 near the younger samples from PA4. The two samples 2619.60 and 2614.71 have a spore and pollen palynological signature similar to samples from PA3/4 during the CIE (plot lower on axis 1) and are either not in the correct location (it cannot be discounted that these samples were misplaced during drilling operations core handling), or represent very short episodes of both marine and terrestrial ecologic change to CIE-type conditions. The rapid and transient nature of these two shifts appears

to suggest that the latter explanation may be unlikely, and we have therefore shaded samples from these two depths in Figs. 3, 6, 7, 8, 9.

The majority of the morphospecies in our study represent taxonomic groupings of terrestrial plant species, from generic to higher level groupings, such that any palynology diversity measure will underestimate vegetation diversity. However, although subtle changes may not be resolved, large changes to the diversity of regional vegetation are likely to be reflected in palynology assemblages. Palynological data have therefore previously been used to estimate plant diversity in the geological record (e.g. Ogaard, 2001; Harrington, 2004; Jaramillo et al., 2010). The number of pollen and spore species in each sample (Fig. 6d) fluctuates, trending towards increasing number over the CIE onset (from PA2 to PA3). To account for the changing number of specimens counted in each sample (more species will be encountered with higher counts), two statistical indices were used (Fig. 6e, f). Both confirm the significant increase in pollen and spore diversity from before to after the CIE onset. We don't use the range-through method, as we are interested in changes to local vegetation habitats. The increase in the number of pollen and spore species appears to be largely driven by pteridophytes (largely ferns, Fig. 6g), although angiosperm diversity also increases over the CIE onset (Fig. 6h) and fungal spores become more prevalent (Fig. 8m). Gymnosperms are only represented by two morphospecies and so diversity was not calculated for this group.

4.2. Isotope changes and age model

The Lista and Sele Formation boundary (Fig. 2) occurs in the lower part of magnetochron 24r in the Faeroe-Shetland Basin (Mudge and Bujak, 2001), giving a date of >56.6 Ma for the base of the studied section (Gradstein et al., 2004). The top of the studied section is within nannofossil zone NP9 (Knox, 1996), dated as <55.7 Ma (Gradstein et al., 2004). The Paleocene-Eocene marker event *Apectodinium augustum* (Bujak and Brinkhuis 1998) occurs in 22/10a-4 at 2617.35 m (Fig. 3), approximately at the CIE onset (as was found at 30/14-1, Fig. 1, Sluijs et al., 2007). The global CIE is determined by the negative shift in both marine and terrestrial $\delta^{13}C$ of between ~2‰ and ~7‰ (see Schouten et al., 2007) and has recently been radioisotopically dated in Spitsbergen to ~55.8 Ma (Charles et al., 2011). The global CIE onset is present in 22/10a-4 within the interval ~2614 to ~2612 m (Fig. 3), where a negative $\delta^{13}C$ shift of ~5‰ occurs along with a peak in *Apectodinium*. The palynological results (from samples containing sufficient material for analysis; red symbols in Fig. 3) show that %wood/plant tissue varies from 10% to 88% throughout this interval (Fig. S3). We found that the samples containing >30% wood/plant tissue (empty red symbols in Fig. 3) in the palynological residues have consistently heavier $\delta^{13}C_{TOC}$ values than those with less wood/plant tissues (solid red symbols). The presence of transported C_{org} thus precludes an

unambiguous interpretation of the rate of atmospheric carbon release from the shape of the CIE onset at 22/10a-4, but the initial negative $\delta^{13}C$ shift between 2614.3 and 2613.5 m (shaded box, Fig. 3) can be taken as marking the earliest evidence of $\delta^{13}C$-depleted atmospheric carbon release and the onset of the global CIE. This positioning of the CIE onset is supported by $\delta^{13}C$ analysis of isolated AOM (Fig. 3), likely of marine origin (see Supplementary Material) and less likely to be composed of mixtures of different sources of C_{org} (although it is still susceptible to reworking).

As we observe only a ~1‰ 'recovery' in the CIE, the age of the top of the studied section appears to be no more than ~75 ka from the CIE onset by comparison with the cyclostratigraphically correlated Longyearbyen Section in Spitsbergen (Charles et al., 2011), giving an average sedimentation rate of ≥8 cm/ka after the CIE onset (if sand horizons (Fig. 3) are removed, assuming rapid turbidite deposition). The long term ~0.7‰ positive shift in $\delta^{13}C$ at 2609 m (Fig. 3) may correlate to a similar shift in the Longyearbyen Section (Charles et al., 2011), which is cyclostratigraphically correlated to ~45 ka after the CIE onset (Charles et al., 2011), and would give a sedimentation rate of on average ~8 cm/ka for this section. The finely laminated part of the core may also provide temporal insight. There is no direct evidence for the period of deposition of each lamination couplet, but as modern marine lamination-forming basins produce annual laminae pairs (e.g. the Black Sea: Arthur, 1994; Carioaco Basin: Tadesco and Thunell, 2003; Santa Barbara Basin: Thunell et al., 1995), the ~40,000 estimate of the number of laminae pairs present between ~2613 and ~2609 m (see Section 3.1) may represent ~40 ka, and a sedimentation rate of ~7.5 cm/ka.

4.3. Dinoflagellate cysts and surface water changes

Dinoflagellate cysts have been used extensively for reconstructing paleoenvironments in the Paleogene (see overview in Sluijs et al., 2005), as they are particularly sensitive to changes in salinity, temperature and nutrient levels (Powell et al., 1992; Pross and Brinkhuis 2005; Sluijs et al., 2005). We calculate "%low salinity dinoflagellate cysts" (Figs. 7–9) by grouping cysts of similar inferred ecologic preferences (see Fig. 7, and discussion in the Supplementary Material) to provide an indication of environmental change, and by excluding species of uncertain affinity such as *Apectodinium*. Samples with fewer than 20 specimens were also excluded. Despite the limitations of this method, the large variation in the %low salinity dinoflagellate cysts (ranging from 0–80%) clearly indicates that significant environmental changes in surface water conditions occurred during the CIE onset in the central North Sea, and is supported by coeval changes in the sedimentary carbon/nitrogen (C/N) ratio (Fig. 10) which reflects changes in the proportion of terrestrial/marine

organic material deposited in the North Sea Basin due to terrestrial runoff and productivity (see Section 4.4).

Dinoflagellate cyst assemblage 1 (DA1, ~2632 to 2618 m, Fig. 7p) contains high proportions of typically open marine and hence normal marine salinity associated *Achomosphaera/Spiniferites* spp., undifferentiated chorate cysts and *Areoligera/Glaphyrocysta* spp. DA1 also contains on average ~5% peridinoid cysts including *Deflandrea* spp. regarded as a coastal/neritic taxon indicating high productivity and nutrient availability (Brinkhuis, 1994; Pross and Brinkhuis, 2005). These characteristics indicate that a somewhat restricted but fully marine shelf environment was present before the onset of the CIE in the central North Sea, with availability of nutrients indicated by the presence of *Deflandrea* spp.

DA2 (~2618 to 2614 m) contains a similar abundance of *Achomosphaera/Spiniferites* spp. and chorate cysts to DA1, but with a marked increase in the abundance of low salinity tolerate *Cerodinium depressum* and *Senegalinium* spp., which may also have been a heterotrophic genus indicative of elevated nutrient levels (Sluijs and Brinkhuis, 2009). Undifferentiated peridinoid cysts also peak in abundance, and may also be indicative of elevated nutrient and reduced salinity conditions (although they are not included in the %low salinity dinoflagellate cysts, Fig. 7c). There is also an increase in the abundance of *Areoligera/Glaphyrocysta* spp., thought to indicate unrestricted neritic environments of more typical marine salinity. In summary, the higher proportion of low salinity tolerant dinoflagellate cysts in DA2 (Fig. 7c) appears to indicate continuous or episodic freshening surface waters below typical marine salinities (< ~31‰ in the modern ocean). In addition, DA2 is characterised by elevated numbers of dinoflagellate cysts per gram (~70 per 0.05g in DA1, ~110 per 0.05g in DA2) indicating a possible increase in cyst production which would be consistent with increased fluvial runoff carrying nutrients from nearby landmasses. DA2 also contains an abundance maxima of *Apectodinium*, an extinct genus with a somewhat uncertain ecologic affinity (see Crouch et al., 2003b), although likely reflecting relatively warm and eutrophic conditions (Crouch et al., 2003b; Sluijs et al., 2007; see Supplementary Material). The transient appearance of *Apectodinium* occurs with a reduced abundance of open marine dinoflagellates, and a peak in *Deflandrea* indicating possibly lower salinity and higher nutrient availability.

DA3 (~2614 to 2612 m) is characterised by *Apectodinium* making up on average ~40% of the assemblage, and a reduction in the abundance of all other species apart from *Deflandrea* and *Senegalinium*. DA3 may therefore indicate a continuation of high nutrient surface water with an elevated freshwater input. In this respect, it is perhaps similar to DA2, although the low abundances of dinoflagellates with known ecologic affinities make interpretations more tentative. The appearance of bottom water anoxia (laminations) and sporadic high accumulation of marine AOM (see Supplementary Material) is consistent with highly productive surface waters.

DA4 (~2612 to 2609 m) shows an increase in the number of low salinity/high nutrient species *Cerodinium depressum* and *Senegalinium* spp., with a continued low abundance of normal marine salinity *Achomosphaera/Spiniferites* spp., such that the proportion of low salinity tolerant dinoflagellate cysts is at a maximum (Fig. 7c) indicating the presence of a pronounced halocline. The lack of freshwater fern and algal spores *Azolla*, *Pediastrum* and *Botryococcus* (Table S1) indicates that surface water salinities were probably not below ~5‰ (the limit for *Azolla*, Brinkhuis et al., 2006), and may have been greater than ~20‰ as *Botryococcus* was found living in salinities as high as 20‰ in modern Australian lakes (de Deckker, 1988). This minimum salinity value is consistent with Zacke et al. (2009), who found continuous occurrences of shark teeth in shallow marine North Sea facies throughout the late Paleocene/early Eocene, and noted that sharks do not live in salinities below ~20‰ in the modern ocean. Highly productive surface waters are indicated by dinoflagellate cysts (Fig. 7c), and consistent with bottom water anoxia (laminations and disappearance of benthic foraminifera) and very high accumulation of marine AOM.

DA5 (~2609 to 2607 m) shows a decrease in low salinity tolerant dinoflagellate cysts and an increase in unrestricted (normal marine salinity) *Areoligera/Glaphyrocysta* spp. and other chorate cysts. The reduction in AOM together with the loss of lamination (possibly end of bottom water anoxia) indicates a return towards marine salinities that existed before the onset of the CIE (represented by DA1), although the persistence of *Apectodinium* may indicate a long term change in marine ecology.

4.4. Enhanced terrigenous deposition

The increase in abundance of low salinity tolerant *Cerodinium depressum* and *Senegalinium* spp. before and after the CIE onset (DA2 to DA4, Fig. 7) most likely indicates a reduction in surface water salinity and elevated nutrient levels (see Section 4.3). This appears to have been associated with increased terrigenous deposition, as evidenced by a concomitant increase in the C/N ratio of 22/10a-4 (Figs. 9c, S4), and an elevated kaolinite contribution to the clay assemblage (Fig. 9e). Sedimentary atomic C/N ratios can be used to differentiate the origin of organic matter (Meyers, 1997; Storme et al., 2012), with values in 22/10a-4 averaging ~10–15 (Fig. 10) indicating a mix of terrestrial land plant-derived and marine/lacustrine algal-derived carbon (Meyers, 1997). C/N values broadly follow %low salinity dinoflagellate cysts (Fig. 9), with an increase at the Lista/Sele Formation boundary, ~4 m before the onset of the CIE, and within the early CIE. As dinoflagellate cyst assemblages suggest higher productivity (%low salinity dinoflagellate cysts, *Apectodinium*), the C/N ratio increase likely indicates an increased flux of terrigenous material to 22/10a-4, rather than a lower marine carbon flux. Kaolinite also increases before and after the onset

of the CIE in 22/10a-4 (Fig. 9e). Kaolinite forms as a result of intense chemical weathering that typically develops on well-drained surfaces receiving high precipitation (Robert and Kennett, 1994). Increased kaolinite at the PETM had been regarded as indicative of an increase in chemical weathering and hence humidity in the source region (e.g. Robert and Kennett, 1994; Knox, 1996), but the long formation time of thick soil kaolinite (>1 myr) suggests these increased proportions probably resulted from erosion of previously-formed kaolinite (Thiry and Dupuis, 2000; Schmitz et al., 2001).

4.5. Pollen, spores and vegetation shifts

The pollen and spore assemblages (PA) that characterise the pre-CIE interval in 22/10a-4 (PA1 and PA2, Fig. 8n) are dominated by *I. hiatus* and bisaccate pollen. Both taxa are produced in abundance by a variety of coniferous plants, and are typical in the Paleogene of the northern UK and Greenland region (Boulter and Manum 1989; Jolley and Whitham 2004; Jolley and Morton 2007), as well as mid-latitude North America (Smith et al., 2007) and Arctic Canada (Greenwood and Basinger, 1993). *I. hiatus* is a member of the Cupressaceae family (coniferous trees) and most likely represents *Metasequoia* and/or *Glyptostrobus* swamp conifers (Greenwood and Basinger, 1993; Jolley and Morton, 2007; Jolley et al., 2009). In terrestrial Paleocene deposits from western Scotland, abundant *I. hiatus* was recorded in association with *Momipites*, *Cupuliferoipollenites*, *Platycaryapollenites*, *Plicapollis pseudoexcelsus*, and *Alnipollenites* interpreted as derived from a channel-margin bog community on a wet substrate (Jolley et al., 2009). Bisaccate pollen (family Pinaceae) was likely derived from temperate coniferous trees possibly on dryer substrates, with an elevation from several meters within swamps (Greenwood and Basinger, 1993) to possibly much higher altitude (Jolley and Whitham 2004). Both are relative overproducers of pollen (Smith et al., 2007). Lowland swamp vegetation appears to have been present before the CIE on the Scotland-Faeroe-Shetland platform (this study; Jolley and Morton 2007), the catchment area for 22/10a-4 (Fig. S1, Knox 1996), with the relative increase of *I. hiatus* over bisaccate pollen from PA1 to PA2 possibly due to sea level fall at the Lista/Sele boundary (Knox, 1996) allowing lowland swamp conifers to expand and/or come into closer proximity to 22/10a-4.

The most significant floral change at 22/10a-4 occurs at the onset of the CIE (PA2 to PA3), with a large drop in the proportion of *I. hiatus*, increasing diversity (Fig. 6), higher proportions of angiosperms (*Alnipollenites* and *P. pseudoexcelsus*), and fern and fungal spores (Fig. 8). *Alnipollenites* was probably from the birch *Alnus* (alder), and in this setting represents generalist vegetation as it occurred in all the terrestrial Paleocene communities from western Scotland (Jolley et al., 2009). *Alnus* and associated ferns were most common where *Metasequoia* (possible source of

I. hiatus) was rare in Arctic Canada, where exceptionally well-preserved Paleocene-Eocene *Metasequoia* and *Glyptostrobus* swamp deposits are prevalent, and interpreted as *Alnus*–fern bogs (Greenwood and Basinger, 1993). *Alnus* is a known nitrogen-fixing pioneer species in nutrient-depleted soils (Hobbie et al., 1998). *P. pseudoexcelsus* is similar to modern *Juglans* (walnut) pollen sometimes associated with wetland plants (Jolley and Whitham, 2004). The fern spores *Cicatricosisporites* and *Laevigatosporites* (family Schizaeaceae) were generalists, associated with all environments from the terrestrial Paleocene communities analysed in western Scotland (Jolley et al., 2009). Since dinoflagellate cysts indicate increased halocline stratification of the North Sea at the CIE (Fig. 9d), and C/N ratios indicate a greater proportion of terrestrial carbon (Fig. 9c), it seems likely that the large reduction in *I. hiatus* in 22/10a-4 reflects a change in vegetation cover in the source region rather than simply a general reduction in the supply of pollen due to proximity of the coast.

Replacement of *I. hiatus*-dominated swamp communities (PA2) with generalist taxa (PA3) indicative of *Alnus*–fern bogs (Greenwood and Basinger, 1993) would be consistent with local sea level fall (partially draining coastal plains) or sea level rise (flooding established coastal swamp plains). There is evidence for sea level rise at the onset of the CIE in southeast England (Powell et al., 1996) and in Spitsbergen (Harding et al., 2011), although any uplift associated with the proximal NAIP could have caused local sea level fall near the Faeroe-Shetland platform. Alternatively, the reduction in gymnosperm swamp conifers and pines at the expense of angiosperms may have been a climatic response to the PETM, as a similar floristic change was also recorded during the PETM in the Bighorn Basin, Wyoming (Smith et al., 2007), and the Lomonosov Ridge (Sluijs et al., 2006). The early ephemeral peak in *Alnipollenites* and *P. pseudoexcelsus* and drop in coastal swamp *I. hiatus* at 2617.35 m, associated with the initial peak in *Apectodinium* (Fig. 8), may indicate a brief episode of coastal flooding from increased precipitation, in association with lower surface water salinity (peak in *Deflandrea*, Fig. 7i).

A further ecologic shift occurs within the core of the CIE (PA4) as *Caryapollenites* (Fig. 8h) begins to dominate, moss (*Stereisporites*) and fungal spores increase in relative abundance, bisaccate pollen and *Alnipollenites* decreases (Fig. 8), and diversity remains relatively high (Fig. 6). *Caryapollenites*, similar to modern *Carya* (hickory) pollen, is common in terrestrial Paleocene deposits from western Scotland interpreted as bogs (Jolley et al., 2009), and many may have been adapted as primary colonisers in wet substrates (Jolley and Whitham 1994). Its association with moss and fern spores in PA4 indicates a possible predominance of bog environments, consistent with high regional precipitation and poorly drained acidic bedrock. As bogs receive water and nutrients directly from precipitation (Price and Waddington, 2000), the change from PA3 to PA4 appears to be the result of climatic change (e.g. precipitation and temperature) rather than sea level.

Other changes include a further increase in the proportion of angiosperm over gymnosperm pollen (largely a reduction in pine), consistent with previous observations from the Lomonosov Ridge (Sluijs et al., 2006) and Bighorn Basin, Wyoming (Smith et al., 2007).

4.6. Environmental and climatic changes before the CIE

The North Sea Basin became restricted before the PETM, as indicated by a lithologic change throughout the North Sea Basin (the Lista/Sele boundary, Figs. 2, 9) and the presence of low oxygen benthic foraminifera in 22/10a-4 (Knox, 1996). This relative sea level fall was recognised as a lithologic change from marine to lagoonal/shallow marine facies in southeast England (Knox et al., 1994) and considered to have been largely the result of major regional uplift possibly by ~100 m (Knox, 1996). The documented major tectonic uplift at the Lista/Sele boundary best explains the concomitant increase in terrigenous input to 22/10a-4 (increase C/N ratio, Fig. 9c), domination of lowland swamp vegetation (swamp conifers, Fig. 8), and the lowering of surface water salinity (dinoflagellate cysts, Fig. 9d) as the basin became more restricted. The gradual lowering of surface water salinity and increase in the C/N ratio from ~4 m before the CIE onset at 22/10a-4 (above ~2618 m, Fig. 9), therefore, may also be the result of further uplift and restriction of the North Sea Basin, bringing the coastline and transported terrestrial material into closer proximity to the centrally-located 22/10a-4, although there is no documented sea level fall in the North Sea preceding the CIE (Knox, 1996; Powell et al., 1996). The increase in kaolinite in this scenario would be coincidental, perhaps having been formed after (>1 myr, Thiry and Dupuis, 2000) the uplift at the List/Sele boundary. As the North Sea Basin was proximal to the NAIP during the Paleocene/Eocene (Figs. 1, S1), regional uplift before the CIE would most likely be related to intrusive activity west of the North Sea Basin, restricting deep-water connections between the North Sea and Atlantic Ocean. Uplift and restriction is therefore consistent with the hypothesis that a mantle-derived magmatic intrusion of organic-rich sediments occurred in the NE Atlantic before the CIE, triggering atmospheric methane release (Svensen et al., 2004).

Alternatively, an increase in regional precipitation could have caused elevated terrestrial runoff (C/N ratios, kaolinite) and lower surface water salinity above ~2618 m before the CIE onset (Fig. 9). The North Sea Basin surrounding landmasses were within the northern rain belt (the southern boundary today is ~40°N), which would have experienced elevated precipitation if the global hydrologic cycle became enhanced (Pagani et al., 2006b; Schmitz et al., 2001). This scenario would be consistent with a gradual increase in the global hydrologic cycle before the CIE, perhaps from gradual warming, which was hypothesised to have triggered ocean circulation changes, methane hydrate destabilisation, and global carbon release at the CIE (Bice and Marotzke, 2002).

We note however that there is currently no evidence for an enhanced hydrologic cycle well-before the CIE in other regions.

Our results provide the first evidence that the North Sea became stratified from ~4 m before the CIE onset (above ~2618 m, Fig. 9). This is significant as Nisbet et al. (2009) hypothesised that the proximal Kilda Basin become stratified and anoxic at before the CIE, allowing significant build-up of methane and CO_2 at depth. They proposed that overturning of this basin could have released greenhouse gases and triggered the CIE, although there is currently no direct evidence as marine records from the Kilda Basin remain rare (Nisbet et al., 2009). Our North Sea records likely indicate enhanced stratification also of the proximal and linked Kilda Basin before the CIE (Fig. 1). Evidence for the linkage of the North Sea, Kilda and Arctic Basins comes from the coincident onset of *A. augustum* and laminated sediments at the CIE onset in sections from the North Sea (this study), Spitsbergen (Harding et al., 2011) and Lomonosov Ridge (Sluijs et al., 2006). Although our results evidence a probable stratified Kilda Basin before the CIE, proxies for overturning are now needed to further test the Kilda basin hypothesis.

The brief peak in *Apectodinium*, AOM and low salinity dinoflagellate cysts (*Deflandrea*) at ~2617.4 m (Fig. 7) indicates a sporadic episode of surface water freshening/eutrophication before the CIE, which is best explained by an increase in regional precipitation due to its rapid nature. A coincident reduction in *I. hiatus* swamp conifers indicates possible disturbance of nearby coastal environments possibly from flooding (see Section 4.5). An associated reduction in $\delta^{13}C$ may have been caused by stratification of the North Sea from an enhanced halocline, trapping ^{12}C-enriched organic carbon at depth. This scenario may also explain the other peaks in *Apectodinium* at 2619.6 and 2614.7 m (although see Section 4.1).

4.7. Environmental and climatic changes within the CIE

The influx of *Apectodinium* (indicating the onset of the CIE) at the Paleocene-Eocene boundary in southeast England occurred in characteristically marine assemblages following non-marine deposition, and was interpreted as an indication of relative sea level rise associated with a transgression immediately preceding a maximum flooding surface within the early PETM (Powell et al., 1996). Sea level rise at the CIE onset was also recorded in Arctic Spitsbergen (Harding et al., 2011). The large reduction lowland swamp pollen at the CIE onset of 22/10a-4 indicates a possible change in regional sea level. As there is no direct evidence for regional uplift causing further restriction of the North Sea Basin associated with the CIE onset, increased regional precipitation may have been the major cause of increased C/N ratios (elevated fluvial runoff) and dinoflagellate cyst changes (lower surface water salinity) at and following the CIE onset in 22/10a-4 (Fig. 9).

Although our records cannot distinguish changes to seasonality, which may have increased in the Pyrenees (Schmitz et al., 2001; Schmitz and Pujalte, 2007) and mid-latitude North America (Wing et al., 2005; Kraus & Riggins 2007), they are consistent with an overall increase in precipitation over NW Europe associated with the CIE. Our records therefore provide evidence consistent with the hypothesis that a northward migration of storm tracks occurred from an intensified hydrologic cycle as a result of global warming at the PETM, proposed by Pagani et al. (2006b) to explain elevated Arctic runoff during the PETM. As Pagani et al. (2006b) could not fully resolve the CIE onset due to incomplete core recovery, our records provide evidence that an enhanced hydrologic cycle may have occurred in approximate concert with global carbon release at the CIE onset. Alternatively, uplift of the NAIP could have caused further restriction and stratification of the North Sea. The reduction in %low salinity dinoflagellate cysts and the C/N ratio above ~2609 m (Fig. 9) may indicate a reduction in precipitation over NW Europe, or more likely tectonic subsidence causing the basin to become less restricted.

5. Conclusions

A negative carbon isotope excursion of ~5‰ has been identified from $\delta^{13}C_{TOC}$ and $\delta^{13}C_{AOM}$ in an expanded Paleocene-Eocene boundary section from the central North Sea Basin. Palynological (dinoflagellate cyst, pollen and spore assemblages) and sedimentologic (C/N ratios and kaolinite) evidence indicates major changes occurred to marine and terrestrial environments in NW Europe both preceding and over the CIE. Enhanced halocline stratification and terrigenous input from ~4 m before the CIE may indicate tectonic uplift, supporting hypotheses for NAIP volcanism as a trigger for the CIE (Svensen et al., 2004), and/or increased terrigenous runoff and regional precipitation, supporting hypotheses of an enhanced hydrologic cycle triggering carbon release (Bice and Marotzke, 2002). A peak in *Apectodinium* before the CIE is interpreted as an ephemeral increase in terrestrial runoff causing local eutrophication. Further enhanced halocline stratification and terrigenous input at and immediately after the CIE onset, coupled with evidence for sea level rise in coastal areas, indicates possible increased regional precipitation over NW Europe. At this location (paleolatitude ~54°N) increased precipitation lends evidence to the hypothesis that a poleward migration of storm tracks from an enhanced hydrologic cycle resulted from global warming during the PETM (Pagani et al., 2006b).

Palynological spore and pollen assemblages from 22/10a-4 record a rapid major shift in vegetation at the CIE onset, with dominant swamp conifer communities and pines largely replaced by generalist taxa including fern and fungal spores and various angiosperms. A change to a dominance of angiosperm over gymnosperm pollen at the CIE onset has previously been recorded

in the Arctic (Sluijs et al., 2006) and Wyoming (Smith et al., 2007). The rapid reduction in lowland gymnosperm swamp pollen at the CIE onset may indicate a change in lowland topography from sea level alteration, and/or it may indicate ecologic changes driven by climate (e.g. precipitation and temperature). This floral shift occurs simultaneously with the first persistent appearance of *Apectodinium* in 22/10a-4, indicating that precursor CIE ecologic changes identified in NW Atlantic and North Sea marine records (Sluijs et al., 2007) had a terrestrial counterpart in the North Sea region. Longer term vegetation changes after the CIE onset indicate a move towards more diverse generalist angiosperm and pteridophyte communities (dominance of *Caryapollenites*, fern and fungal spores). The pollen and spore assemblages therefore indicate that long term ecologic change occurred in NW Europe probably in response to temperature and hydrologic changes during the PETM, but that the most dramatic changes recorded in 22/10a-4 occurred abruptly at the onset of the CIE.

Supplementary materials related to this article can be found online at doi: xxx

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Fig. 1. Paleogeographic reconstruction of the continents at ~54 Ma (Mosar et al., 2002), showing the location of core 22/10a-4 (this study), 30/14-1 (Sluijs et al., 2007), 208/19-1 (Mudge and Bujak, 2001), Spitsbergen Central Basin (Harding et al., 2011) and M0002A (Pagani et al., 2006b). Grey shading indicates regions of volcanism affected by the North Atlantic Igneous Province (NAIP).

Fig. 2. Lithologic column of borehole 22/10a-4 showing Gamma Ray Attenuation (GRA), stratigraphic and lithologic subdivisions, and previously published low resolution palynological and sedimentologic results (Knox, 1996). Depth is logging depth (all other depths in this paper are core depth). The GRA spike and onset of abundant *Apectodinium* was previously used to identify the position of the CIE (Knox, 1996). The extent of core analysed in this study is also shown. Yellow = sandstone; blue = massive grey claystone; light grey = intermittently bedded grey claystone; dark grey = well-bedded grey mudstone (Knox, 1996).

Fig. 3. Carbon isotopic results of total organic matter ($\delta^{13}C_{TOC}$) and amorphous organic matter ($\delta^{13}C_{AOM}$), against core 22/10a-4 lithology and *Apectodinium* spp. (%). Blue = bulk rock $\delta^{13}C_{TOC}$; black = $\delta^{13}C_{AOM}$; solid red symbols = bulk rock $\delta^{13}C_{TOC}$ from samples with <30% wood/plant tissue (determined from palynological residue of the sample); open red symbols = bulk rock $\delta^{13}C_{TOC}$ from samples with >30% wood/plant tissue. The first appearance of *Apectodinium augustum* identifies the PETM in the North Sea (Bujak and Brinkhuis, 1998), and the first negative shift in $\delta^{13}C$ identifies the approximate position of the CIE onset and the Paleocene-Eocene boundary. Values shaded at 2614.7 and 2619.6 m are considered possible outliers based on statistical analysis of the palynological residues (see Section 4.1). Lithologic column shows position of sand intervals (yellow), claystone intervals (brown; predominantly laminated claystone, dark brown), and ash layers (pink).

Fig. 4. Correspondence analysis for (a) dinoflagellate cysts, and (b) spores and pollen, from each sample (total counts per gram) in core 22/10a-4. First three axes are plotted against depth, with % variance indicated. Solid vertical lines indicate mean, dashed lines indicate standard deviation ($\pm 1\sigma$). Values outside $\pm 1\sigma$ are considered statistically meaningful and used to define pollen assemblages (PA) and dinoflagellate cyst assemblages (DA). The two circled values indicate samples that fall outside PA2, and either represent ephemeral episodes of vegetation changes to post-CIE onset type conditions, or possible misplaced samples during drilling operations core handling. Analysis carried out using the software of Hammer et al. (2005).

Fig. 5. Correspondence analysis (first two axes) for (a) spores and pollen species (total counts per gram, axis 1 = 39% of total variance, axis 2 = 17% of total variance), and (b) dinoflagellate cysts (total counts per gram, axis 1 = 31% of total variance, axis 2 = 11% of total variance), in core 22/10a-4. Symbols indicate depths for each pollen assemblage (PA) and dinoflagellate cyst assemblage (DA), and their correspondingly most associated palynomorph species. Marked depths (red) indicate the samples with pre-CIE peaks in *Apectodinium* and $\delta^{13}C_{TOC}$. Samples at 2619.60 m and 2614.71 m (see Fig. 4) plot close to post-CIE onset assemblages PA3 and PA4, and either represent ephemeral episodes of marine and vegetation changes to post-CIE onset type conditions, or possible misplaced samples during drilling operations core handling. Analysis carried out using the software of Hammer et al. (2005).

Fig. 6. Spore and pollen abundance and diversity data from 22/10a-4. (a) Lithologic formations, (b) bulk total organic carbon (TOC) stable isotopes, (c) total abundance of all pollen/spores, (d) total number of pollen and spore species recorded in each sample, (e) diversity index Shannon H (calculated using the software of Hammer et al., 2005) which takes into account the number of specimens in each sample in addition to species, (f) diversity index Fisher alpha (calculated using the software of Hammer et al., 2005) which takes into account the number of specimens in each sample in addition to species, (g) total number of pteridophyte spore species recorded in each sample, (h) total number of angiosperm pollen species recorded in each sample.

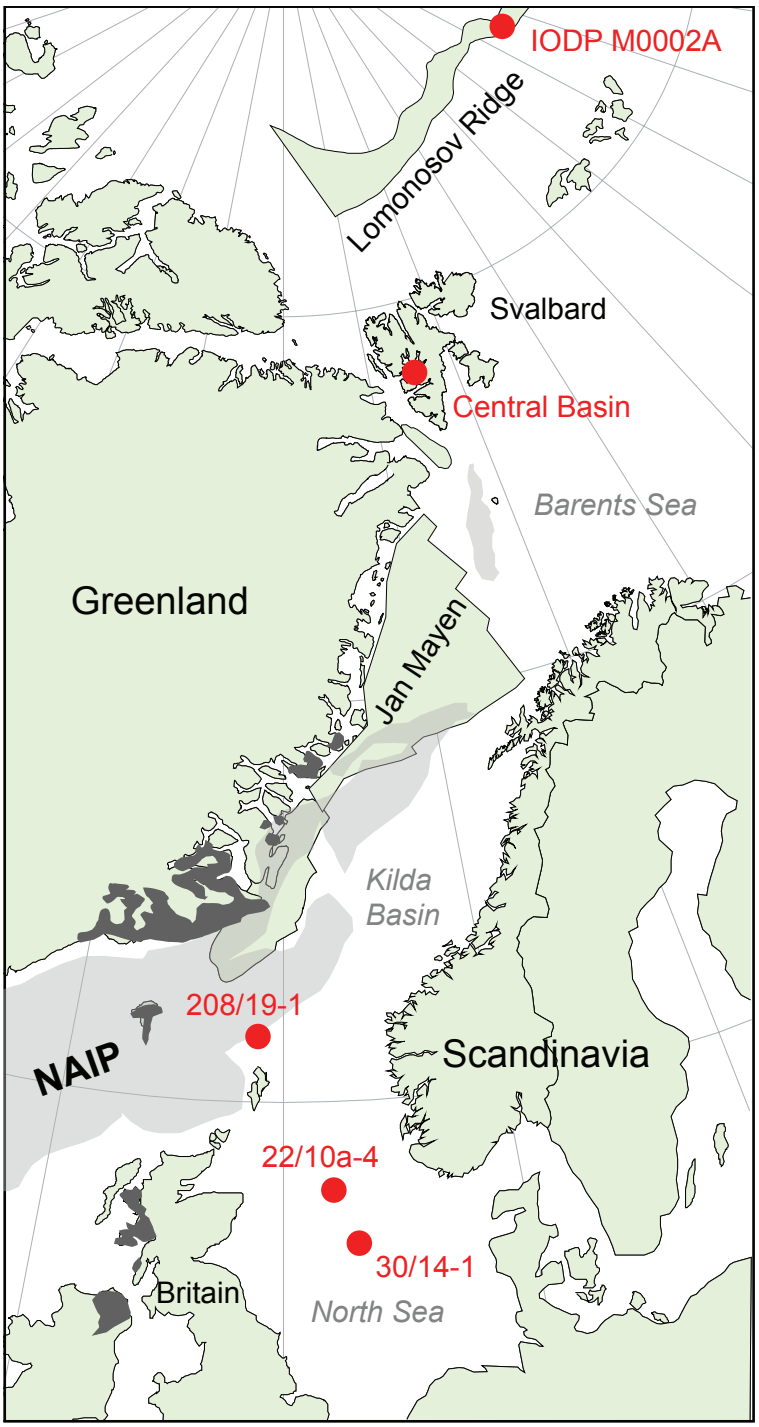
Fig. 7. Sedimentologic, isotopic and micropaleontologic data for North Sea core 22/10a-4 (this study). Blue shaded area represents time of lowest surface water salinity (from dinoflagellate cysts) and inferred halocline stratification. (a) Lithologic formations, (b) bulk total organic carbon (TOC) isotopes, (c) dinoflagellate cysts tolerant of low salinity and high nutrients as percentage of all dinoflagellate cysts with known salinity preference, (d) benthic foraminifera (agglutinated), (e) amorphous organic matter (AOM) as a percentage of all palynomorphs, (f) position of finely laminated sediment, (g) % dinoflagellate cysts to spores and pollen, (h) *Apectodinium* spp. as percentage of all dinoflagellate cysts, (i-l) dinoflagellate cysts likely tolerant to low salinity and high nutrient surface water, (m-o) dinoflagellate cysts representative of typical marine salinity surface water, (p) dinoflagellate cyst assemblages (DA).

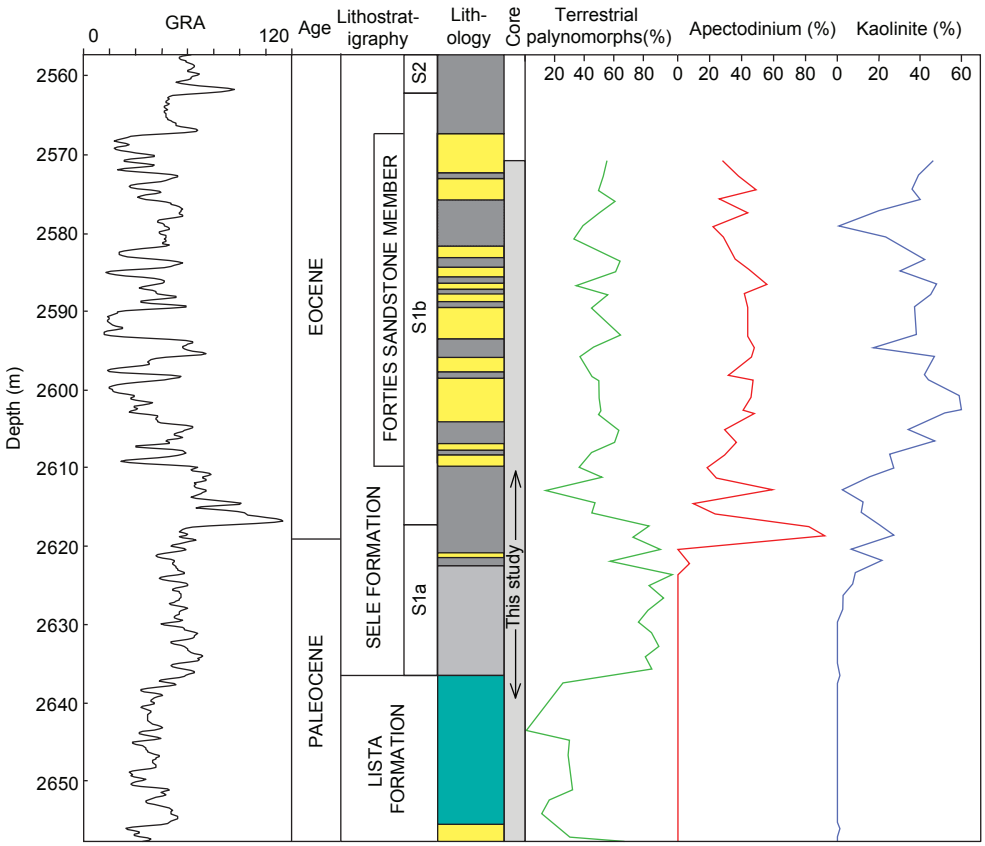
Fig. 8. Sedimentologic, isotopic and micropaleontologic data for North Sea core 22/10a-4 (this study). (a) Lithologic formations, (b) bulk total organic carbon (TOC) isotopes, (c) dinoflagellate cysts tolerant of low salinity and high nutrients as percentage of all dinoflagellate cysts with known

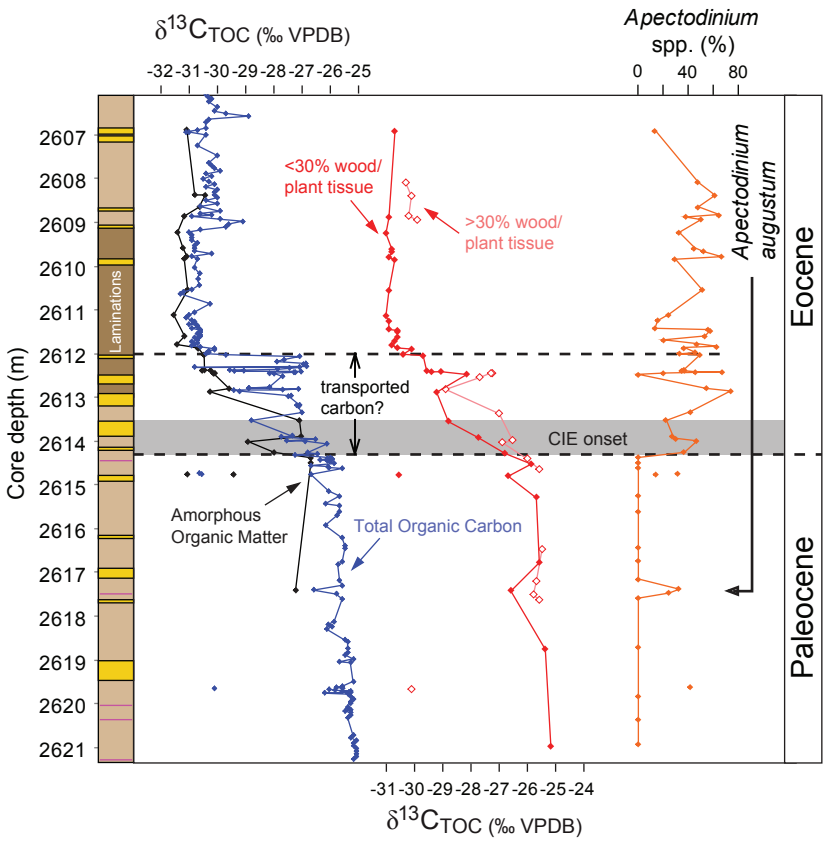
salinity preference, (d) angiosperm pollen as percentage of all angiosperm and gymnosperm pollen, (e) total abundance of all pollen/spores, (f-g) pre-CIE pollen, (h-l) pollen and spores prevalent in the CIE, (m) fungal spores, (n) pollen/spore assemblages (PA).

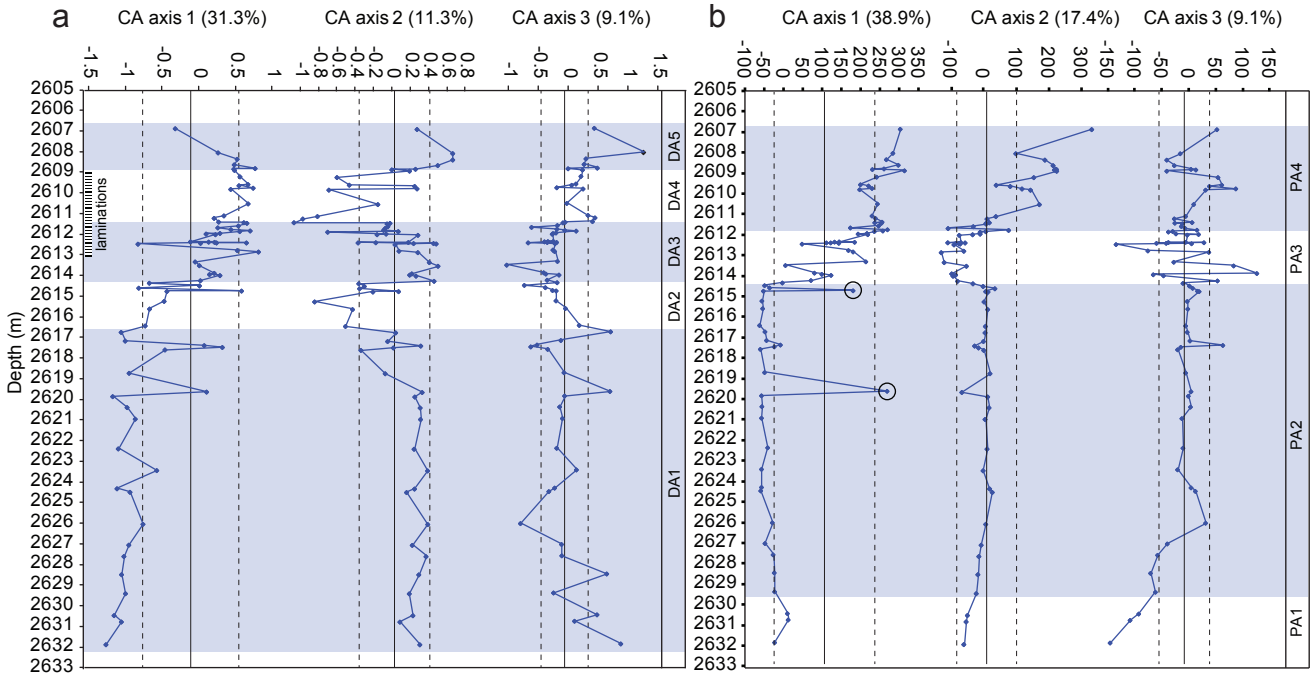
Fig. 9. Geochemical, micropaleontologic and sedimentologic data for North Sea core 22/10a-4. (a) Lithologic formations, (b) bulk total organic carbon (TOC) isotopes, (c) carbon/nitrogen ratio, (d) dinoflagellate cysts tolerant of low salinity and high nutrients as percentage of all dinoflagellate cysts with known salinity preference, (e) percentage kaolinite (from Knox, 1996).

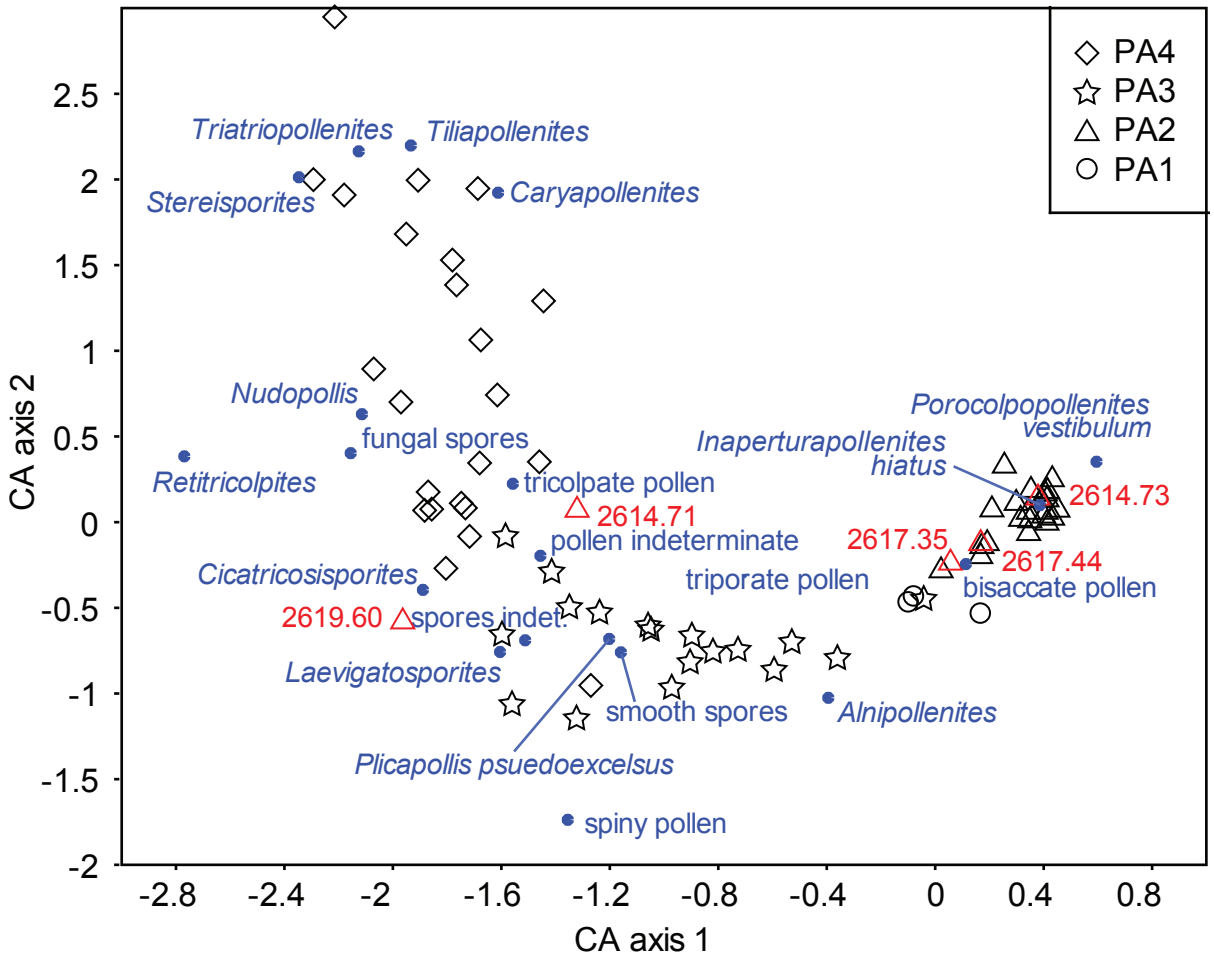
Fig. 10. Atomic C/N ratio distribution from all studied samples (Fig. S4). Shaded areas show typical values for modern algae and land plants (Meyers, 1997).

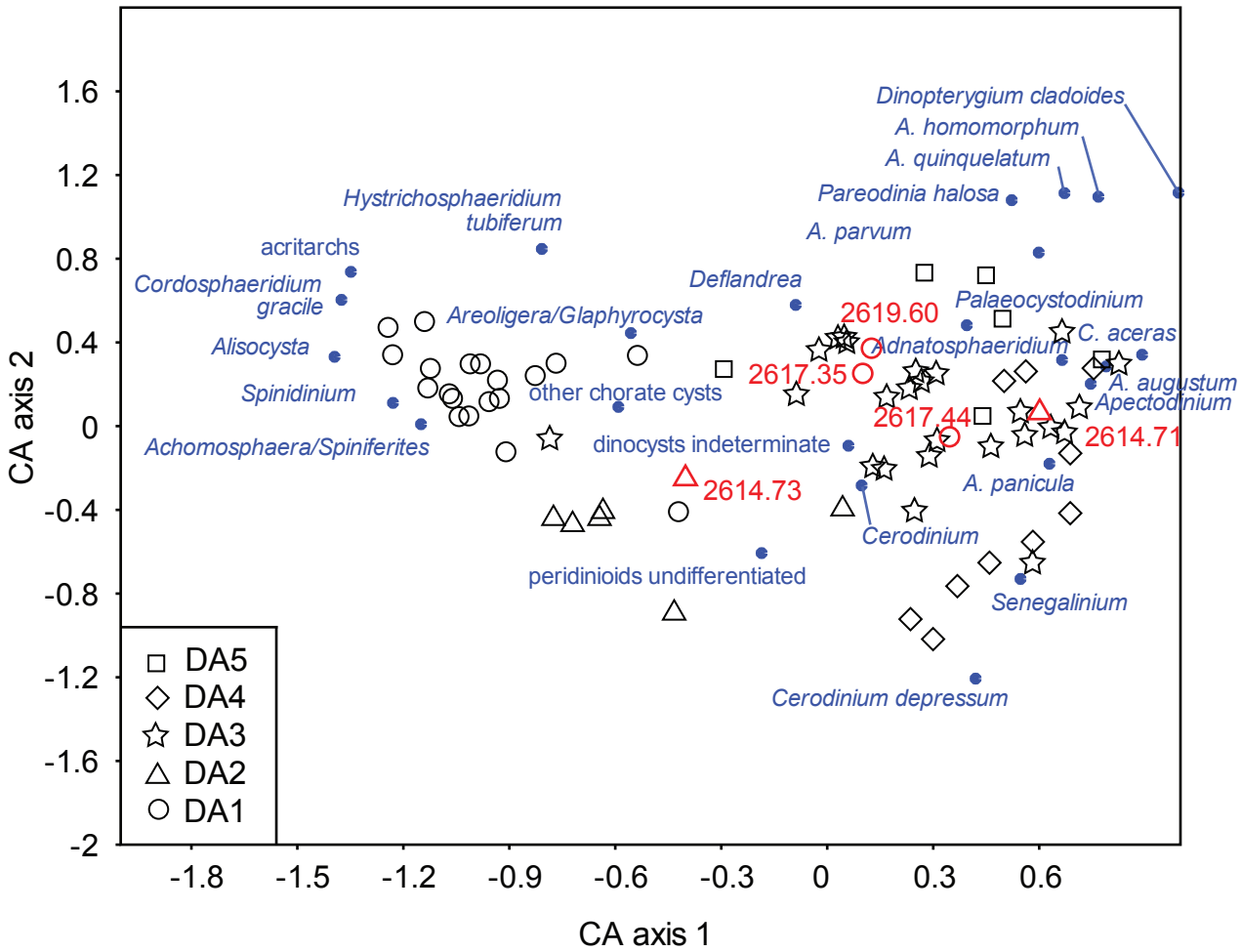

IODP M0002A
Lomonosov Ridge
Svalbard
Central Basin
Barents Sea
Greenland
Jan Mayen
Kilda
Basin
208/19-1
NAIP
Scandinavia
22/10a-4
30/14-1
Britain
North Sea

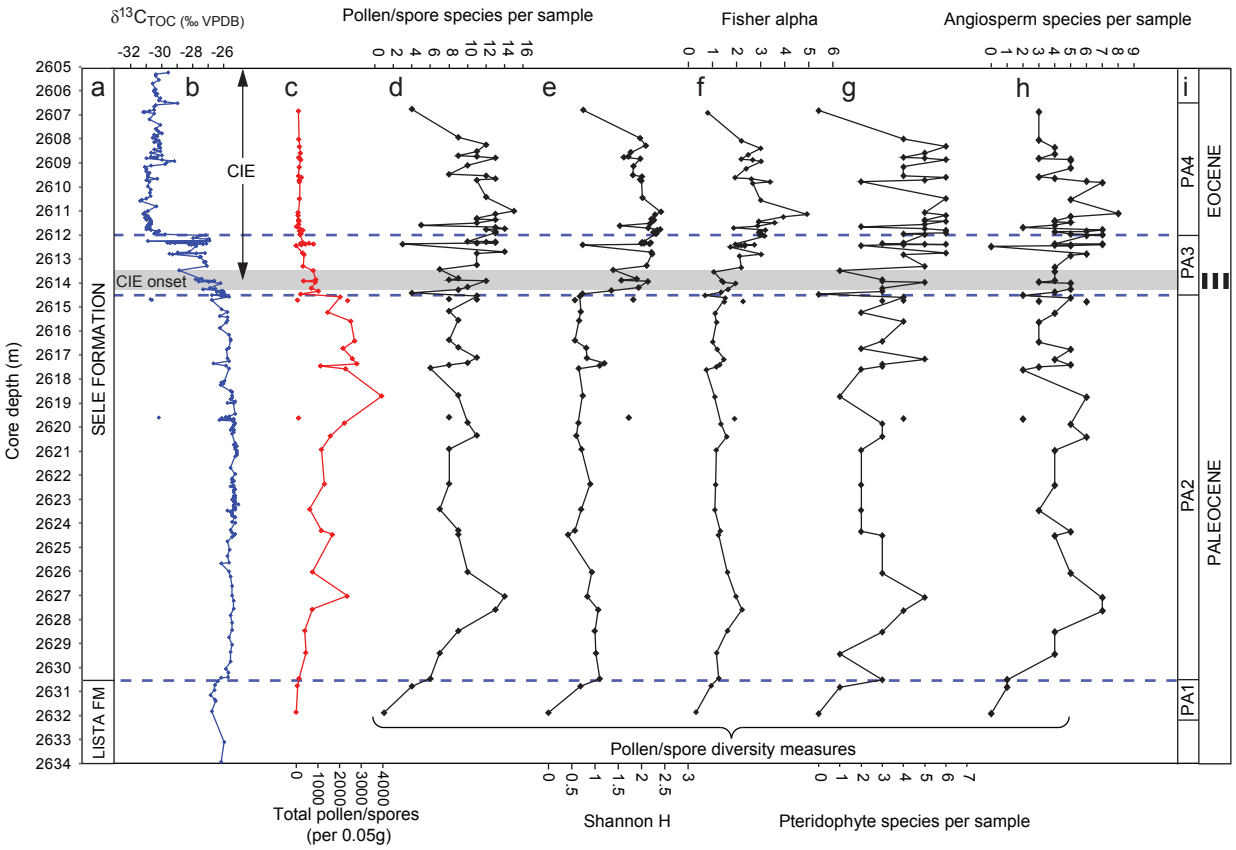

GRA
0
120
Age
Lithostrat-igraphy
Lith-ology
Core
Terrestrial palynomorphs(%)
Apectodinium (%)
Kaolinite (%)
20 40 60 80
0 20 40 60 80
0 20 40 60
Depth (m)
2560
2570
2580
2590
2600
2610
2620
2630
2640
2650
EOCENE
PALEOCENE
SELE FORMATION
FORTIES SANDSTONE MEMBER
LISTA FORMATION
S2
S1b
S1a
This study

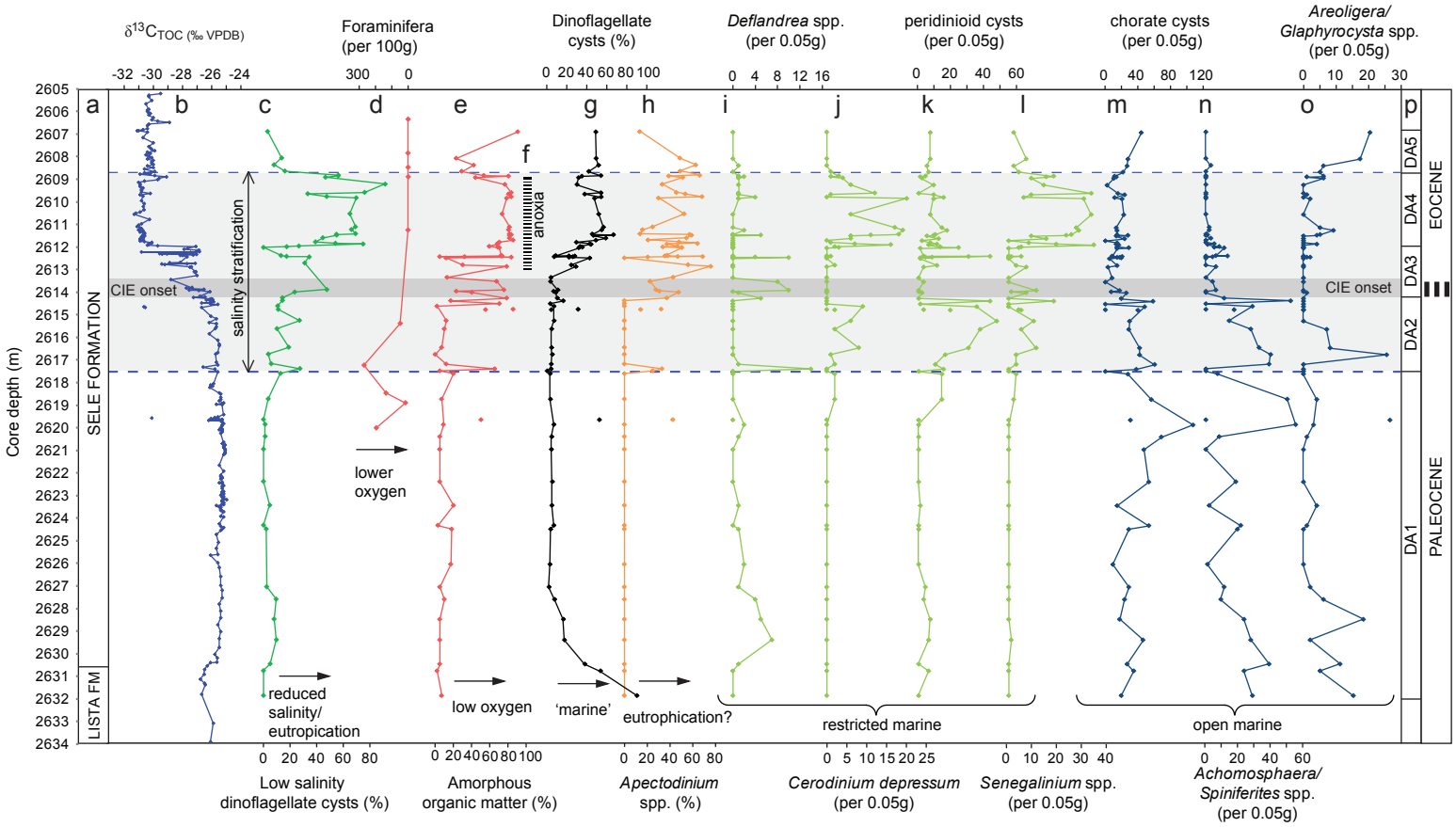

δ13CTOC (‰ VPDB)
-32 -31 -30 -29 -28 -27 -26 -25
Apectodinium
spp. (%)
0 40 80
Core depth (m)
2607
2608
2609
2610
2611
2612
2613
2614
2615
2616
2617
2618
2619
2620
2621
Laminations
<30% wood/
plant tissue
>30% wood/
plant tissue
transported
carbon?
CIE onset
Amorphous
Organic Matter
Total Organic Carbon
Apectodinium
augustum
Eocene
Paleocene
-31 -30 -29 -28 -27 -26 -25 -24
δ13CTOC (‰ VPDB)

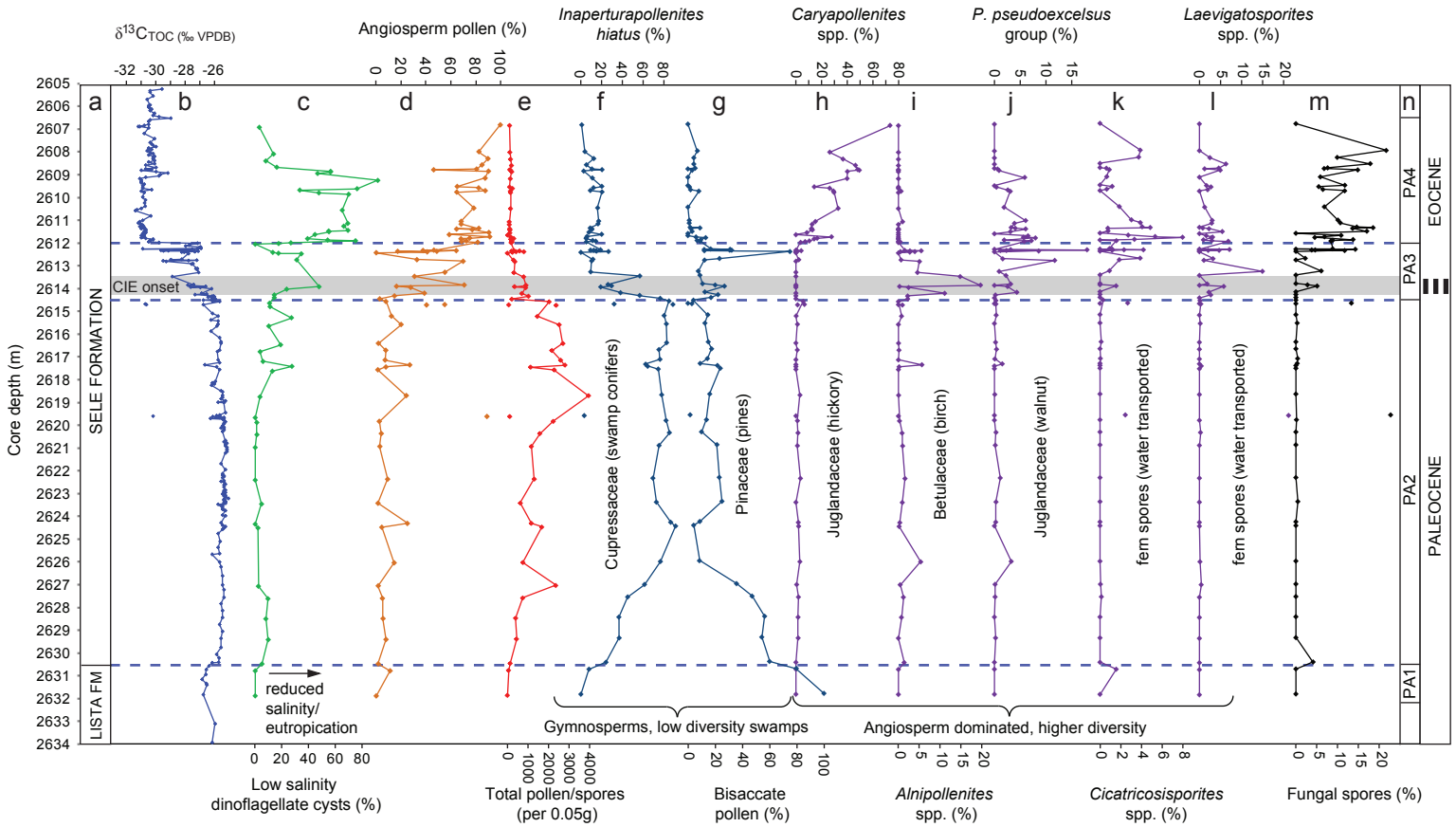

a
CA axis 1 (31.3%)
CA axis 2 (11.3%)
CA axis 3 (9.1%)
Depth (m)
laminations
DA5
DA4
DA3
DA2
DA1
b
CA axis 1 (38.9%)
CA axis 2 (17.4%)
CA axis 3 (9.1%)
PA4
PA3
PA2
PA1

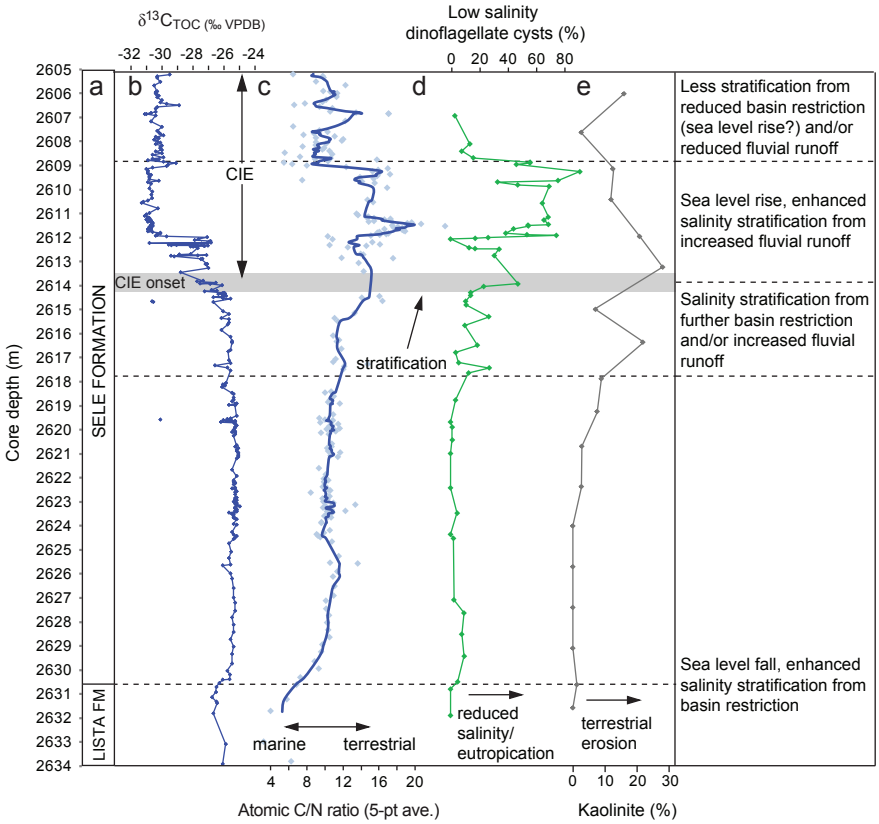

PA4
PA3
PA2
PA1
Triatriopollenites
Tiliapollenites
Stereisporites
Caryapollenites
Nudopollis
fungal spores
Retitricolpites
tricolpate pollen
2614.71
pollen indeterminate
Cicatricosisporites
2619.60
spores indet.
Laevigatosporites
smooth spores
Plicapollis psuedoexcelsus
spiny pollen
Porocolpopollenites vestibulum
Inaperturapollenites hiatus
2614.73
2617.35
2617.44
triporate pollen
bisaccate pollen
Alnipollenites
CA axis 2
CA axis 1
2.5
2
1.5
1
0.5
0
-0.5
-1
-1.5
-2
-2.8
-2.4
-2
-1.6
-1.2
-0.8
-0.4
0
0.4
0.8

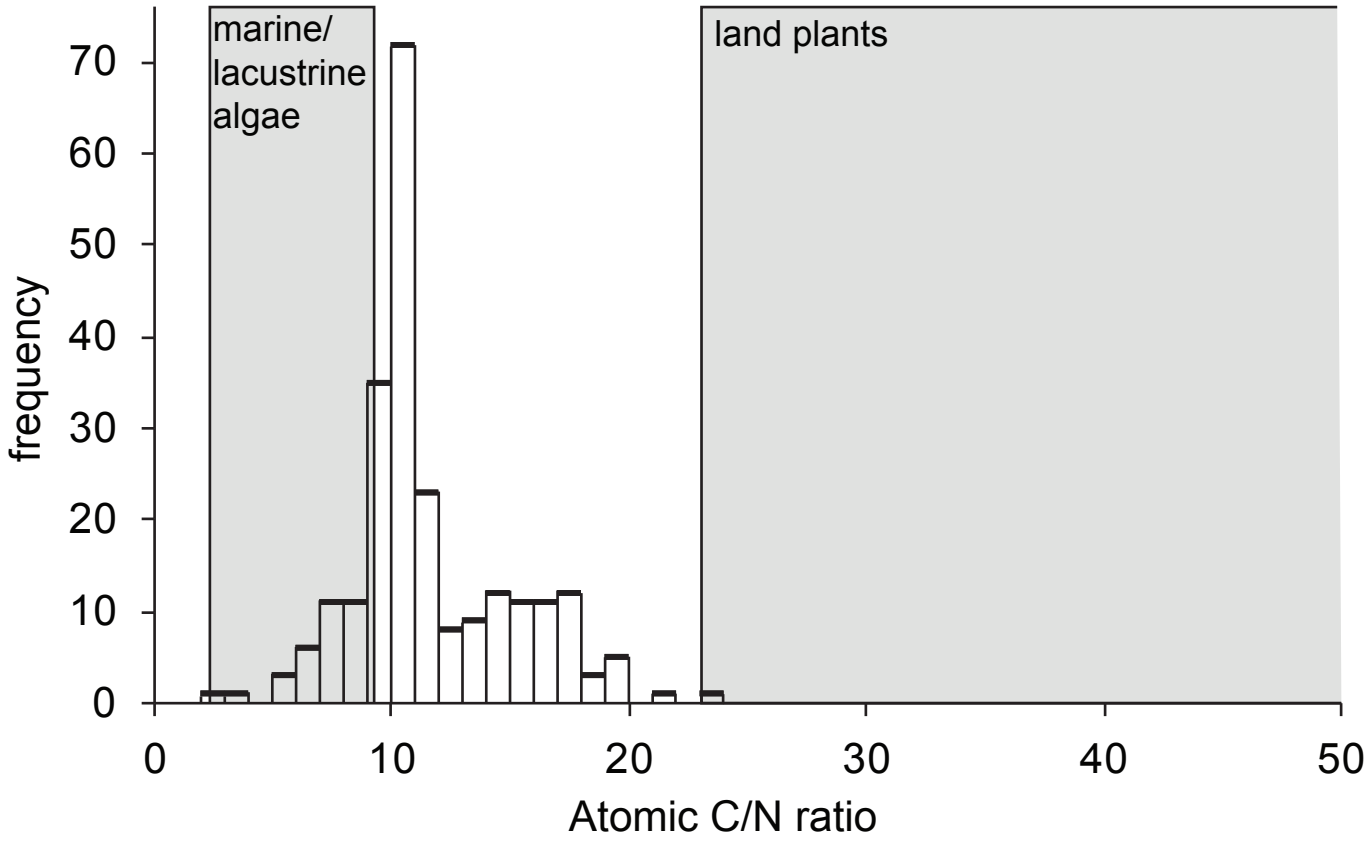

Dinopterygium cladoides
A. homomorphum
A. quinquelatum
Pareodinia halosa
Hystrichosphaeridium tubiferum
A. parvum
acritarchs
Cordosphaeridium gracile
Deflandrea
Palaeocystodinium
Areoligera/Glaphyrocysta
2619.60
C. aceras
Alisocysta
Adnatosphaeridium
2617.35
A. augustum
Spinidinium
other chorate cysts
Apectodinium
2617.44
2614.71
Achomosphaera/Spiniferites
dinocysts indeterminate
2614.73
A. panicula
Cerodinium
peridinioids undifferentiated
Senegalinium
Cerodinium depressum
DA5
DA4
DA3
DA2
DA1
CA axis 2
CA axis 1
1.6
1.2
0.8
0.4
0
-0.4
-0.8
-1.2
-1.6
-2
-1.8
-1.5
-1.2
-0.9
-0.6
-0.3
0
0.3
0.6


δ13CTOC (‰ VPDB)
Pollen/spore species per sample
Fisher alpha
Angiosperm species per sample
Core depth (m)
SELE FORMATION
LISTA FM
CIE
CIE onset
EOCENE
PALEOCENE
PA4
PA3
PA2
PA1
Pollen/spore diversity measures
Total pollen/spores (per 0.05g)
Shannon H
Pteridophyte species per sample


δ13CTOC (‰ VPDB)
Foraminifera (per 100g)
Dinoflagellate cysts (%)
Deflandrea spp. (per 0.05g)
peridinioid cysts (per 0.05g)
chorate cysts (per 0.05g)
Areoligera/ Glaphyrocysta spp. (per 0.05g)
Core depth (m)
SELE FORMATION
LISTA FM
CIE onset
salinity stratification
anoxia
lower oxygen
reduced salinity/ eutropication
low oxygen
'marine'
eutrophication?
restricted marine
open marine
DA5
DA4
DA3
DA2
DA1
EOCENE
PALEOCENE
Low salinity dinoflagellate cysts (%)
Amorphous organic matter (%)
Apectodinium spp. (%)
Cerodinium depressum (per 0.05g)
Senegalinium spp. (per 0.05g)
Achomosphaera/ Spiniferites spp. (per 0.05g)


δ13CTOC (‰ VPDB)
Angiosperm pollen (%)
Inaperturapollenites hiatus (%)
Caryapollenites spp. (%)
P. pseudoexcelsus group (%)
Laevigatosporites spp. (%)
Core depth (m)
SELE FORMATION
LISTA FM
CIE onset
Cupressaceae (swamp conifers)
Pinaceae (pines)
Juglandaceae (hickory)
Betulaceae (birch)
Juglandaceae (walnut)
fern spores (water transported)
fern spores (water transported)
reduced salinity/ eutropication
Gymnosperms, low diversity swamps
Angiosperm dominated, higher diversity
PA4
PA3
PA2
PA1
EOCENE
PALEOCENE
Low salinity dinoflagellate cysts (%)
Total pollen/spores (per 0.05g)
Bisaccate pollen (%)
Alnipollenites spp. (%)
Cicatricosisporites spp. (%)
Fungal spores (%)


δ13CTOC (‰ VPDB)
-32 -30 -28 -26 -24
Low salinity dinoflagellate cysts (%)
0 20 40 60 80
a
b
c
d
e
CIE
CIE onset
stratification
SELE FORMATION
LISTA FM
Core depth (m)
2605
2606
2607
2608
2609
2610
2611
2612
2613
2614
2615
2616
2617
2618
2619
2620
2621
2622
2623
2624
2625
2626
2627
2628
2629
2630
2631
2632
2633
2634
marine
terrestrial
reduced salinity/ eutropication
terrestrial erosion
4 8 12 16 20
Atomic C/N ratio (5-pt ave.)
0 10 20 30
Kaolinite (%)
Less stratification from reduced basin restriction (sea level rise?) and/or reduced fluvial runoff
Sea level rise, enhanced salinity stratification from increased fluvial runoff
Salinity stratification from further basin restriction and/or increased fluvial runoff
Sea level fall, enhanced salinity stratification from basin restriction


marine/
lacustrine
algae
land plants
frequency
70
60
50
40
30
20
10
0
0
10
20
30
40
50
Atomic C/N ratio

Marine and terrestrial environmental changes in NW Europe preceding carbon release at the Paleocene-Eocene transition

Supplementary Material

1. Isotope changes at the Paleocene-Eocene boundary

Bulk sediment total organic carbon (TOC) isotopes ($\delta^{13}C_{TOC}$) in core 22/10a-4 shift from consistently > –27‰ to < –30‰ within the interval ~2614 to 2612 m core depth (Fig. 3). The ~5‰ excursion in $\delta^{13}C_{TOC}$ is comparable in magnitude to organic carbon $\delta^{13}C$ records of the PETM from the Lomonosov Ridge (Sluijs et al., 2006), Wilson Lake (Sluijs et al., 2007) and the North Sea (Sluijs et al., 2007), but larger than those from Spitsbergen (Harding et al., 2011) and Bass River (Sluijs et al., 2007). These differences in magnitude of the CIE may be partly due to simultaneous changes in proximal terrestrial vegetation type and/or shifts in marine plankton assemblages (Schouten et al., 2007; Handley et al., 2008). The first occurrence of the dinoflagellate marker species *Apectodinium augustum*, diagnostic of the Paleocene-Eocene boundary in the North Sea (Bujak and Brinkhuis, 1998; Sluijs et al., 2007), occurs in 22/10a-4 at ~2617 m (Fig. 3).

In contrast to the low variability of $\delta^{13}C_{TOC}$ preceding (SD = 0.36‰, n = 106) and following (SD = 0.52‰, n = 117) the onset of the CIE, $\delta^{13}C_{TOC}$ values fluctuate by up to 4‰ within the interval associated with the CIE onset at ~2614 to 2612 m (dashed lines in Fig. 3). These $\delta^{13}C_{TOC}$ fluctuations are not seen in other marginal marine records of the CIE (e.g. Harding et al., 2011), and are unlikely to be representative of the atmospheric carbon pool as the estimated speed of global carbon sequestration is too slow (Dickens et al., 1995; Bains et al., 2000; Zeebe et al., 2009). As the $\delta^{13}C_{TOC}$ signature of organic carbon (C_{org}) from the marine and terrestrial realms can be different, the fluctuations in $\delta^{13}C_{TOC}$ observed at the onset of the CIE likely reflect varying contributions to C_{org} deposited within the North Sea Basin rather than changes within the global carbon system. The palynological results (from samples containing sufficient material for analysis; red symbols in Fig. 3) indeed show that %wood/plant tissue varies from 10% to 88% throughout this interval (Fig. S3). We found that the samples containing >30% wood/plant tissue (empty red symbols in Fig. 3) in the palynological residues have consistently higher $\delta^{13}C_{TOC}$ values than those with less wood/plant tissues (solid red symbols). These heavier values may indicate the presence of material reworked from older sediments with pre-CIE $\delta^{13}C$ values, and if the hydrological

cycle was enhanced, excavation and re-deposition of latest Paleocene carbon might be expected. An equally valid alternate hypothesis is that they may partially reflect the high abundance of wood/plant tissue with potentially more positive $\delta^{13}C_{TOC}$ values (terrestrial lignite deposit $\delta^{13}C_{TOC}$ from southeast England ranged from –24.5‰ before the CIE to –27‰ after the CIE, Collinson et al., 2007). Note here we are essentially arguing for more negative marine $\delta^{13}C_{TOC}$ values than is typical for marine C_{org} (e.g. North Atlantic New Jersey Margin –27‰, Sluijs et al., 2007). Usually Paleogene marine C_{org} exhibits similar or more positive $\delta^{13}C_{TOC}$ than terrestrial, but in the restricted North Sea (22/10a-4, see also Sluijs et al., 2007) a build up of nutrients may have lowered seawater $\delta^{13}C$ to values below those typically found in the North Atlantic during the PETM – a hypothesis supported by $\delta^{13}C_{AOM}$, where AOM is probably of marine origin (see below). We note similarly low $\delta^{13}C_{TOC}$ PETM values in marine sections from Spitsbergen (Harding et al., 2011) and the Lomonosov Ridge (Sluijs et al., 2006) – water masses linked to the restricted North Sea (Fig. 1). Restriction of the Arctic–Kilda–North Sea basins (Fig. 3) is also supported by laminations, marking anoxia at the CIE onset in all three sections from the North Sea (this study), Spitsbergen (Harding et al., 2011) and the Lomonosov Ridge (Sluijs et al., 2006). The presence of transported C_{org} thus precludes an unambiguous interpretation of the rate of atmospheric carbon release at the CIE onset, but the initial negative $\delta^{13}C$ shift between 2614.3 and 2613.5 m (Fig. 3), which is especially pronounced in the $\delta^{13}C_{AOM}$ data, can be taken as marking the earliest onset of the CIE.

A significant component of the palynological residues is amorphous organic matter (AOM), which becomes abundant during the CIE both in our records (Fig. 7e), and in Arctic Spitsbergen (Fig. 1, Harding et al., 2011). A marine origin for the AOM is indicated by evidence for enhanced primary productivity during the CIE, namely an increase in the abundance of low salinity tolerant dinoflagellate cysts, likely reflecting increased fluvial nutrient supply which would have stimulated primary productivity, and mm-scale laminations (Fig. 7f) reflecting anoxic bottom waters resulting from high fluxes of phytoplankton detritus. The %dinoflagellate cysts (compared to spores and pollen, Fig. 7g) also increases over the interval of increased AOM, indicating that the AOM is associated with an increase in marine dinoflagellate cysts and/or a reduction in terrestrial palynomorph input. The weak orange/brown fluorescence of the AOM is also typical of marine plankton-derived organic matter (Tyson, 1995; Omura and Hoyanagi, 2004; Harding et al., 2011), as is its association with abundant pyrite inclusions (Tyson, 1995). Carbon isotopic measurements of palynological residues dominated by AOM ($\delta^{13}C_{AOM}$, black symbols in Fig. 3) have lighter

values than $\delta^{13}C_{TOC}$ in the majority of samples. If the AOM is of marine origin as we suggest, these lighter values appear to indicate that the North Sea was greatly enriched in ^{12}C compared to the New Jersey Shelf, North Atlantic ($\delta^{13}C$ measured on marine dinoflagellate cysts, Sluijs et al., 2007), which ranged from –22‰ before the CIE to –27‰ after. This apparent –3.5‰ offset between the North Sea and the North Atlantic could be due to the restricted nature of the North Sea Basin, with high productivity and apparent sluggish ventilation allowing the build-up of ^{12}C, and/or a function of different fractionations between marine algae/bacteria (AOM) and dinoflagellate cysts. Although at a lower temporal resolution, the $\delta^{13}C_{AOM}$ record (Fig. 3) supports $\delta^{13}C_{TOC}$ positioning of the CIE onset at ~2614 m.

2. Dinoflagellate paleoecology

***Achomosphaera/Spiniferites* spp.** was typically associated with open marine and hence normal marine salinity environments in the Paleogene of northern Italy (Brinkuis, 1994) and the New Jersey Shelf (Sluijs and Brinkhuis, 2009). Brinkuis (1994) points out that the analogous extant *Spiniferites ramosus* is present in a wide variety of settings in both oceans and marginal seas (not specialised to restricted environments) but may be more prevalent in shelf areas. *Spiniferites* was also recorded in high abundance in marine PETM sediments in Spitsbergen (Harding et al., 2011) and interpreted as a distal marine influence. Undifferentiated chorate cysts, although unidentifiable to a species level, have a similar morphology and distribution to *Achomosphaera/Spiniferites* spp. (Fig. 7n) and may therefore have a similar, normal marine, ecologic signal.

***Areoligera/Glaphyrocysta* spp.** were regarded as inhabiting inner neritic but not restricted localities (i.e. normal marine salinity) by Brinkhuis (1994), and are part of the un-restricted 'coastal taxa' of Pross and Brinkhuis (2005). *Achomosphaera/Spiniferites* spp. and *Areoligera/Glaphyrocysta* spp. are characteristic of the "*Hystrichosphaera* Association" and "*Areoligera* Association" from Paleogene sediments of Southeast England (Downie et al., 1971), suggested as representing open sea environments.

***Deflandrea* spp.** was regarded as a coastal/neritic taxon indicating high productivity and nutrient availability (Brinkhuis, 1994; Pross and Brinkhuis, 2005), occurs in association with low salinity dinoflagellate cysts and freshwater algae in the Arctic (Sluijs et al., 2008), and was associated with biosiliceous horizons interpreted as high productivity in the North Atlantic (Firth, 1996).

Cerodinium depressum and ***Senegalinium* spp.** are both thought to tolerate very low salinity conditions based on their co-occurrence with terrestrial spores and pollen and freshwater algae (including *Botryococcus* and *Pediastrum*) in the Arctic (Brinkhuis et al., 2006; Sluijs et al., 2006; Sluijs et al., 2008). *Senegalinium* may also have been a heterotrophic genus indicative of elevated nutrient levels (Sluijs and Brinkhuis, 2009), and occurs during the PETM in the Southern Ocean (Sluijs et al., 2011) and Arctic Spitsbergen (Harding et al., 2011) in association with elevated levels of river-transported spores and pollen.

Undifferentiated peridinoid cysts ecology is less well constrained (and they are therefore not included in the %low salinity dinoflagellate cysts, Fig. 7c), they may also be indicative of elevated nutrient and reduced salinity conditions due to their similarity in form to other Paleogene peridinoids and the modern heterotrophic dinoflagellate *Protoperidinium* (Powell et al., 1992).

The migration of ***Apectodinium* spp.** from low to high latitudes during Paleocene-Eocene warming suggests a certain temperature control on its distribution (Bujak and Brinkhuis, 1998; Crouch et al., 2003b; Sluijs and Brinkhuis, 2009). An overriding temperature control on *Apectodinium* has been questioned however by (1) canonical correspondence analysis of sites at the New Jersey margin which show it correlated only weakly with temperature (Sluijs and Brinkhuis, 2009), (2) its occurrence in the Arctic at SSTs of only 23°C during the PETM (Sluijs et al., 2006), and (3) its pre-CIE peak in abundance in the Southern Ocean prior to any significant global warming (Sluijs et al., 2011). There is evidence that the occurrence of *Apectodinium* is associated with increased nutrient supply during the PETM, as it peaked in abundance coincident with periods of enhanced terrigenous input off New Zealand (Crouch et al., 2003a), elevated kaolinite percentages off Tunisia (Crouch et al., 2003b), peak runoff (terrestrially-derived matter) off Spitsbergen (Harding et al., 2011) and peak productivity (dinoflagellate cyst accumulation rates) on the New Jersey margin (Sluijs et al., 2007). An affinity for high nutrient supply is also consistent with the association of *Apectodinium* with elevated abundances of low salinity/high nutrient tolerant dinoflagellate cysts, increased terrigenous runoff (increased C/N ratio and kaolinite) in 22/10a-4 (Fig. 9), although its occurrence does not precisely follow any one of these parameters. It has been suggested that *Apectodinium* did not thrive at very low salinities, as it was outcompeted by low salinity dinoflagellate cysts in the Arctic Ocean (Sluijs et al., 2006; 2007), and does not always follow the abundance patterns of low salinity tolerant dinoflagellate cysts in 22/10a-4 (Fig. 7c) or the New Jersey margin (Sluijs et al., 2007).

Although no single environmental condition could be responsible for the *Apectodinium* acme during the PETM in so many diverse environments (Crouch et al., 2003b; Sluijs et al., 2007), warming before and during the PETM may have opened the corridor for its migration and invasion of higher latitudes (Crouch et al., 2003b), where it appears to have outcompeted existing species that may have been less well adapted. Due to the sporadic migration of *Apectodinium* over the PETM, it may not be possible to use this species as a faithful indicator for specific environmental conditions at higher latitudes.

3. Foraminifera

Ten samples were analysed for foraminifera (Fig. 7d) in order to characterise benthic conditions. Abundant and low diversity assemblages were recovered between ~2620 and 2615 m (Table S2), containing an average of 130 specimens/g of dried sediment, and nine species of benthic foraminifera were recovered in total. These samples were dominated by *Haplophragmoides walteri* and *Hyperammina* spp., and all species observed are agglutinated non-calcareous forms. It is possible that calcareous species were never present in the central North Sea at this time, or that secondary dissolution has preferentially removed these forms. This low diversity, non-calcareous, agglutinated fauna is indicative of low oxygen marine environments (Gradstein and Berggren, 1981; Charnock and Jones, 1990; Gradstein et al., 1992). Samples from the interval ~2611 to 2606 m associated with full PETM conditions and the upper part of the laminated section, are found to be barren of all foraminifera including a near absence of foraminiferal test linings in the palynological residues (Table S1). The lack of benthic foraminifera within the laminated sediments is consistent with the interpretation of anoxic bottom water.

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Supplementary tables

Table S1. Raw counts (apart from *) of palynomorphs from 22/10a-4. All specimens on a single slide were counted. As each slide was made with 0.05 g of sediment, all counts represent specimens/0.05 g. * = Estimate of total specimens per slide (specimens/0.05 g), these specimens counted to >300, and subsequently scaled up to an estimate for the whole slide by number of traverses taken.

Table S2. Raw counts of benthic foraminifera from 22/10a-4.

Table S3. C and N wt% data from 22/10a-4.

Table S4. Carbon isotope data from 22/10a-4.

Supplementary figures

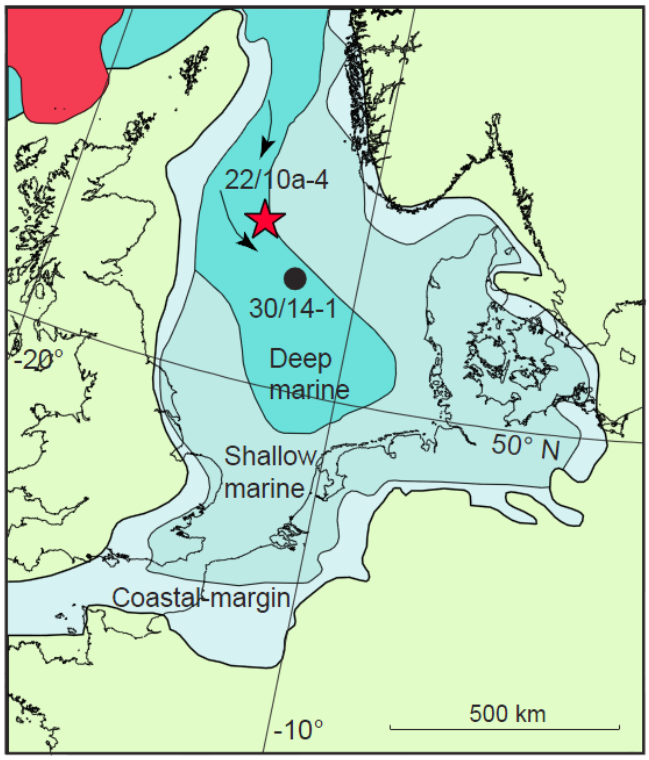

Fig. S1. Paleogeographic reconstruction of the North Sea area for the Paleocene/Eocene at ~55.5 Ma (Coward et al., 2003), with locations of cores 22/10a-4 (this study, 57°44'8.47''N. 1°50'26.59''E.) and 30/14-1 (Sluijs et al., 2007). Arrows indicate principal direction of sediment flow, red is area of basaltic volcanism, deep marine equates to middle bathyal (500-1000 m water depth).

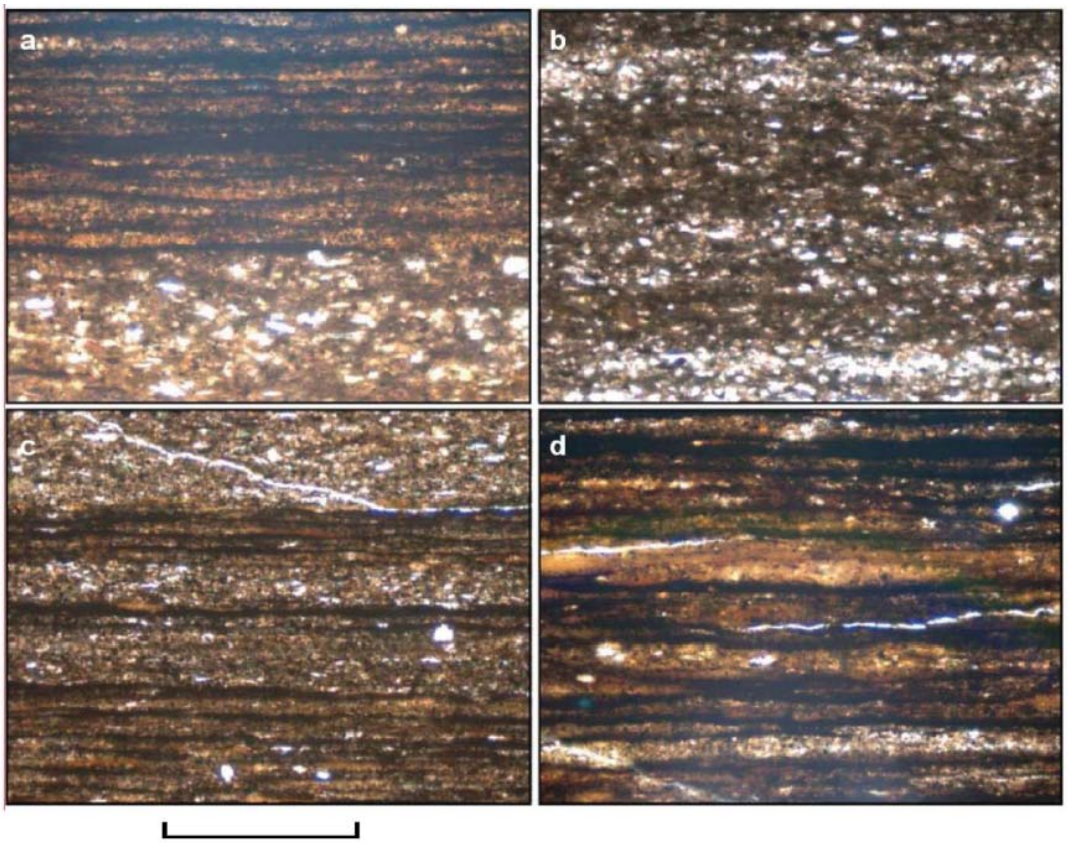

Fig. S2. Transmitted light microscope images of thin sections from core 22/10a-4 from 4 horizons. (a) 2612.266 mbsf, (b) 2612.226 mbsf, (c) 2612.176 mbsf, and (d) 2611.986 mbsf. Laminations are laterally continuous and vary in thickness. Dark laminae are organic rich, light laminae are clay and sand rich. An average of ~13 laminae per mm occur throughout the section. Scale bar = 0.5 mm.

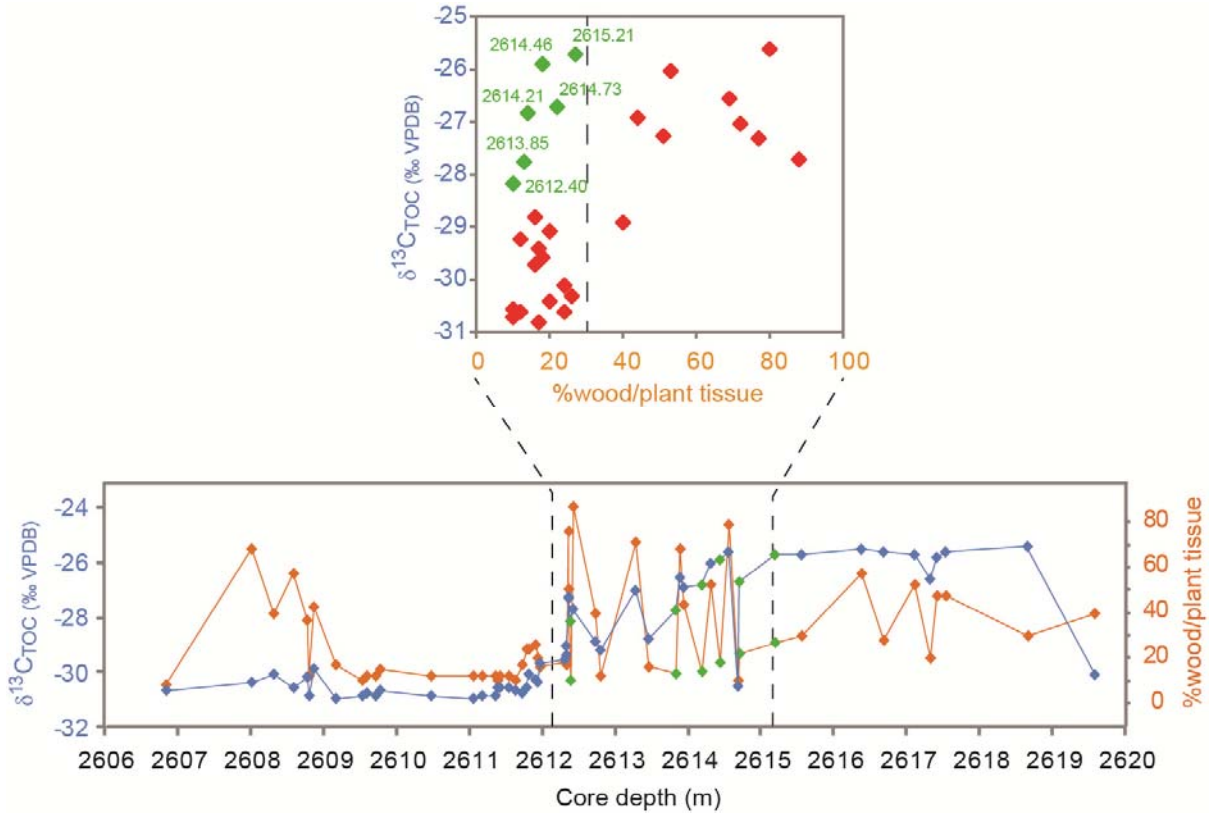

Fig. S3. Core 22/10a-4 %wood/plant tissues (from all palynological residues) plotted with their corresponding bulk carbon isotopes ($\delta^{13}C_{TOC}$). All samples with >30% wood/plant tissue within the CIE (~2615-2612 m) exhibit heavier $\delta^{13}C_{TOC}$ values (upper plot) uncharacteristic of PETM values, signifying either re-deposited pre-CIE terrestrial material and/or transported terrestrial material with more positive $\delta^{13}C_{TOC}$ values than contemporaneous North Sea marine organic carbon. Positive $\delta^{13}C_{TOC}$ samples within the CIE onset that do not have >30% wood/plant tissue are marked in green. The majority fall within the early CIE onset as expected. One sample at 2612.40 m however has relatively high $\delta^{13}C_{TOC}$. As there is <30% wood/plant tissue in this sample, it is possible that it represents a pulse of non-woody terrestrial material transported to 22/10a-4, possibly peat.

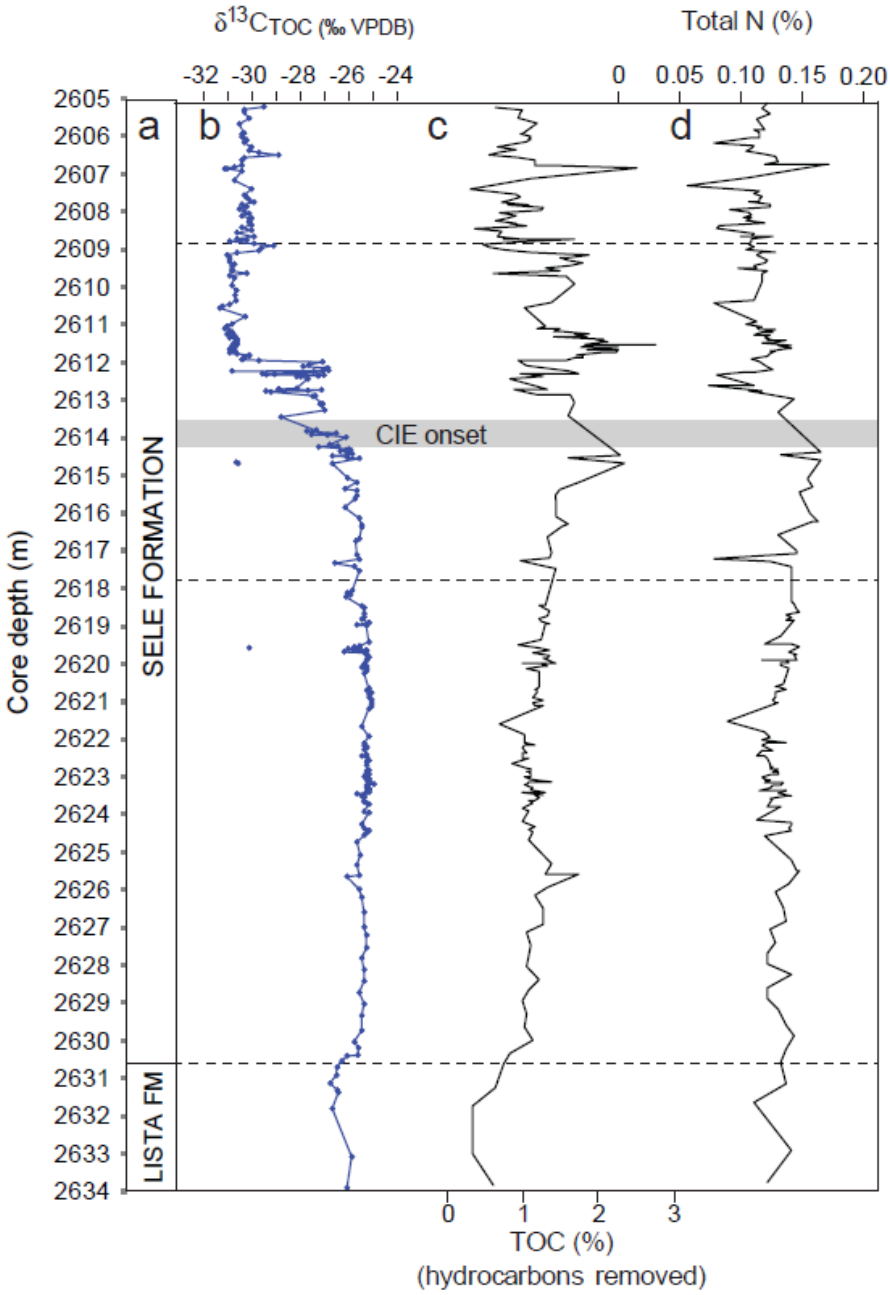

Fig. S4. Total organic carbon (TOC) and total nitrogen (TN) wt% values for 22/10a-4.