Mobilization of lithospheric mantle carbon during the Palaeocene-Eocene thermal maximum

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Abstract

The early Cenozoic exhibited profound environmental change influenced by plume magmatism, continental breakup, and opening of the North Atlantic Ocean. Global warming culminated in the transient (170 thousand year, kyr) hyperthermal event, the Palaeocene-Eocene thermal maximum (PETM) 56 million years ago (Ma). Although sedimentary methane release has been proposed as a trigger, recent studies have implicated carbon dioxide (CO\textsubscript{2}) emissions from the coeval North Atlantic igneous province (NAIP). However, we calculate that volcanic outgassing from mid-ocean ridges and large igneous provinces associated with the NAIP yields only one-fifth of the carbon required to trigger the PETM. Rather, we show that volcanic sequences spanning the rift-to-drift phase of the NAIP exhibit a sudden and ~220-kyr-long intensification of volcanism coincident with the PETM, and driven by substantial melting of the sub-continental lithospheric mantle (SCLM). Critically, the SCLM is enriched in metasomatic carbonates and is a major carbon reservoir. We propose that the coincidence of the Iceland plume and emerging asthenospheric upwelling disrupted the SCLM and caused massive mobilization of this deep carbon. Our melting models and coupled tectonic-geochemical simulations indicate the release of >10\textsuperscript{12} gigatons of carbon, which is sufficient to drive PETM warming. Our model is consistent with anomalous CO\textsubscript{2} fluxes during continental breakup, while also reconciling the deficit of deep carbon required to explain the PETM.

During the early Cenozoic, a major episode of continental breakup in the North Atlantic region (Fig. 1a) resulted in widespread magmatism and the opening of the North Atlantic Ocean\textsuperscript{13}. Initially the Iceland plume caused regional uplift and a phase of subaerial volcanism lasting ~5 million years (Myr)\textsuperscript{2,3,9,10}. Then, at ~56 million years ago (Ma), an incipient mid-ocean ridge developed (Fig. 1d), coinciding with an order-of-magnitude increase in melt production rates\textsuperscript{7,8}, and a celerated westward migration of Greenland via ridge push\textsuperscript{8} (Fig. 1e). This transitional phase of volcanism in the North Atlantic igneous province (NAIP) involved peak eruption rates of ~2.4 km\textsuperscript{3} yr\textsuperscript{-1} (ref.\textsuperscript{9}) and occurred contemporaneously with intense early Eocene warming, including the PETM\textsuperscript{11}. During the PETM, sea surface temperatures increased by ~5°C (ref.\textsuperscript{12}), ocean acidification occurred\textsuperscript{13,14} and there was an abrupt and short-lived (~170 thousand year, kyr)\textsuperscript{11,15} negative \delta\textsuperscript{13}C carbon isotope excursion (Fig. 1c). It is thought that the release of approximately 1–1.5 × 10\textsuperscript{20} gigatons (Gt) of carbon into the ocean-atmosphere system is necessary to account for these changes\textsuperscript{14,16}. This massive carbon flux has been attributed to methane release associated with clathrate destabilization along continental shelves\textsuperscript{17} and/or hydrothermal venting during interaction of magmas and organic-rich mudrocks\textsuperscript{18}. However, on the basis of paired \delta\textsuperscript{11}B–\delta\textsuperscript{13}C data and carbon-cycle modeling\textsuperscript{14}, and B/Ca in planktic foraminifera\textsuperscript{16}, it has been proposed that triggering of the PETM required a short, sharp injection of a \delta\textsuperscript{13}C depleted source of carbon, possibly tied to volcanic outgassing across the NAIP.

To test this hypothesis, we estimated the combined CO\textsubscript{2} emissions from incipient mid-ocean ridge volcanism and large igneous provinces (LIPs) in the NAIP, parameterized by plate-tectonic reconstructions (Fig. 1b,d) and existing estimates of magmatic productivity\textsuperscript{9} (Methods). In our model, we considered pre-eruptive CO\textsubscript{2} concentrations of 2 wt%, which are typical of flood basalt eruptions\textsuperscript{19}. The associated CO\textsubscript{2} release from the ocean crust is then calculated from the ratio between the observed levels of magmatic production along the present-day mid-ocean ridge system (~18 km\textsuperscript{2} yr\textsuperscript{-1}) and estimated CO\textsubscript{2} degassing fluxes along the modern global ridge system (7 × 10\textsuperscript{11} mol yr\textsuperscript{-1})\textsuperscript{20}. This calculation suggests that ~10% of the CO\textsubscript{2} in the juvenile ocean crust is degassed at ridges. Using this estimate, the NAIP ridges likely produced ~1.1 × 10\textsuperscript{12} Gt C during the PETM. This is an order of magnitude lower than the requisite ~1–1.5 × 10\textsuperscript{20} Gt C (refs.\textsuperscript{14,16}). Inclusion of the effects of LIP magmatism—assuming most probable eruption rates\textsuperscript{9} and total degassing—yields less than one-fifth of the carbon needed to drive and sustain the PETM\textsuperscript{14,16}. Furthermore, seafloor isochrons indicate that magmatic productivity along...
the North Atlantic ridge peaked after, not during, early Eocene
hyperthermals (Fig. 1d). Therefore, some other major, but tran-
sient, source of volcanic carbon appears to be required if the
volcanic outgassing hypothesis is correct.

We have investigated several volcanic sequences spanning
the Palaeocene-Eocene boundary (Fig. 1c). The Deep Sea
Drilling Project Leg 81 Site 555 lies on the Rockall Plateau
(Fig. 1a), near the proto-North Atlantic ridge (Fig. 1b). Here,
Phase 1 volcanism\(^2\) (Fig. 2a) is coeval with the Milne Land
basalts in East Greenland and the Middle to Upper Series lavas
in the Faroe Islands\(^1\) (Fig. 2b–c). In the Rockall sequence, we
found a sharp increase in the frequency of volcanic tufts just be-
low the Palaeocene–Eocene boundary (Fig. 2a) (Methods). The
PETM is defined by \(\delta^{13}C\), however the volcanoostratigraphy in
our study area is not conducive to developing a high-resolution
carbon isotope stratigraphy (Methods). Therefore, we rely on a
combination of radiometric, magnetostratigraphic and paleon-
tological age constraints, in addition to well-defined sediment
accumulation rate estimates (Methods). Mudstones interbedded
with the uppermost tufts contain the dinoflagellate cyst, Apec-
todinium augustum\(^2\), which is biostratigraphically diagnostic

of the PETM as it signifies a sudden prevalence of tropical sea-
surface temperatures\(^2\). Based on sedimentation rates (50 cm
ka\(^{-1}\))\(^4\), this volcanic flare-up lasted for 171–213 kyr, similar to
the duration of the coeval PETM\(^1,11,15\), and was followed by a
sharp decline in volcanism\(^2\). The tufts exhibit wide composi-
tional diversity from basanites to dacites (Supplementary Figs.
11,15), the duration of the coeval PETM\(^1,11,15\), and have similar compositions to some of the lowermost ('neg-
ative ash series') tufts of the Danish Basin, which likely derive from
nearby volcanoes along the continental shelf\(^29\). The geo-
chemical similarities between tufts from Rockall and the Dan-
ish Basin is consistent with paleogeography (Fig. 1b), palyno-
logical constraints\(^27\) and the stratigraphic position of these tufts
towards the end of the PETM elsewhere in the NAIP\(^6\).

Figure 1. Early Cenozoic tectonic and magmatic evolution of the North Atlantic region | a. Map of the present-day North Atlantic region showing the
distribution of Palaeocene–Eocene lava flows and intrusives\(^1\), with dated volcanics denoted by colored symbols\(^7\). b. Plate tectonic reconstruction showing nascent
ridge systems developing along the Labrador Sea and North Atlantic. c. Ages of the volcanic sections discussed (Up=Upper; VFF=Vandfaldsdalen Fm), defined by
radiometric dates\(^1,13\), magnetostratigraphy and nannofossil zonation\(^4,5,8\), and corresponding carbon and oxygen isotope records showing the PETM isotope
excursions (solid and faint lines show 1 Myr and 20 kyr locally weighted functions, respectively)\(^7\). d. Seafloor production rates for the Labrador Sea and North
Atlantic, derived from GPlates (Methods), shown alongside the timing of Eocene hyperthermals. (E) Palaeolongitude of Greenland\(^3\) indicating the onset of ocean
crustal production in the North Atlantic and ridge push at 56 Ma.
Fig. 2. Palaeocene-Eocene volcanostratigraphy and geochemistry of the proto-North Atlantic ridge. a, Simplified log of the Rockall ‘Phase 1’ sequence showing lithologies, Mg# (i.e., 100 × molecular MgO/(MgO + FeO), where FeO is assumed to be 0.9FeOT), and εNd (Supplementary Fig. 1). b, Simplified log of the Faroes Basalt Formations, with Mg# and (Eu/Yb)n (chondrite-normalized) data from the Faroe Islands (Faroese Basalt Formations), showing lithologies, Mg# (i.e., 100 × molecular MgO/(MgO + FeO), where FeO is assumed to be 0.9FeOT), and εNd (Supplementary Fig. 1). c, East Greenland showing lithologies, Mg# (i.e., 100 × molecular MgO/(MgO + FeO), where FeO is assumed to be 0.9FeOT), and εNd (Supplementary Fig. 1). d, (La/Yb)n vs (Eu/Yb)n of the Faroes and Hold with Hope (HwH) lavas (chondrite-normalized) and modeled non-modal batch melting of a lherzolitic mantle source, adopted from, showing different degrees of melting of a garnet lherzolite (green, blue and red curves). e, (Sm/Yb)n vs (Ce/Sm)n and an REE melting model (Methods), showing percentage melt along the top and the relative proportions of garnet- and spinel-lherzolites from 100% garnet-lherzolite (red curve) to 100% spinel-lherzolite (green curve). Both models indicate that the Faroes Middle Lava Formation (i.e., high Mg# basalts in the lower 500 m of the MLF; see (b), that erupted immediately prior to and during the PETM, experienced the highest degrees of melting of a mantle source containing ≥10% garnet.
Interpretation of geochemical data from the Rockall tuffs is complicated because they derive from diverse sources and have experienced varying degrees of seafloor weathering. Hence, we next studied the coeval thick basaltic lava sequences that were emplaced subaerially near the ridge axis (Fig. 1). The Palaeocene-Eocene lavas of the Faroe and East Greenland land (Fig. 1b) are ideally suited because, unlike the contemporaneous lavas in SE Greenland, they are minimally altered by crustal contamination. The base of the Milne Land Formation in East Greenland, which correlates with the base of the Middle Lava Series of the Faroes (Fig. 2b-c), hereafter referred to as ‘MLF’, is dated at 56.1 ± 0.4 Ma and the duration of activity is well constrained (Fig. 2). Thus, the base of the MLF correlates with (or shortly predates) the intensification of volcanism at Rockall Plateau (Fig. 2a).

We estimate that the 1.25 km-thick MLF package, emplaced over 200–300 kyr, yielding an average eruption rate of ~4–6 m kyr\(^{-1}\), but potentially an order of magnitude higher early in the eruptive cycle. At the base of the sequence, thick (>100 m) pyroclastic deposits, also reported in Greenland (106), signal an early volatile-rich explosive phase. The overlying tholeiitic lavas are characterized by a sharp increase in Mg\(^{#}\), from values of ~52 throughout the Lower Lava Formation to >80 in the basal MLF (Fig. 2b-c). The basal lavas are coincident (within dating uncertainties) with the onset of the PETM (11). Many of these lavas are highly magnesian (e.g., with MgO = 24%), implying hot liquidus temperatures, and are locally characterized by massive olivine accumulation. They also exhibit high TiO\(_2\) contents (typically 1.5–2.5 wt.%) and enrichment in Light Rare Earth Elements (LREEs), e.g., \((La/Yb)_{\text{a}} = 2–3,\) and (Eu/Yb)\(_{\text{a}} > 2.5\) (Fig. 2b) (24,25). This unusual pulse of high-Ti magmatism, similar to that of the Jurassic Karoo flood basalts in southern Africa\(^{26}\), generated the long-lived (~300 kyr) Skaergaard layered igneous intrusion at 55.75 ± 0.35 Ma (Fig. 1a). Finally, the Upper Lava Formation (Fig. 2b-c) signifies an abrupt shift to low-Ti depleted MORB-like basalts (22,24,25) (Fig. 2d-e), thus, there is evidence that the early (high Mg\(^{#}\)) MLF volcanism was both transient (~227 kyr) and chemically anomalous (Fig. 2).

The geochemical compositions of these lavas have been attributed to partial melting of the SCLM during asthenospheric upwelling, a model that is supported by studies of their feeder intrusions. To test this hypothesis further, we applied two models of mantle melting. First, we adopted a non-modal batch melting model of a garnet-bearing lherzolitic mantle source (Fig. 2d). For the second model, we estimated melt percentage based on average lherzolite mineralogy, average partition coefficients, and a plume composition based on primitive basalt (Fig. 2e) (Methods). Both models suggest that the MLF experienced a high degree of melting, up to ~13%, but typically in the range of 4–8%, with their high (Sm/Yb)\(_{\text{a}}\) ratios (2–2.5) requiring ~20–50% of garnet lherzolite in the mantle source (Fig. 2e). Metasomatism in the mantle source region has already been documented in this region at precisely this time and voked to explain the compositional characteristics of these high-Ti basalts, notably their LREE enrichment and variable Nb and Ta anomalies (31,32). The composition of the MLF is compatible with enhanced melting of metasomatized SCLM by the Iceland plume, just prior to full seafloor spreading at c. 55.8 Ma (Figs. 2b-c).

The trace element compositions show that the degree of melting varies systematically through the Faroes sequence (Fig. 2e), with the Lower Lava Formation (Fig. 2b-c) representing a relatively low degree of melting (2–5%; Fig. 2e). To expand the dataset, lavas from farther along the ridge axis in northeast Greenland are included (Hold with Hope) (1b), which have an upper age of ca. 57 Ma (C25n–C24r) (34). These lavas exhibit geochemical characteristics that have been attributed to the incorporation of SCLM during melting (31,32). Our model confirms a low degree of melting (0.5–2%) of a variably garnet-rich lherzolite source (Fig. 2e). Taken together, these observations confirm an up-section increase in the involvement of SCLM in melting during the late Palaeocene, peaking at the PETM (Fig. 2e), then rapidly declining in intensity prior to full continental breakup. There is possible evidence for this peak in Palaeocene–Eocene basaltic lavas of the Vadsfaldsalen Formation in central east Greenland. These lavas exhibit Nd, Sr, and Pb isotope signatures indicative of an SCLM component (35). Assimilation of the deep SCLM is particularly important because this zone is metasomatically enriched in carbonates, and is thus a major carbon reservoir (6,37,38).

Figure 3. Simulations of volcanic carbon release during the PETM. Results are plotted as cumulative distribution functions (CDFs). The gray lines show the estimated carbon output from ridge volcanism and LIPs alone; with S1 and S2 showing low (0.6 km\(^3\) yr\(^{-1}\)) and high (2.4 km\(^3\) yr\(^{-1}\)) LIP eruption rate scenarios, respectively (Supplementary Fig. 3) (see Methods). The coloured lines show the effects of adding 4% to 8% carbonated (~50 C) SCLM melt along the incipient ridge during breakup. The gray vertical bars denote the carbon output necessary to drive and sustain PETM warming estimated by Gutjahr et al. (labelled G: 10,200–12,200 Gt C), and Haynes & Honisch (labelled H: 14,900 Gt C).

The process of ocean basin formation significantly disrupts the basal thick SCLM (39). Because carbonates are only stable in the mantle at high pressures (~3 GPa at 1300°C) (39), significant carbon release will only occur when extending crusts are at least 130 km thick. Seismic tomography indicates that the lithosphere in central Greenland (i.e., the locus of the Iceland plume at 56 Ma\(^{40}\)) is 180 km thick, firmly meeting this criterion. Xenoliths provide additional evidence for coeval volatile-rich metasomatism in the deep SCLM be-
neath Greenland, as does the occurrence of carbonatite magmatism across the wider NAIP at this time. Therefore, breakup of the North Atlantic craton provided a perfect combination of conditions: the availability of sufficiently thick metasomatized SCLM, analogous to the present-day Tanzanian craton in the East African Rift System; the long-term thermo-mechanical weakening and erosion of the SCLM by the Iceland plume, broadly analogous to that proposed for the Permian-Triassic Siberian Traps; and the initiation of asthenospheric upwelling that induced vigorous interaction between anomalously hot melt and the weakened SCLM. A key aspect of our hypothesis is that the most extensive period of volcanic CO$_2$ outgassing (~50 Mt C yr$^{-1}$) occurred over a short period (~171–227 kyr), and likely peaked just prior to PETM onset (Fig. 2b-c). While continental rift volcanism is associated with enhanced CO$_2$ fluxes over millions of years, petrological and numerical models suggest that there is an initial short period of deep redox controlled carbonatic to aillikitic melting (i.e., high CO$_2$) that is followed by more widespread metasomatism of the shallower asthenosphere and generation of less CO$_2$-rich melts through decompression melting.

Both the thickness and composition of the NAIP lithospheric mantle make it a viable source of large amounts of carbon from the deep, carbonated SCLM during the PETM. A fundamental aspect of our model is that the occurrence of continental rifting opens a wide area along the incipient ridge to SCLM influence. This contrasts with the end-Permian Siberian Traps, where there is no evidence for large-scale lithospheric stretching during eruption. Indeed, it has been suggested that relics of this SCLM that delaminated during North Atlantic breakup can explain residual enriched ‘EM1’ components present in the Icelandic mantle today. The question is then whether this process can contribute enough carbon to help explain the PETM carbon isotope excursion. Recent estimates suggest that the SCLM may contain 5–8% CO$_2$, or possibly more. The present-day East African Rift offers a lower bound. Here, lateral advection of SCLM along the craton margins results in an order of magnitude increase in the volcanic CO$_2$ flux compared to the background crustal contribution.

To more accurately quantify the potential influence of rift-related SCLM disturbance, we advance on the approach taken by Sobolev et al. to estimate the most probable carbon release scenarios for the PETM. We performed 10,000 Monte Carlo calculations using CO$_2$ contents ranging from 1–8%, and length-scales of the carbonated SCLM in the narrow melting zone below the nascent ridge crest based on tectonic reconstructions, chemical tomography and lithospheric models (tales S7–S8). These calculations suggest that only 4–8% of this zone needs to melt to exceed the requisite CO$_2$ fluxes from SCLM to the crust (Fig. 2d-e). This is consistent with the modeled 5–8% melting of a garnet-rich lherzolite zolite during the PETM (Fig. 2d-e), and earlier estimates that the intrusive feeders of these Ti-rich basalts formed by 4%–7% batch melting of fertile, metasomatized lherzolites in the SCLM.

The step change in NAIP magmatic productivity immediately prior to the PETM gave rise to widespread volcanic unrest (Figs. 2, 4). While some volcanic tufts at Rockall Plateau are likely linked to the MLF (especially those with high Mg#), many of these tufts are likely genetically unrelated, but are rather a manifestation of the general increase in mantle melting, and fluxes of magma and volatiles through volcanic centres along the continental shelf at this time. The Rockall sequence places firm constraints on both the timing and duration of this climate-altering phase of volcanism at ~171–213 kyr, which is similar to the duration of the lower, high Mg#, part of the MLF (Fig. 2). This surge in regional volcanism could explain the wide variability in the composition of tufts belonging to the Danish negative ash series. Indeed, the chemical weathering of extensive tephra blankets may also explain the large decrease in Os observed just prior to PETM onset at Svalbard. Enhanced melting of the SCLM during continental breakup resulted from an unusual combination of conditions in the NAIP (Fig. 4) and provides a direct mechanism to dramatically increase the outgassing of deep carbon. We have shown that this process reached peak intensity just prior to, and during, the PETM (Fig. 2), satisfying the requirement for a massive surge of depleted carbon into the LIP volcanism (see curves S1–S2 on Fig. 3). The melting of SCLM can result in a fivefold increase in volcanic CO$_2$ output, reconciling an apparent major deficit of carbon from ‘background’ ridge and LIP volcanism (see curves S1–S2 on Fig. 3). The mobilization of carbon from the SCLM is consistent with the inferred disruption of the metasomatized lithosphere during breakup of the North Atlantic craton, and the anomalously high CO$_2$ fluxes from SCLM along craton margins during extension.
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Our data-driven models of the PETM demonstrably support the proposal that large-scale lithospheric melting can induce global warming, if the tectonic setting is primed to facilitate intensive volcanic CO2 degassing (Fig. 4). Our study highlights the critical role that solid Earth degassing plays in driving abrupt shifts in climate, and in promoting fundamental reorganisation of Earth’s surface environment and biosphere.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s12345-111-2222-3.
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Methods

Calculating seafloor production rates

Seafloor production (SP) rates (Fig. 1d), which were utilised in our volcanic CO$_2$ flux calculations (see page 277), 10, were calculated using the plate model of Müller et al. (2016). We used the open source python library, pyGPlates (https://www.gplates.org/docs/pygplates/) in order to filter and extract the data from the plate model. For the target time-steps, $(t, \text{in Myr})$, we broke the complete mid-ocean ridge system into a series of spreading and transform segments in order to isolate the spreading segments $(j)$ where new oceanic crust is formed. This approach has recently become common in analysing tectonic scenarios in ocean basins. At each segment, for each time-step, we extracted the full spreading rate $(u, \text{in km Myr}^{-1})$ and the length of the spreading segment $(L, \text{in km})$. The full spreading rate was calculated by summing the half spreading rate of each individual flank of each ridge segment, thereby sidestepping any issues pertaining to asymmetric spreading. We did this because, for the purposes of our analysis, we were only interested in the total amount of new seafloor generated. We then took the product of the spreading segment length and full spreading rate to calculate the seafloor production rate (in km$^2$ Myr$^{-1}$), and then summed the area of all segments per time-step, to obtain a total seafloor production per Myr (equation 1), as follows:

$$SP(t) = \sum_j uL$$

Geochemical analysis of the volcanic tufts

We carried out major, trace and isotopic analysis (variably) on 20 tuft layers sampled from DSDP Site 555 on the Rockall Plateau. X-ray fluorescence (XRF) analyses were carried out in the Grant Institute of Earth Science at the University of Edinburgh. Samples were then digested via a mixed acid, closed vessel HNO$_3$-HF approach on a hotplate at 120°C for at least 1 hour, and a precisely-weighed 1-g aliquot ignited at 1100°C to determine loss on ignition (LOI). The fused and pressed samples were analysed using a PANalytical PW 2404 automatic X-ray fluorescence spectrometer with a Rh-anode X-ray tube. Trace-element background positions were placed as close as possible to peaks, and long count times were used at both peak and background positions. Where background count rates were measured on either side of the peak, as in most trace-element determinations, the count time was divided between the two positions. Analytical conditions are given in refs. 50,51.

Corrections for matrix effects on the intensities of major-element lines were made using theoretical alpha coefficients calculated on-line using the PANalytical software. The coefficients were calculated to allow for the amount of extra flux replacing volatile components in the sample so that analytical totals should be 100% less than the measured LOI. Intensities of the longer wavelength trace-element lines (La, Ce, Nd, Ni, Co, Cr, V, Ba, and Sc) were corrected for matrix effects using alpha coefficients based on major-element concentrations measured at the same time on the powder samples. Matrix corrections were applied to the intensities of the other trace-element lines by using the count rate from the RhK$_\alpha$ Compton scatter line as an internal standard. Line-overlap corrections were applied using synthetic standards.

The spectrometer was calibrated against USGS and CRPG geochemical reference standards using the values given by Govindaraju (1994), except that the values of Jochum et al. (1990) were used for Nb and Zr in BCR-1 and BHVO-1. Excellent calibration lines were obtained using these standards. Analytical precision and accuracy are comparable to the values reported in refs. 50,51. Four USGS geostandards were analysed along with the samples and the data from these (Supplementary Table 2) were used to assess accuracy and precision.

Trace element analysis was performed on a representative selection of four of the tufts using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS). Samples were homogenised using a unidirectional crushing machine prior to dissolution via a three-stage, mixed acid (HF-HNO$_3$-HCl) closed-vessel approach. After digestion, samples were dried down prior to resuspension in 2% HNO$_3$ for analysis, carried out using a Thermo Scientific X-Series-2 at the University of Southampton. Alongside samples, blanks and International standard JA-2 (ref. 56) and Southampton internal basalt standard BRR-1 (ref. 57) were prepared and analysed (Supplementary Table 4). Recoveries of JA-2 and BRR-1 are shown in Supplementary Table 5.

Finally, we analysed the $^{143}$Nd/$^{144}$Nd isotope composition of several samples of tuft from Site 555 (Fig. 2a; Supplementary Table 3). Here, ~25 mg of freeze-dried sediment was homogenised using a pestle and mortar prior to leaching in 6M HCl for 2 hours to remove any diagenetic, alteration-related material. Samples were then digested via a mixed acid, closed vessel HNO$_3$-HF approach on a hotplate at 120°C. Digests were then taken to incipient dryness prior to resuspension in 2% HNO$_3$ for analysis. All acids were sub-boiled to ensure purity and prepared at the University of Southampton. Aliquots were then purified via first cation (AG50-X8 200-400 mesh compressed against a polished tungsten carbide disc in a hydraulic press at 0.6 tons cm$^{-2}$. The fused and pressed samples were analysed using a PANalytical PW 2404 automatic X-ray fluorescence spectrometer with a Rh-anode X-ray tube. Trace-element background positions were placed as close as possible to peaks, and long count times were used at both peak and background positions. Where background count rates were measured on either side of the peak, as in most trace-element determinations, the count time was divided between the two positions. Analytical conditions are given in refs. 50,51.
resin) and then reverse phase (LN Spec, Eichrom Industries) chromatography to remove isotopic interferences. All measurements were made on a Thermo Fisher Neptune Multi-Collector ICP-MS at the University of Southampton. Mass bias corrections were performed using repeat measurements of the standard JNd-1 (ref. 58) with a $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.512115. Nd isotope ratios are presented using epsilon notation as follows:

$$\epsilon\text{Nd} = \left(\frac{^{143}\text{Nd}}{^{144}\text{Nd}}_{\text{sample}} / \frac{^{143}\text{Nd}}{^{144}\text{Nd}}_{\text{CHUR}} - 1\right) \times 10^4$$

where $^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}}$ is the Nd isotopic composition of the chondrite uniform reservoir, with a value of 0.512638 (ref. 59).

Precision of $^{143}\text{Nd}/^{144}\text{Nd}$ (2 SE) was between 0.000005 and 0.00001, corresponding to $\epsilon\text{Nd}$ precision of between 0.1 and 0.2.

Establishing a chronostratigraphic framework

We established a chronostratigraphic framework for Site 555, on the Rockall Plateau, building on a number of existing constraints at this site (Fig. 1a). The PETM as geochemically defined could not be identified at this site, because of the high proportion of volcanic and siliciclastic material (Fig. 2a), in contrast to the adjacent sediments likely experienced temperatures $>650^\circ\text{C}$, thermal conditions which are known to seriously affect $^{13}C$compilation in mudrocks and carbonates (ref. 60). Accordingly, Nd and $^{143}\text{Nd}/^{144}\text{Nd}$ thermal ages at this site were interpreted as $\sim$0.4 Ma. Storey et al. (2007) use the relative age of the well-dated Danish Ash-17 to firmly place the PETM interval after the onset of MLF volcanism. A late Palaeocene age is also supported by palynologic studies of the underlying (non-marine) coal-bearing Prestfjall Formation, and its position near the boundary between magnetic polarity chronos c24r and c25n (i.e., 57.1 Ma; ref. 65). For the Upper Lava Formation (ULF), we assumed an upper age of 55.5 Ma; this is based on an average of three radiometric ages, including one obtained from the upper ULF in the Faeroes (55.1 ± 0.5 Ma; ref. 1). We used two radiometric ages obtained from the Skåergaard intrusion (55.75 ± 0.3 Ma, and 55.65 ± 0.3 Ma; ref. 66), where the parental magma has been genetically linked to the Milne Land Formation (ref. 67), and accordingly its crystallisation is considered to provide an upper age constraint for Milne Land Formation volcanism (and thus, the lower Geikie Plateau Formation22). Using this age model, we calculate the duration of the lowermost high Mg# part of the MLF (i.e., the lowermost 800 m; Fig. 2b) to be 227 kyr, similar to that of the volcanic phase at Rockall.

Establishing a chronostratigraphic framework for the Faroese Lower Lava Formation (correlative with the Nansen Fjord Formation of east Greenland) is more challenging because of the large uncertainties associated with radiometric ages through this succession. However, this inherent uncertainty does not impact our results in any way; data for the lower formations are shown only for reference purposes on our stratigraphic section (Fig. 2b–c). As a reasonable solution, we used magnetic polarity chrons for the Faroes sequence68, combined with the chemical stratigraphy of Larsen et al. (1999)22 and using a standard geomagnetic polarity time scale (ref. 65). We calibrated the ages at certain depths in the succession using the ages of chron boundaries as tie points, and applying a linear interpolation between these known points (which assumes constant lava accumulation rates). We found that this age model was broadly consistent with existing radiometric ages (i.e., in most cases within uncertainty).

REE melting model

A simple batch melting equation was used in the construction of Figure 2e:

$$C_L/C_0 = \frac{1}{D + F - PF}$$

where $C_0$ is the initial concentration of some element in the mantle source, $C_L$ is its concentrations in the liquid, $F$ is the melt fraction, $D$ is the average distribution coefficient for the mantle phases weighted by their respective mass fractions.
before the onset of melting, and $P$ is the average distribution coefficient for the mantle phases weighted by their respective contribution to the melt. We use a peridotite mantle source with a mineralogical composition given by McKenzie and O’Nions (1991): (1) 57.8% olivine, 27.0% orthopyroxene, 11.9% clinopyroxene and 3.3% spinel (spinel lherzolite); and (2) 59.8% olivine, 21.1% orthopyroxene, 7.6% clinopyroxene and 11.5% garnet (garnet lherzolite). Partition coefficients (D) vary significantly with pressure, temperature and liquid composition, and this limits the reliability of partial melting models since the choice of values is always somewhat subjective. Here we have attempted to be more objective by using averages of all appropriate published D values (Supplementary Table 6). We use the following mantle melting proportions for spinel lherzolite (equation 4; ref. 71) and garnet lherzolite (equation 5; ref. 71):

$$\text{liquid} = -0.22\text{ol} + 0.38\text{opx} + 0.71\text{cpx} + 0.13\text{sp}$$

$$\text{liquid} = 0.08\text{ol} - 0.19\text{opx} + 0.81\text{cpx} + 0.30\text{gt}$$

where ol is olivine, opx is orthopyroxene, sp is spinel, cpx is clinopyroxene and gt is garnet.

The concentrations of REE in our mantle source (Ce 1.23 ppm, Sm 0.346 ppm, Yb 0.432 ppm) are calculated from the composition of primitive basalt from the Ontong Java Plateau and are thought to be typical of mantle plumes. Please note that Supplementary Table 6 cites additional references.

**Modelling of CO$_2$ fluxes**

We perform a simple Monte Carlo simulation, sampling from probability distributions for six uncertain parameters (Supplementary Tables 7–8) to estimate the probable combined CO$_2$ emissions from incipient ridge volcanism and active large igneous provinces (LIPs). This enables us to evaluate the relative contributions to carbon release during the PETM due to ridge production and LIP formation. We also calculate the effect of incorporating carbonated SCLM melt in different amounts during enhanced melting along the ridge axis. In our calculations, we assumed a PETM duration of 170 kyr (ref. 15), which is supported by recent astrochronological solutions.

We use Beta distributions to represent uncertainty in the parameters (Supplementary Table 8). The Beta distribution is a continuous distribution over a fixed interval [0, 1], but can be rescaled to any desired range. It is defined by two shape parameters $\alpha$ and $\beta$, which can be estimated from the distribution mean ($\mu$) and variance ($\sigma^2$):

$$\alpha = \mu \left(\frac{1 - \mu}{\sigma^2} - 1\right)$$

$$\beta = \alpha \left(\frac{1}{\mu} - 1\right)$$

where $\mu$ = the mean, $\sigma$ = standard deviation, and $\sigma^2$ = the variance. To estimate Beta distribution parameters, we use best estimates (from published data and observations) of the minimum, mean and maximum values for each variable (Supplementary Table 8, and discussed below), and apply these to equations 6 and 7 above (using re-scaled values for $\mu$ and $\sigma$). For simplicity we assume that the standard deviation for a given variable is 20% of the range, $\sigma_r = 0.2$ (max–min). The inputs to the calculations (sampled, constant and fixed) and outputs are listed in Supplementary Tables 7–8 and the corresponding histograms for each of the sampled variables are shown in Supplementary Fig. 3.

To quantify the CO$_2$ degassing flux from ridges in the NAIP (i.e., the North Atlantic and the Labrador Sea), we used seafloor production rates parameterised from plate-tectonic reconstructions (see above) for the ridges at 55 Ma—the first time step at which seafloor generation is recorded in the North Atlantic. In these calculations we assumed an oceanic crustal thickness of 6 km, the global average. Only a small fraction of the total amount of CO$_2$ available in the basaltic oceanic crust is degassed at ridges. To estimate this, we used the ratio between the oceanic crustal productivity (i.e., the total amount of crust formed, which is ~18 km$^3$ yr$^{-1}$) and the estimated CO$_2$ fluxes from the present-day global ridge system (7 $\times$ 10$^{11}$ mol yr$^{-1}$; a middle value of ref. 20). This analysis indicates that about 10% of the CO$_2$ available in the ocean crust is degassed at ridges (see main text, corresponding to the upper 600 m. Thus, in our simulations we used a Beta distribution with a mean value of 10% (i.e., 10% of the total available CO$_2$ is lost from the crust), and minimum and maximum values of 5% and 15%, respectively. For the pre-eruptive CO$_2$ content of basaltic magmas, we used a Beta distribution with a mean value of 0.5 wt%, and minimum and maximum values of 0.2 wt% and 2 wt%, considered reasonable for flood basalts and ocean crust at this time.

To estimate the CO$_2$ fluxes from LIPs, we used existing eruptive rate estimates for the NAIP. We consider two LIP eruptive rate scenarios: (S1) low flood basalt productivity (0.6 km$^3$ yr$^{-1}$); and (S2) high productivity (2.4 km$^3$ yr$^{-1}$; ref. 9). The combined fluxes from incipient ridge volcanism and each of these eruptive scenarios for LIPs are shown as the grey lines on our cumulative distribution functions (Fig. 3). We assumed near-total loss of CO$_2$ from flood basalt volcanism (mean 95%, minimum 90%, maximum 100% loss), based on observations from fresh basaltic glass in flood basalts that show most of the CO$_2$ is lost to degassing. Ridge and LIP CO$_2$ outgassing was calculated using equation 8:

$$W_{CO_2,\text{Ocean}} = \frac{12}{44} \frac{t_{\text{er}}}{10^{12}} (O_{\text{Floss}} \times \text{FractProduction}\times \text{FractCO}_2) \times (C_{\text{FractLIP}} \times \text{FractProduction} \times \text{FractCO}_2)$$

where $W_{CO_2,\text{Ocean}}$ = total weight of C released from the ocean crust and LIPs; $t_{\text{er}}$ = the duration of extension in years; $O_{\text{Floss}}$ = fraction of CO$_2$ lost from the ocean crust; $F_{\text{basalt}}$ = volumetric rate of basalt production from the NAIP ridges (see equation 1); $F_{\text{basalt}}$ = density of basalt (3000 kg m$^{-3}$); $B_{FractCO_2}$ = CO$_2$ content (weight percent) of basalt, expressed as a fraction;
where $W_{\text{CSCLM}} = \frac{l_{\text{CSCLM}} \cdot w_{\text{CSCLM}} \cdot b_{\text{CSCLM}} \cdot \rho_{\text{CSCLM}}}{10^{12}}$ (9)

where $W_{\text{CSCLM}} = \text{total weight of carbon in the SCLM (Gt)}$;

$L_{\text{CSCLM}} = \text{length of SCLM involved in melting along the North Atlantic mid-ocean ridge system}$;

$w_{\text{CSCLM}} = \text{width of the SCLM zone (km)}$;

$b_{\text{CSCLM}} = \text{thickness of the SCLM (km)}$,

and $\rho_{\text{CSCLM}} = \text{density of SCLM (herzolite; 3200 kg m}^{-3}$). A factor of $10^{12}$ converts weight from kg to Gt. Finally, we calculated the total weight of carbon ($W_{\text{TotalC}}$) produced from the ocean crust, LIPs and enhanced melting of the c-SCLM using equation 10:

$$W_{\text{TotalC}} = W_{\text{CO2Ocean}} + F_{\text{CSCLM}} W_{\text{CSCLM}}$$ (10)

where $F_{\text{CSCLM}}$ is the fraction of c-SCLM melted during extension (calculated with 0.04, 0.05 and 0.08; see Fig. 3).

Please note that additional references are cited in the Supplementary Figures and Tables.
M.P. provided support with geochemical analysis and interpretation. G.F. carried out the melt modelling. A.M. calculated the seafloor production rates and provided support with GPlates and pyGPlates. T.G. wrote the manuscript with input from all co-authors.

**Competing interests:**

The authors declare no competing interests.

**Additional information**

*Supplementary information* is available for this paper at https://doi.org/10.1038/s12345-111-2222-3.

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*Peer review information* *Nature Geoscience* thanks [reviewers] for their contribution to the peer review of this work.

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