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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 (CO₂) 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 >10⁴ gigatons of carbon, which is sufficient to drive PETM warming. Our model is consistent with anomalous CO₂ fluxes during continental breakup, while also reconciling the deficit of deep carbon required to explain the PETM.

1 During the early Cenozoic, a major episode of continental
2 breakup in the North Atlantic region (Fig. 1a) resulted in
3 widespread magmatism and the opening of the North Atlantic
4 Ocean^{1,3}. Initially the Iceland plume caused regional uplift
5 and a phase of subaerial volcanism lasting ~5 million years
6 (Myr)^{2,9,10}. Then, at ~56 million years ago (Ma), an incipient
7 mid-ocean ridge developed¹ (Fig. 1d), coinciding with an
8 order-of-magnitude increase in melt production rates³, and ac-
9 celerated westward migration of Greenland via ridge push⁸
10 (Fig. 1e). This transitional phase of volcanism in the North
11 Atlantic igneous province (NAIP) involved peak eruption rates
12 of ~2.4 km³ yr⁻¹ (ref.⁹) and occurred contemporaneously with
13 intense early Eocene warming, including the PETM¹¹.

14 During the PETM, sea surface temperatures increased by
15 ~5°C (ref.¹²), ocean acidification occurred^{13,14} and there was
16 an abrupt and short-lived (~170 thousand year, kyr)^{11,15} nega-
17 tive δ¹³C carbon isotope excursion (Fig. 1c). It is thought that
18 the release of approximately 1–1.5 × 10⁴ gigatons (Gt) of car-
19 bon into the ocean-atmosphere system is necessary to account
20 for these changes^{14,16}. This massive carbon flux has been at-
21 tributed to methane release associated with clathrate destabi-
22 lization along continental shelves¹⁷ and/or hydrothermal vent-
23 ing during interaction of magmas and organic-rich mudrocks¹⁸.

However, on the basis of paired δ¹¹B–δ¹³C data and carbon-
cycle modeling¹⁴, and B/Ca in planktic foraminifera¹⁶, it has
been proposed that triggering of the PETM required a short,
sharp injection of a ¹³C depleted source of carbon, possibly tied
to volcanic outgassing across the NAIP.

To test this hypothesis, we estimated the combined CO₂
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 productivity⁹ (Methods). In our model, we consid-
ered pre-eruptive CO₂ concentrations of 2 wt%, which are typ-
ical of flood basalt eruptions¹⁹. The associated CO₂ 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 km³ yr⁻¹), and estimated CO₂
degassing fluxes along the modern global ridge system (7 ×
10¹¹ mol yr⁻¹)²⁰. This calculation suggests that ~10% of the
CO₂ in the juvenile ocean crust is degassed at ridges. Using
this estimate, the NAIP ridges likely produced ~1.1 × 10³ Gt
C during the PETM. This is an order of magnitude lower than
the requisite ~1–1.5 × 10⁴ Gt C (refs.^{14,16}). Inclusion of the
effects of LIP magmatism—assuming most probable eruption
rates⁹ and total degassing—yields less than one-fifth of the car-
bon needed to drive and sustain the PETM^{14,16}. Furthermore,
seafloor isochrons indicate that magmatic productivity along

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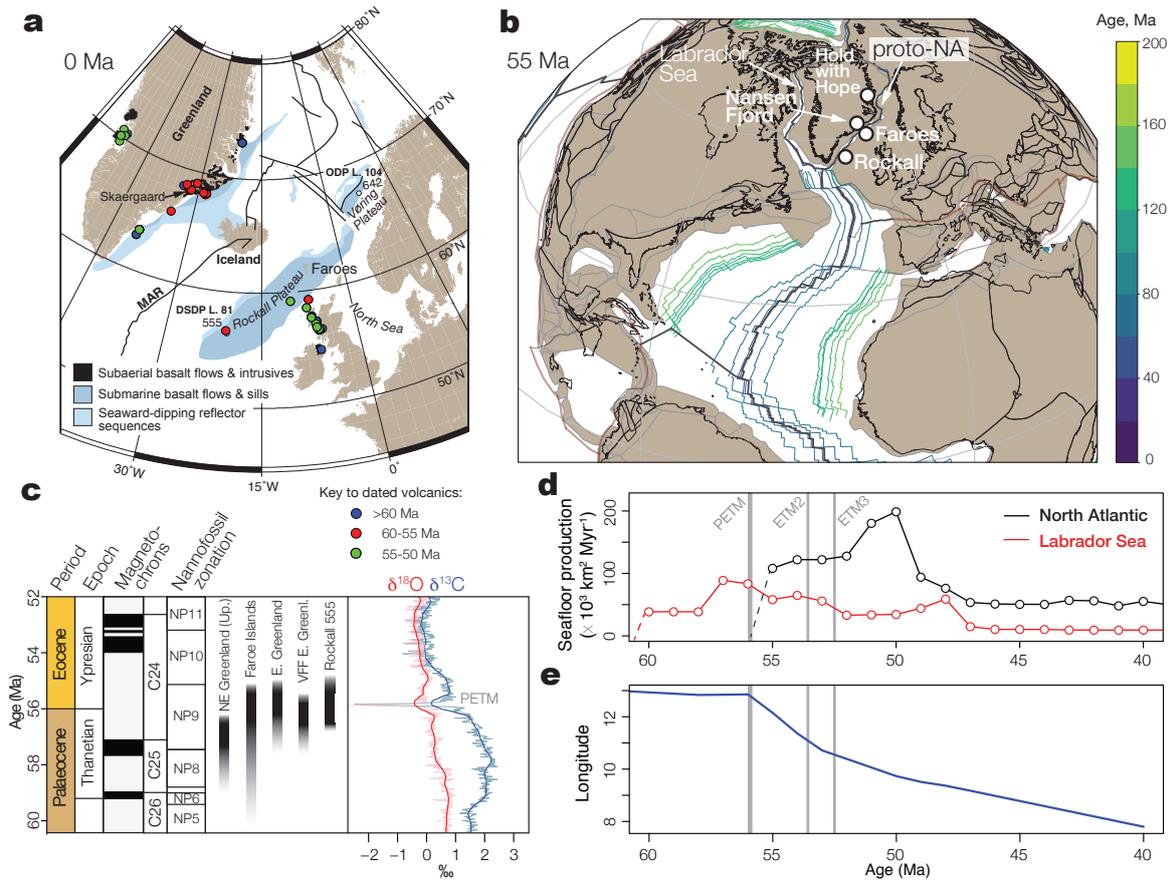


Figure 1. Early Cenozoic tectonic and magmatic evolution of the North Atlantic region | **a**, Map of the present-day North Atlantic region showing the distribution of Palaeocene–Eocene lava flows and intrusives¹, with dated volcanics denoted by colored symbols². **b**, Plate tectonic reconstruction 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^{1,3}, magnetostratigraphy and nannofossil zonation^{4,5,6}, 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**, Sea floor production rates for the Labrador Sea and North Atlantic, derived from GPlates (Methods), shown alongside the timing of Eocene hyperthermals. **(E)** Palaeolongitude of Greenland⁸ indicating the onset of ocean crustal production in the North Atlantic and ridge push at 56 Ma.

the North Atlantic ridge peaked after, not during, early Eocene hyperthermals (Fig. 1d). Therefore, some other major, but transient, source of volcanic carbon appears to be required if the volcanic outgassing hypothesis^{14,16} 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 volcanism²¹ (Fig. 2a) is coeval with the Milne Land basalts in East Greenland and the Middle to Upper Series lavas in the Faroe Islands¹ (Fig. 2b–c). In the Rockall sequence, we found a sharp increase in the frequency of volcanic tuffs just below the Palaeocene–Eocene boundary (Fig. 2a) (Methods). The PETM is defined by $\delta^{13}C$, however the volcanostratigraphy 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 paleontological age constraints, in addition to well-defined sediment accumulation rate estimates (Methods). Mudstones interbedded with the uppermost tuffs contain the dinoflagellate cyst, *Apectodinium augustum*²⁷, which is biostratigraphically diagnostic

of the PETM as it signifies a sudden prevalence of tropical sea-surface temperatures²⁸. 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^{11,15}, and was followed by a sharp decline in volcanism²¹. The tuffs exhibit wide compositional diversity from basanites to dacites (Supplementary Figs. 1, 2a–b), an increased range of magnesium number (Mg#; to a maximum observed value of 63), and a marked shift to highly negative ϵNd signatures at ca. 56.03 Ma (Fig. 2a). This activity signals a step change in magmatic processes and volcanic unrest along the ridge, as recorded across a major area of the NAIP (>130,000 km²)^{1,29}. Sampled PETM-age tuffs are enriched in Rb and Ba and depleted in Nb and Sr (Supplementary Fig. 2c), and have similar compositions to some of the lowermost (‘negative ash series’) tuffs of the Danish Basin, which likely derive from nearby volcanoes along the continental shelf²⁹. The geochemical similarities between tuffs from Rockall and the Danish Basin is consistent with paleogeography (Fig. 1b), palynological constraints²⁷ and the stratigraphic position of these tuffs towards the end of the PETM elsewhere in the NAIP⁶.

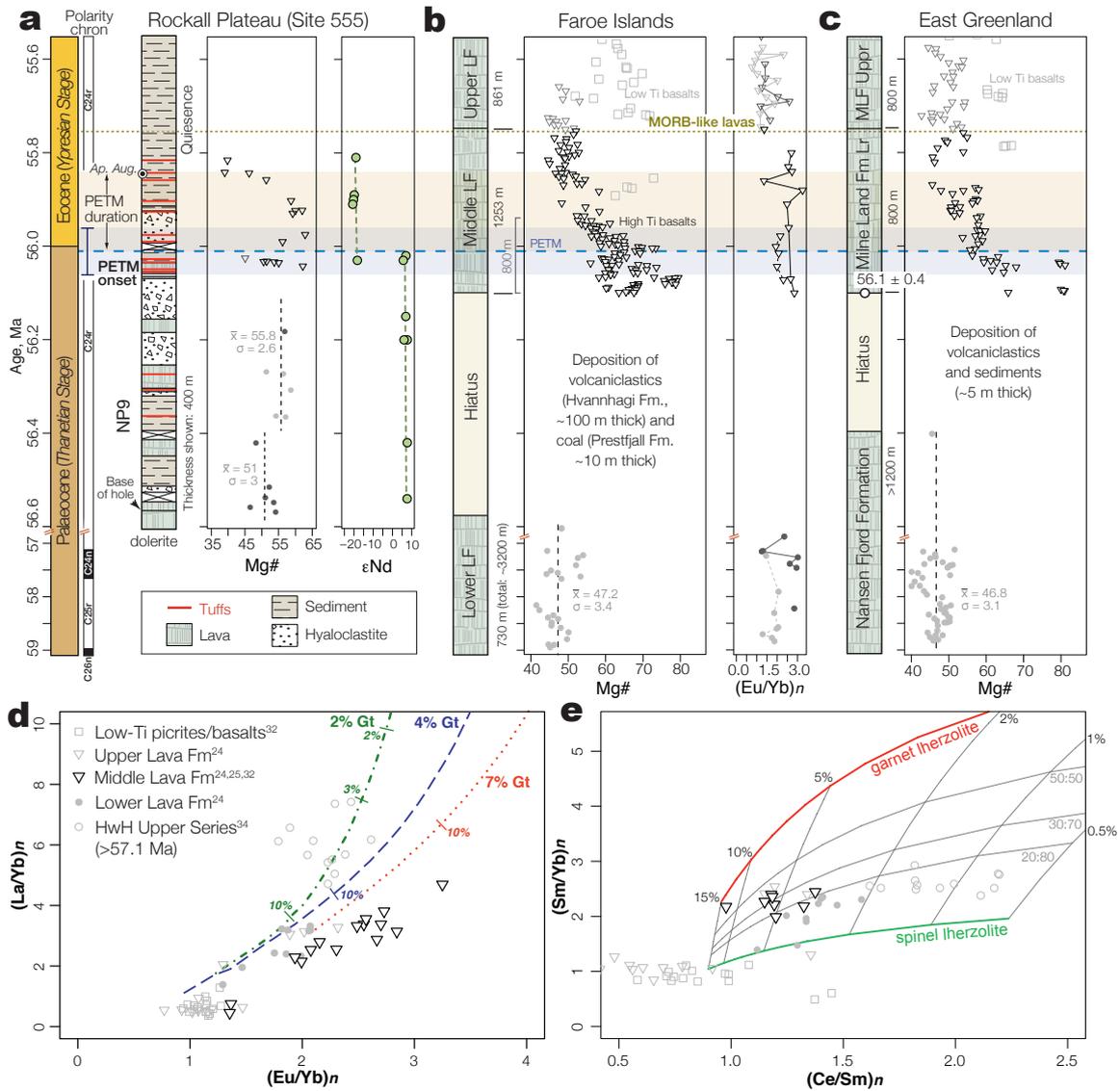


Fig. 2. Palaeocene-Eocene volcanostratigraphy and geochemistry of the proto-North Atlantic ridge. **a**, Simplified log of the Rockall ‘Phase 1’ sequence²¹ showing lithologies, Mg# (i.e., $100 \times \text{molecular MgO}/(\text{MgO} + \text{FeO})$, where FeO is assumed to be 0.9FeO), and ϵ_{Nd} (Supplementary Fig. 1). **b**, Simplified log of the Faroes Basalt Formations²², with Mg# and $(Eu/Yb)_n$ (chondrite-normalized²³); Mg# data are from²² and $(Eu/Yb)_n$ are from^{24,25}, note the sharp transition to high Mg# (and enriched REE contents) at ca. 56.1 Ma¹, which is also observed in **c**, east Greenland (Milne Land Formation)²². **d**, $(La/Yb)_n$ vs $(Eu/Yb)_n$ of the Faroes and Hold with Hope (HwH) lavas (chondrite-normalized²³) and modeled non-modal batch melting of a lherzolitic mantle source, adopted from²⁶, 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% gt-lherzolite (red curve) to 100% sp-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 $\geq 10\%$ garnet.

90 Interpretation of geochemical data from the Rockall tuffs is¹⁴⁷
 91 complicated because they derive from diverse sources and have¹⁴⁸
 92 experienced varying degrees of seafloor weathering²¹. Hence,¹⁴⁹
 93 we next studied the coeval thick basaltic lava sequences that¹⁵⁰
 94 were emplaced subaerially near the ridge axis (Fig. 1). The¹⁵¹
 95 Palaeocene-Eocene lavas of the Faroe Islands and East Green-¹⁵²
 96 land (Fig. 1b) are ideally suited because, unlike the contempo-¹⁵³
 97 raneous lavas in SE Greenland³⁰, they are minimally affected¹⁵⁴
 98 by crustal contamination^{22,24,31}. The base of the Milne Land¹⁵⁵
 99 Formation in East Greenland, which correlates with the base of¹⁵⁶
 100 the Middle Lava Series of the Faroes^{1,22} (Fig. 2b-c; hereafter¹⁵⁷
 101 both referred to as ‘MLF’), is dated at 56.1 ± 0.4 Ma and the¹⁵⁸
 102 duration of activity is well constrained^{1,3} (Fig. 2). Thus, the¹⁵⁹
 103 base of the MLF correlates with (or shortly predates) the inten-¹⁶⁰
 104 sification of volcanism at Rockall Plateau (Fig. 2a).¹⁶¹

105 We estimate that the 1.25 km-thick MLF package^{1,22} was¹⁶²
 106 emplaced over 200–300 kyr, yielding an average eruption rate¹⁶³
 107 of $\sim 4\text{--}6$ m kyr⁻¹, but potentially an order of magnitude higher¹⁶⁴
 108 early in the eruptive cycle¹. At the base of the sequence, thick¹⁶⁵
 109 (>100 m) pyroclastic deposits, also reported in Greenland²²,¹⁶⁶
 110 signal an early volatile-rich explosive phase. The overlying¹⁶⁷
 111 tholeiitic lavas are characterized by a sharp increase in Mg#,¹⁶⁸
 112 from values of ~ 52 throughout the Lower Lava Formation²² to¹⁶⁹
 113 >80 in the basal MLF (Fig. 2b-c). The basal lavas are coinci-¹⁷⁰
 114 dent (within dating uncertainties) with the onset of the PETM¹¹.
 115 Many of these lavas are highly magnesian³² (e.g., with MgO
 116 = 24%), implying hot liquidus temperatures, and are locally
 117 characterized by massive olivine accumulation²². They also
 118 exhibit high TiO₂ contents (typically 1.5–2.5 wt.%), and enrich-
 119 ment in Light Rare Earth Elements (LREEs), e.g., $(La/Yb)_n$
 120 = 2–3, and $(Eu/Yb)_n > 2.5$ (Fig. 2b)^{24,25}. This unusual pulse of
 121 high-Ti magmatism, similar to that of the Jurassic Karoo flood
 122 basalts in southern Africa²⁶, generated the long-lived (~ 300
 123 kyr)¹ Skaergaard layered igneous intrusion at 55.75 ± 0.35 Ma
 124 (Fig. 1a)¹. Finally, the Upper Lava Formation (Fig. 2b-c) signi-
 125 fies an abrupt shift to low-Ti depleted MORB-like basalts^{22,24,25}
 126 (Fig. 2d-e), thus, there is evidence that the early (high Mg#)
 127 MLF volcanism was both transient¹ (~ 227 kyr) and chemically
 128 anomalous (Fig. 2).

129 The geochemical compositions of these lavas have been attrib-
 130 uted to partial melting of the SCLM during asthenospheric
 131 upwelling²⁴, a model that is supported by studies of their feeder
 132 intrusions^{31,32}. To test this hypothesis further, we applied two
 133 models of mantle melting. First, we adopted a non-modal batch
 134 melting model of a garnet-bearing lherzolitic mantle source²⁶
 135 (Fig. 2d). For the second model, we estimated melt percent-
 136 age based on average lherzolite mineralogy, average partition
 137 coefficients, and a plume composition based on primitive basalt
 138 (Fig. 2e) (Methods). Both models suggest that the MLF experi-¹⁷¹
 139 enced a high degree of melting, up to $\sim 13\%$, but typically in the¹⁷²
 140 range of 4–8%, with their high $(Sm/Yb)_n$ ratios (2–2.5) requir-¹⁷³
 141 ing $\sim 20\text{--}50\%$ of garnet lherzolite in the mantle source (Fig.¹⁷⁴
 142 2e). Metasomatism in the mantle source region has already¹⁷⁵
 143 been documented in this region at precisely this time³³ and in-¹⁷⁶
 144 voked to explain the compositional characteristics of these high-¹⁷⁷
 145 Ti basalts, notably their LREE enrichment and variable Nb and¹⁷⁸
 146 Ta anomalies^{31,32}. The composition of the MLF is compatible¹⁷⁹

with enhanced melting of metasomatized SCLM by the Iceland
 plume, just prior to full seafloor spreading at c. 55.8 Ma²⁵ (Figs.
 2b–c).

The trace element compositions show that the degree of melt-
 ing varies systematically through the Faroes sequence (Fig. 2e),
 with the Lower Lava Formation (Fig. 2b-c) representing a rela-
 tively 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)³⁴ (Fig. 1b), which
 have an upper age of ca. 57 Ma (C25n–C24r)¹⁰. These lavas ex-
 hibit geochemical characteristics that have been attributed to the
 incorporation of SCLM during melting^{10,34}. Our model con-
 firms a low degree of melting (0.5–2%) of a variably garnet-
 rich lherzolite source (Fig. 2e). Taken together, these obser-
 vations 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–Eocene⁵ basaltic lavas of the Vandfaldsdalen For-
 mation in central east Greenland³⁵. These lavas exhibit Nd, Sr
 and Pb isotope signatures indicative of an SCLM component³⁵.
 Assimilation of the deep SCLM is particularly important be-
 cause this zone is metasomatically enriched in carbonates, and
 is thus a major carbon reservoir^{36,37,38}.

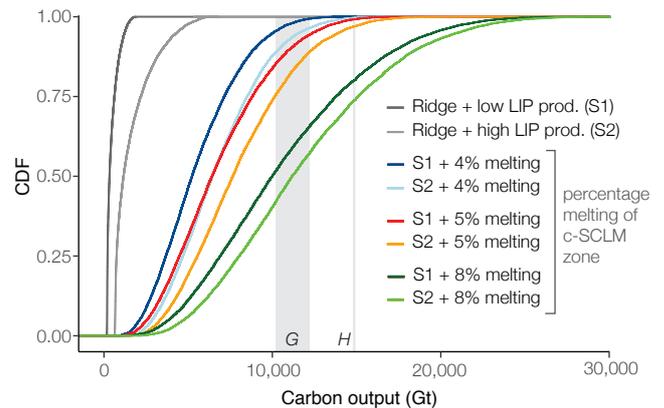


Figure 3. Simulations of volcanic carbon release during the PETM.
 Results are plotted as cumulative distribution functions (CDFs). The gray lines show the estimated carbon output from ridge volcanism and LIPs alone; with S1 and S2 showing low ($0.6 \text{ km}^3 \text{ yr}^{-1}$) and high ($2.4 \text{ km}^3 \text{ yr}^{-1}$) LIP eruption rate scenarios⁹, respectively (Supplementary Fig. 3) (see Methods). The coloured lines show the effects of adding 4% to 8% carbonated (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.¹⁴ (labelled G; 10,200–12,200 Gt C), and Haynes & Hönisch¹⁶ (labelled H; 14,900 Gt C).

The process of ocean basin formation significantly disrupts the >100 km thick SCLM³⁹. Because carbonates are only stable in the mantle at high pressures³⁷ (~ 3 GPa at 1300°C)³⁹, significant carbon release will only occur when extending cratons are at least 130 km thick³⁹. Seismic tomography indicates that the lithosphere in central Greenland (i.e., the locus of the Iceland plume at 56 Ma²) is 180 km thick⁴⁰; firmly meeting this criterion. Xenoliths provide additional evidence for coeval volatile-rich metasomatism in the deep SCLM be-

180 neath Greenland, as does the occurrence of carbonatite magma-
 181 tism across the wider NAIP at this time³³. Therefore, breakup
 182 of the North Atlantic craton provided a perfect combination
 183 of conditions: the availability of sufficiently thick metasoma-
 184 tized SCLM, analogous to the present-day Tanzanian craton
 185 in the East African Rift System³⁶; the long-term thermo-
 186 mechanical weakening and erosion of the SCLM by the Ice-
 187 land plume², broadly analogous to that proposed for the Permo-
 188 Triassic Siberian Traps³⁸; and the initiation of asthenospheric
 189 upwelling that induced vigorous interaction between anomalously
 190 hot melt and the weakened SCLM^{37,39}. A key aspect
 191 of our hypothesis is that the most extensive period of volcanic
 192 CO₂ outgassing (~50 Mt C yr⁻¹) occurred over a short period
 193 (~171–227 kyr), and likely peaked just prior to PETM onset
 194 (Fig. 2b-c). While continental rift volcanism is associated with
 195 enhanced CO₂ fluxes over millions of years, petrological and
 196 numerical models suggest that there is an initial short period
 197 of deep redox controlled carbonatitic to aillikitic melting (i.e.,
 198 high CO₂) that is followed by more widespread metasomatism
 199 of the shallower asthenosphere and generation of less CO₂-rich
 200 melts through decompression melting^{37,39}.

201 Both the thickness and composition of the NAIP lithospheric
 202 mantle make it a viable source of large amounts of carbon from
 203 the deep, carbonated SCLM during the PETM. A fundamental
 204 aspect of our model is that the occurrence of continental rift-
 205 ing opens a wide area along the incipient ridge to SCLM in-
 206 fluence. This contrasts with the end-Permian Siberian Traps,
 207 where there is no evidence for large-scale lithospheric stretch-
 208 ing during eruption³⁸. Indeed, it has been suggested that relics
 209 of this SCLM that delaminated during North Atlantic breakup²³⁷
 210 can explain residual enriched ‘EMI’ components present in the
 211 Icelandic mantle today^{41,42,43}. The question is then whether this²³⁹
 212 process can contribute enough carbon to help explain the PETM²⁴⁰
 213 carbon isotope excursion^{14,16}. Recent estimates suggest that²⁴¹
 214 the SCLM may contain 5–8% CO₂, or possibly more³⁷. The²⁴²
 215 present-day East African Rift offers a lower bound. Here, lat-²⁴³
 216 eral advection of SCLM along the craton margins results in an²⁴⁴
 217 order of magnitude increase in the volcanic CO₂ flux compared²⁴⁵
 218 to the background crustal contribution³⁶.

219 To more accurately quantify the potential influence of rift-²⁴⁷
 220 related SCLM disturbance, we advance on the approach taken²⁴⁸
 221 by Sobolev et al.³⁸ to estimate the most probable carbon re-²⁴⁹
 222 lease scenarios for the PETM. We performed 10,000 Monte²⁵⁰
 223 Carlo calculations using CO₂ contents ranging from 1–8%, and²⁵¹
 224 length-scales of the carbonated SCLM in the narrow melting²⁵²
 225 zone below the nascent ridge crest³⁹ based on tectonic recon-²⁵³
 226 structions, chemical tomography and lithospheric models (ta-²⁵⁴
 227 bles S7–S8) (see Methods). These calculations suggest that²⁵⁵
 228 only 4–8% of this zone needs to melt to exceed the requisite²⁵⁶
 229 >10⁴ Gt C for PETM warming^{14,16} (Fig. 3). This is con-²⁵⁷
 230 sistent with the modeled 5–8% melting of a garnet-rich lher-²⁵⁸
 231 zolite during the PETM (Fig. 2d-e), and earlier estimates that²⁵⁹
 232 the intrusive feeders of these Ti-rich basalts formed by 4%³² to²⁶⁰
 233 7%³¹ batch melting of fertile, metasomatized lherzolites in the²⁶¹
 234 SCLM³¹.

235 The step change in NAIP magmatic productivity³ immedi-²⁶³
 236 ately prior to the PETM gave rise to widespread volcanic un-²⁶⁴

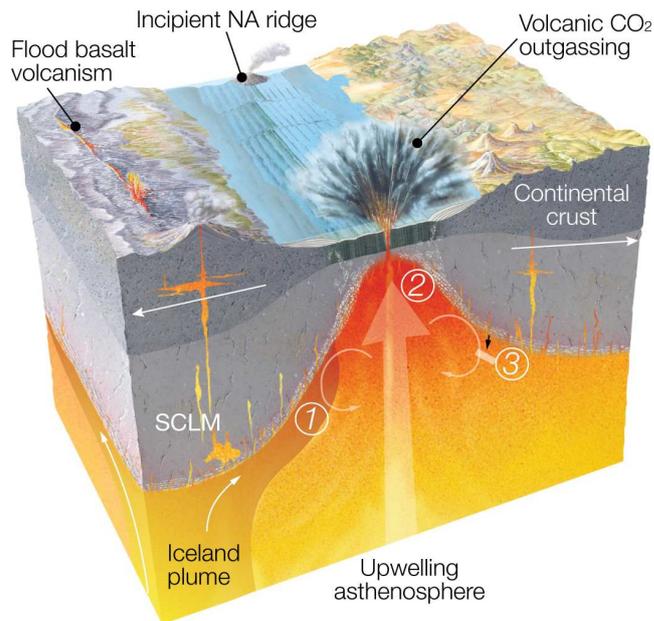


Figure 4. Volcanic carbon release in the North Atlantic during the PETM.

Model stages: **1**, progressive thermo-mechanical weakening of the sub-continental lithospheric mantle (SCLM) by the Iceland plume; **2**, disruption of the ruptured metasomatized SCLM zone by incipient asthenospheric upwelling; **3**, delamination of carbonated fragments of the deep SCLM during lithospheric stretching, which generate the enduring enriched components (‘EMI’) still present in the Icelandic mantle today⁴¹.

rest (Figs. 2, 4). While some volcanic tuffs at Rockall Plateau are likely linked to the MLF (especially those with high Mg#), many of these tuffs 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 time²⁹. 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 tuffs belonging to the Danish negative ash series²⁹. Indeed, the chemical weathering of extensive tephra blankets may also explain the large decrease in ¹⁸⁷Os/¹⁸⁸Os observed just prior to PETM onset at Svalbard⁴⁴.

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 ¹³C depleted carbon^{14,16} (Fig. 3). The melting of SCLM can result in a fivefold increase in volcanic CO₂ 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 craton⁴¹, and the anomalously high CO₂ fluxes from SCLM along craton margins during extension^{36,39}.

265 Our data-driven models of the PETM demonstrably support the
 266 proposal that large-scale lithospheric melting can induce global
 267 warming³⁸, if the tectonic setting is primed to facilitate intensive
 268 volcanic CO₂ degassing (Fig. 4). Our study highlights the
 269 critical role that solid Earth degassing plays in driving abrupt
 270 shifts in climate, and in promoting fundamental reorganisation
 271 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>.

References

1. M. Storey, R.A. Duncan, and C.C. Swisher. Paleocene-Eocene Thermal Maximum and the opening of the Northeast Atlantic. *Science*, 316:587–589, 2007.
2. B. Steinberger, E. Bredow, S. Lebedev, A. Schaeffer, and Trond H. T. Widespread volcanism in the Greenland–North Atlantic region explained by the Iceland plume. *Nature Geoscience*, 12(1):61–68, 2019.
3. M. Storey, R. A. Duncan, and C. Tegner. Timing and duration of volcanism in the North Atlantic Igneous Province: Implications for geodynamics and links to the Iceland hotspot. *Chemical Geology*, 241:264–281, 2007.
4. J. Backman, A. C. Morton, D. G. Roberts, S. Brown, K. Krumsiek, and R. M. Macintyre. Geochronology of the Lower Eocene and Upper Paleocene sequences of Leg 81 - Deep Sea Drilling Project (Rockall Plateau). In *DSDP Initial Reports*, volume 81, chapter 38, pages 877–882. 1984.
5. H. Nøhr-Hansen. Palynostratigraphy of the Cretaceous–lower Palaeogene sedimentary succession in the Kangerlussuaq Basin, southern East Greenland. *Review of Palaeobotany and Palynology*, 178:59–90, 2012.
6. E. W. Stokke, M. T. Jones, J. E. Tierney, H. H. Svensen, and J. H. Whiteside. Temperature changes across the Paleocene-Eocene Thermal Maximum – a new high-resolution TEX₈₆ temperature record from the Eastern North Sea Basin. *Earth and Planetary Science Letters*, 544:116388, 2020.
7. T. Westerhold, N. Marwan, A. J. Drury, D. Liebrand, C. Agnini, E. Anagnostou, J. S. K. Barnett, S. M. Bohaty, D. De Vleeschouwer, F. Florindo, T. Frederichs, D. A. Hodell, A. E. Holbourn, D. Kroon, V. Lauretano, K. Littler, L. J. Lourens, M. Lyle, H. Pälike, U. Röhl, J. Tian, R. H. Wilkens, P. A. Wilson, and J. C. Zachos. An astronomically dated record of Earth's climate and its predictability over the last 66 million years. *Science*, 369(6509):1383–1387, 2020.
8. R. N. Mitchell, T. M. Kilian, and D. A. D. Evans. Supercontinent cycles and the calculation of absolute palaeolongitude in deep time. *Nature*, 482(7384):208–211, 2012.
9. O. Eldholm and K. Grue. North Atlantic volcanic margins: Dimensions and production rates. *Journal of Geophysical Research*, 99(B2):2955–2968, 1994.
10. A. D. Saunders, J. G. Fitton, A. C. Kerr, M. J. Norry, and R. W. Kent. The north atlantic igneous province. In J. J. Mahoney and M. F. Coffin, editors, *Large Igneous Provinces*, volume 100, pages 45–94. AGU Geophysical Monograph Series, 1997.
11. R. E. Zeebe and L. J. Lourens. Solar System chaos and the Paleocene–Eocene boundary age constrained by geology and astronomy. *Science*, 365(6456):926–929, 2019.
12. Joost Frieling, Holger Gebhardt, Matthew Huber, Olabisi A. Adekeye, Samuel O. Akande, Gert-Jan Reichart, Jack J. Middelburg, Stefan Schouten, and Appy Sluijs. Extreme warmth and heat-stressed plankton in the tropics during the Paleocene-Eocene Thermal Maximum. *Science Advances*, 3(3):e1600891, 2017.
13. J. C. Zachos, G. R. Dickens, and R. E. Zeebe. An early Cenozoic perspective on greenhouse warming and carbon-cycle dynamics. *Nature*, 451:279–283, 2008.
14. M. Gutjahr, A. Ridgwell, P. F. Sexton, E. Anagnostou, P. N. Pearson, H. Pälike, R. D. Norris, E. Thomas, and G. L. Foster. Very large release of mostly volcanic carbon during the Palaeocene–Eocene Thermal Maximum. *Nature*, 548(7669):573–577, 2017.
15. U. Röhl, T. Westerhold, T. J. Bralower, and J. C. Zachos. On the duration of the Paleocene-Eocene thermal maximum (PETM). *Geochemistry, Geophysics, Geosystems*, 8(12):Q12002, 2007.
16. L. L. Haynes and B. Hönisch. The seawater carbon inventory at the Paleocene–Eocene Thermal Maximum. *Proceedings of the National Academy of Sciences*, page 202003197, 2020.
17. G. R. Dickens, J. R. O'Neil, D. K. Rea, and R. M. Owen. Dissociation of oceanic methane hydrate as a cause of the carbon isotope excursion at the end of the Paleocene. *Paleoceanography*, 10(6):965–971, 1995.
18. H. Svensen, S. Planke, A. Malthe-Sørenssen, B. Jamtveit, R. Myklebust, T. Rasmussen-Eldem, and S.S. Ray. Release of methane from a volcanic basin as a mechanism for initial Eocene global warming. *Nature*, 429:542–545, 2004.
19. S. Self, T. Thordarson, and M. Widdowson. Gas fluxes from flood basalt eruptions. *Elements*, 1(5):283–287, 2005.
20. D. Chavrit, E. Humler, and O. Grasset. Mapping modern CO₂ fluxes and mantle carbon content all along the mid-ocean ridge system. *Earth and Planetary Science Letters*, 387:229–239, 2014.
21. A. C. Morton and J. B. Keene. Paleogene pyroclastic volcanism in the southwest Rockall Plateau - Deep Sea Drilling Project Leg 81. In *DSDP Initial Reports*, volume 81, chapter 19, pages 633–643. 1984.
22. L. M. Larsen, R. Waagstein, A. K. Pedersen, and M. Storey. Trans-Atlantic correlation of the Palaeogene volcanic successions in the Faeroe Islands and East Greenland. *Journal of the Geological Society*, 156(6):1081–1095, 1999.
23. W. V. Boynton and P. Henderson. *Chapter 3 - Cosmochemistry of the Rare Earth Elements: Meteorite Studies*, volume 2, pages 63–114. Elsevier, 1984.
24. C. Gariépy, J. Ludden, and C. Brooks. Isotopic and trace element constraints on the genesis of the Faeroe lava pile. *Earth and Planetary Science Letters*, 63(2):257–272, 1983.
25. J-G. Schilling and A. Noe-Nygaard. Faeroe-Iceland plume: Rare-Earth evidence. *Earth and Planetary Science Letters*, 24(1):1–14, 1974.
26. F. Jourdan, H. Bertrand, U. Schärer, J. Blichert-Toft, G. Féraud, and A. B. Kampunzu. Major and trace element and Sr, Nd, Hf, and Pb isotope compositions of the Karoo Large Igneous Province, Botswana–Zimbabwe: Lithosphere vs mantle plume contribution. *Journal of Petrology*, 48(6):1043–1077, 2007.
27. S. Brown and C. Downie. Dinoflagellate cyst biostratigraphy of late Paleocene and early Eocene sediments from holes 552, 553A, and 555 - Deep Sea Drilling Project Leg 81 (Rockall Plateau). In *DSDP Initial Reports*, volume 81, chapter 13, pages 565–579. 1984.
28. A. Sluijs, S. Schouten, M. Pagani, M. Woltering, H. Brinkhuis, J. S. Sinninghe Damsté, G. R. Dickens, M. Huber, G. J. Reichart, R. Stein, J. Matthiessen, L. J. Lourens, N. Pedentchouk, J. Backman, K. Moran, and the Exhibition 302 Scientists. Subtropical Arctic Ocean temperature during the Palaeocene/Eocene thermal maximum. *Nature*, 441:610–613, 2006.
29. L. M. Larsen, J. G. Fitton, and A. K. Pedersen. Paleogene volcanic ash layers in the Danish Basin: compositions and source areas in the North Atlantic Igneous Province. *Lithos*, 71:47–80, 2003.
30. J. G. Fitton, L. M. Larsen, A. D. Saunders, B. S. Hardarson, and P. D. Kempton. Palaeogene continental to oceanic magmatism on the SE Greenland continental margin at 63°N: a review of the results of Ocean Drilling Program Legs 152 and 163. *Journal of Petrology*, 41(7):951–966, 2000.
31. J. Hansen, J. Davidson, D. Jerram, C. Ottley, and M. Widdowson. Contrasting TiO₂ compositions in early Cenozoic mafic sills of the Faeroe Islands: An example of basalt formation from distinct melting regimes. *Earth Sciences*, 8(5):235–267, 2019.
32. P. M. Holm, N. Hald, and R. Waagstein. Geochemical and Pb–Sr–Nd isotopic evidence for separate hot depleted and Iceland plume mantle sources for the Paleogene basalts of the Faeroe Islands. *Chemical Geology*, 178(1):95–125, 2001.

33. S. Aulbach, J. Sun, S. Tappe, H. E. Höfer, and A. Gerdes. Volatile-rich metasomatism in the cratonic mantle beneath SW Greenland: Link to kimberlites and mid-lithospheric discontinuities. *Journal of Petrology*, 58(12):2311–2338, 2018.
34. M. F. Thirlwall, B. G. J. Upton, and C. Jenkins. Interaction between Continental Lithosphere and the Iceland Plume—Sr-Nd-Pb Isotope Geochemistry of Tertiary Basalts, NE Greenland. *Journal of Petrology*, 35(3):839–879, 06 1994.
35. P. M. Holm. Nd, Sr and Pb isotope geochemistry of the Lower Lavas, E Greenland Tertiary Igneous Province. *Geological Society, London, Special Publications*, 39(1):181, 1988.
36. J. D. Muirhead, T. P. Fischer, S. J. Oliva, A. Laizer, J. van Wijk, C. A. Currie, H. Lee, E. J. Judd, E. Kazimoto, Y. Sano, N. Takahata, C. Tiberi, S. F. Foley, J. Dufek, M. C. Reiss, and C. J. Ebinger. Displaced cratonic mantle concentrates deep carbon during continental rifting. *Nature*, 582(7810):67–72, 2020.
37. S. F. Foley and T. P. Fischer. An essential role for continental rifts and lithosphere in the deep carbon cycle. *Nature Geoscience*, 10(12):897–902, 2017.
38. S. V. Sobolev, A. V. Sobolev, D. V. Kuzmin, N. A. Krivolutsкая, A. G. Petrunin, N. T. Arndt, V. A. Radko, and Y. R. Vasiliev. Linking mantle plumes, large igneous provinces and environmental catastrophes. *Nature*, 477(7364):312–316, 2011.
39. W. Gorczyk and C. M. Gonzalez. CO₂ degassing and melting of metasomatized mantle lithosphere during rifting—Numerical study. *Geoscience Frontiers*, 10(4):1409–1420, 2019.
40. F. A. Darbyshire, T. B. Larsen, K. Mosegaard, T. Dahl-Jensen, O. Gudmundsson, T. Bach, S. Gregersen, H. A. Pedersen, and W. Hanka. A first detailed look at the Greenland lithosphere and upper mantle, using Rayleigh wave tomography. *Geophysical Journal International*, 158(1):267–286, 2004.
41. V. Debaille, R. G. Trønnes, A. D. Brandon, T. E. Waight, D. W. Graham, and C.-T. A. Lee. Primitive off-rift basalts from Iceland and Jan Mayen: Os-isotopic evidence for a mantle source containing enriched subcontinental lithosphere. *Geochimica et Cosmochimica Acta*, 73(11):3423–3449, 2009.
42. T. H. Torsvik, H. E. F. Amundsen, R. G. Trønnes, P. V. Doubrovine, C. Gaina, N. J. Kusznir, B. Steinberger, F. Corfu, L. D. Ashwal, W. L. Griffin, S. C. Werner, and B. Jamtveit. Continental crust beneath southeast Iceland. *Proceedings of the National Academy of Sciences*, 112(15):E1818, 2015.
43. B. B. Hanan and J. G. Schilling. The dynamic evolution of the Iceland mantle plume: the lead isotope perspective. *Earth and Planetary Science Letters*, 151(1):43–60, 1997.
44. R. Wiczorek, M. S. Fantle, L. R. Kump, and G. Ravizza. Geochemical evidence for volcanic activity prior to and enhanced terrestrial weathering during the Paleocene Eocene Thermal Maximum. *Geochimica et Cosmochimica Acta*, 119:391–410, 2013.

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272 Methods

273 *Calculating seafloor production rates*

274 Seafloor production (*SP*) rates (Fig. 1d), which were
275 utilised in our volcanic CO₂ flux calculations (see page
276 10), were calculated using the plate model of Müller et al.
277 (2016)⁴⁵. We used the open source python library, *pyGPlates*
278 (<https://www.gplates.org/docs/pygplates/>) in order to filter and
279 extract the data from the plate model. For the target time-steps
280 (*t*, in Myr), we broke the complete mid-ocean ridge system into
281 a series of spreading and transform segments in order to isolate
282 the spreading segments (*j*) where new oceanic crust is formed.
283 This approach has recently become common in analysing tectonic
284 scenarios in ocean basins^{46,47,48,49}. At each segment, for
285 each time-step, we extracted the full spreading rate (*u*, in km
286 Myr⁻¹) and the length of the spreading segment (*L*, in km). The
287 full spreading rate was calculated by summing the half spreading
288 rate of each individual flank of each ridge segment, thereby
289 sidestepping any issues pertaining to asymmetric spreading. We
290 did this because, for the purposes of our analysis, we were only
291 interested in the total amount of new seafloor generated. We
292 then took the product of the spreading segment length and full
293 spreading rate to calculate the seafloor production rate (in km²
294 Myr⁻¹), and then summed the area of all segments per time-
295 step, to obtain a total seafloor production per Myr (equation 1),
296 as follows:

$$297 \quad SP(t) = \sum_j uL \quad (1)$$

297 *Geochemical analysis of the volcanic tuffs*

298 We carried out major, trace and isotopic analysis (variously)
299 on 20 tuff layers sampled from DSDP Site 555 on the Rockall
300 Plateau. X-ray fluorescence (XRF) analyses were carried out in
301 the Grant Institute of Earth Science at the University of Edinburgh,
302 using procedures described in refs.^{50,51}. Major-element
303 concentrations (Supplementary Table 1) were determined after
304 fusion with a lithium borate flux containing La₂O₃ as a
305 heavy absorber, using an existing method⁵². Rock powder was
306 dried at 110°C for at least 1 hour, and a precisely-weighed 1-g
307 aliquot ignited at 1100°C to determine loss on ignition (LOI).
308 The residue was then mixed with Johnson Matthey Spectroflux
309 105 in a sample:flux ratio of 1:5, based on the unignited sam-
310 ple mass, and fused in a muffle furnace in a Pt5%Au crucible.
311 After the initial fusion, the crucible was reweighed and any flux
312 weight loss was made up with extra flux. After a second fusion
313 over a Meker burner, the molten mixture was swirled several
314 times to ensure homogeneity, cast onto a graphite mold, and
315 flattened with an aluminium plunger into a thin disk. The mold
316 and plunger were maintained at a temperature of 220°C on a
317 hotplate.

318 Trace-element concentrations (Supplementary Table 1) were
319 determined on pressed-powder samples. Eight grams of rock
320 powder were mixed thoroughly with eight drops of a 2% aqueous
321 solution of polyvinyl alcohol. The mixture was loaded into
322 a 40-mm diameter aluminium cup in a stainless steel die and

323 compressed against a polished tungsten carbide disc in a hy-
324 draulic press at 0.6 tons cm⁻². The fused and pressed samples
325 were analysed using a PANalytical PW 2404 automatic X-ray
326 fluorescence spectrometer with a Rh-anode X-ray tube. Trace-
327 element background positions were placed as close as possible
328 to peaks, and long count times were used at both peak and back-
329 ground positions. Where background count rates were mea-
330 sured on either side of the peak, as in most trace-element de-
331 terminations, the count time was divided between the two posi-
332 tions. Analytical conditions are given in refs.^{50,51}.

333 Corrections for matrix effects on the intensities of major-
334 element lines were made using theoretical alpha coefficients
335 calculated on-line using the PANalytical software. The coef-
336 ficients were calculated to allow for the amount of extra flux
337 replacing volatile components in the sample so that analytical
338 totals should be 100% less than the measured LOI. Intensities of
339 the longer wavelength trace-element lines (La, Ce, Nd, Cu, Ni,
340 Co, Cr, V, Ba, and Sc) were corrected for matrix effects using
341 alpha coefficients based on major-element concentrations mea-
342 sured at the same time on the powder samples. Matrix correc-
343 tions were applied to the intensities of the other trace-element
344 lines by using the count rate from the RhK_α Compton scatter
345 line as an internal standard⁵³. Line-overlap corrections were
346 applied using synthetic standards.

347 The spectrometer was calibrated against USGS and CRPG
348 geochemical reference standards using the values given by
349 Govindaraju (1994)⁵⁴, except that the values of Jochum et al.
350 (1990)⁵⁵ were used for Nb and Zr in BCR-1 and BHVO-1. Ex-
351 cellent calibration lines were obtained using these standards.
352 Analytical precision and accuracy are comparable to the values
353 reported in refs.^{50,51}. Four USGS geostandards were analysed
354 along with the samples and the data from these (Supplementary
355 Table 2) were used to assess accuracy and precision.

356 Trace element analysis was performed on a representative se-
357 lection of four of the tuffs using an Inductively Coupled Plasma
358 Mass Spectrometer (ICP-MS). Samples were homogenised us-
359 ing a unidirectional crushing machine prior to dissolution via
360 a three-stage, mixed acid (HF-HNO₃-HCl) closed-vessel ap-
361 proach. After digestion, samples were dried down prior to
362 resuspension in 2% HNO₃ for analysis, carried out using a
363 Thermo Scientific X-Series-2 at the University of Southamp-
364 ton. Alongside samples, blanks and International standard
365 JA-2 (ref.⁵⁶) and Southampton internal basalt standard BRR-
366 1 (ref.⁵⁷) were prepared and analysed (Supplementary Table 4).
367 Recoveries of JA-2 and BRR-1 are shown in Supplementary
368 Table 5.

369 Finally, we analysed the ¹⁴³Nd/¹⁴⁴Nd isotope composition
370 of several samples of tuff from Site 555 (Fig. 2a; Supplemen-
371 tary Table 3). Here, ~25 mg of freeze-dried sediment was ho-
372 mogenised using a pestle and mortar prior to leaching in 6M
373 HCl for 2 hours to remove any diagenetic, alteration-related
374 material. Samples were then digested via a mixed acid, closed
375 vessel HNO₃-HF approach on a hotplate at 120°C. Digests were
376 then taken to incipient dryness prior to resuspension in 2%
377 HNO₃ for analysis. All acids were sub-boiled to ensure pu-
378 rity and prepared at the University of Southampton. Aliquots
379 were then purified via first cation (AG50-X8 200-400 mesh

resin) and then reverse phase (LN Spec, Eichrom Industries)⁴³² chromatography to remove isobaric interferents. All measure-⁴³³ments were made on a Thermo Fisher Neptune Multi-Collector-⁴³⁴ICP-MS at the University of Southampton. Mass bias correc-⁴³⁵tions were performed using repeat measurements of the stan-⁴³⁶dard JNdi-1 (ref.⁵⁸) with a ¹⁴³Nd/¹⁴⁴Nd value of 0.512115. Nd⁴³⁷ isotope ratios are presented using epsilon notation as follows: ⁴³⁸

$$\epsilon Nd = \left(\frac{{}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{sample}}}{{}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{CHUR}}} - 1 \right) \times 10^4 \quad (2)$$

where ¹⁴³Nd/¹⁴⁴Nd_{CHUR} is the Nd isotopic composition of the ⁴⁴²chondrite uniform reservoir, with a value of 0.512638 (ref.⁵⁹).⁴⁴³ Precision of ¹⁴³Nd/¹⁴⁴Nd (2 SE) was between 0.000005 and⁴⁴⁴ 0.00001, corresponding to ϵ Nd precision of between 0.1 and⁴⁴⁵ 0.2.⁴⁴⁶

Establishing a chronostratigraphic framework

We established a chronostratigraphic framework for Site 555⁴⁵⁰ on the Rockall Plateau, building on a number of existing con-⁴⁵¹straints at this site (Fig. 1a). The PETM as geochemically de-⁴⁵²scribed could not be identified at this site, because of the high⁴⁵³ proportion of volcanic and siliciclastic material (Fig. 2a), in-⁴⁵⁴truded at many levels by dolerite dykes and sills with emplace-⁴⁵⁵ment temperatures likely in the range $\sim 1000^\circ$ to 1250° . Thus,⁴⁵⁶ the adjacent sediments likely experienced temperatures $> 650^\circ$,⁴⁵⁷ thermal conditions which are known to seriously affect $\delta^{13}\text{C}$ ⁴⁵⁸ composition in mudrocks and carbonates⁶⁰. Accordingly, we⁴⁵⁹ rely on a combination of radiometric, magnetostratigraphic and⁴⁶⁰ paleontological age constraints, in addition to well-defined sed-⁴⁶¹iment accumulation rate estimates. The most reliable radiomet-⁴⁶²ric date in this sequence, from an olivine phyric basalt near the⁴⁶³ Palaeocene-Eocene boundary, yielded a K-Ar age of 54.5 ± 2 ⁴⁶⁴ Ma (ref.⁶¹). Although uncertain, this can be narrowed further⁴⁶⁵ by considering the distribution of calcareous nannoplankton, in-⁴⁶⁶cluding *Fasciculithus*, which disappears in the mudstones be-⁴⁶⁷low the hyaloclastites (top NP9), with its last occurrence dated⁴⁶⁸ to ~ 55.6 Ma (<http://www.mikrotax.org>). This suggests that the⁴⁶⁹ age of the lava is most likely in the range 56.5–55.6 Ma, sup-⁴⁷⁰porting the original position of the Palaeocene-Eocene bound-⁴⁷¹ary⁶². Further, the overlying mudstones interbedded with the⁴⁷² tuffs contain the dinoflagellate cyst species, *Apectodinium au-*⁴⁷³*gustum*²⁷, characteristic of the TP5a palynozone⁶³. Therefore,⁴⁷⁴ most of the tuff layers at Site 555 (Fig. 2) were emplaced im-⁴⁷⁵mediately prior to, and during the PETM. We estimate the dura-⁴⁷⁶tion of this eruptive phase by calculating the thickness of strata⁴⁷⁷ between the bottom and upper tuffs in the phase (i.e., 691 and⁴⁷⁸ 605.5 mbsl, respectively), and using the sediment accumulation⁴⁷⁹ rates (50 cm kyr^{-1}) at these levels at Site 555⁴. This gives a dura-⁴⁸⁰tion of 171 kyr, which is near-identical to that of the contem-⁴⁸¹poraneous PETM interval^{11,15}. However, given that the earli-⁴⁸²est tuffs occur just below the Palaeocene-Eocene boundary, the⁴⁸³ true duration is more likely to be of the order of 213 kyr, assum-⁴⁸⁴ing that the PETM section represents the 170 kyr interval^{11,15}.⁴⁸⁵ This interpretation is consistent with the occurrence of tuffs in⁴⁸⁶ the upper part of the sequence (Supplementary Fig. 2) that are⁴⁸⁷ geochemically similar to those reported in the PETM recovery⁴⁸⁸

phase elsewhere in the NAIP⁴. We therefore consider a dura-⁴⁸⁹tion range of 171–213 kyr to be most reasonable for this eruptive⁴⁹⁰ phase. Given the above observations, we estimate the age of basal⁴⁹¹ strata in this hole (~ 950 m) at 56.6 Ma, which is consis-⁴⁹²tent with biostratigraphy and magnetostratigraphy⁴.

The base of the Milne Land Formation (Lower), which is cor-⁴⁹³related with the base of the Faeroes Middle Lava Formation^{1,22}⁴⁹⁴ has been well-dated at 56.1 ± 0.4 Ma. Storey et al. (2007)¹⁴⁹⁵ use the relative age of the well-dated Danish Ash-17 to firmly⁴⁹⁶ place the PETM interval after the onset of MLF volcanism. A⁴⁹⁷ late Palaeocene age is also supported by palynologic studies of⁴⁹⁸ the underlying (non-marine) coal-bearing Prestfjall Formation,⁴⁹⁹ and its position near the boundary between magnetic polarity⁵⁰⁰ chrons c24r and c25n⁶⁴ (i.e., 57.1 Ma; ref.⁶⁵). For the Upper⁵⁰¹ Lava Formation (ULF), we assumed an upper age of 55.5 Ma;⁵⁰² this is based on an average of three radiometric ages, includ-⁵⁰³ing one obtained from the upper ULF in the Faeroes ($55.1 \pm$ ⁵⁰⁴ 0.5 Ma; ref.¹). We used two radiometric ages obtained from⁵⁰⁵ the Skaergaard intrusion (55.75 ± 0.3 Ma, and 55.65 ± 0.3 Ma;⁵⁰⁶ ref.⁶⁶), where the parental magma has been genetically linked⁵⁰⁷ to the Milne Land Formation⁶⁷, and accordingly its crystallisa-⁵⁰⁸tion is considered to provide an upper age constraint for Milne⁵⁰⁹ Land Formation volcanism¹ (and thus, the lower Geikie Plateau⁵¹⁰ Formation²²). Using this age model, we calculate the duration⁵¹¹ of the lowermost high Mg# part of the MLF (i.e., the lowermost⁵¹² 800 m; Fig. 2b) to be 227 kyr, similar to that of the volcanic⁵¹³ phase at Rockall.

Establishing a chronostratigraphic framework for the Faroese⁵¹⁴ Lower Lava Formation (correlative with the Nansen Fjord For-⁵¹⁵mation of east Greenland) is more challenging because of the⁵¹⁶ large uncertainties associated with radiometric ages through-⁵¹⁷out this succession³. However, this inherent uncertainty does not⁵¹⁸ impact our results in any way; data for the lower formations are⁵¹⁹ shown only for reference purposes on our stratigraphic section⁵²⁰ (Fig. 2b–c). As a reasonable solution, we used magnetic polar-⁵²¹ity chrons for the Faroese sequence⁶⁸, combined with the chem-⁵²²ical stratigraphy of Larsen et al. (1999)²² and using a standard⁵²³ geomagnetic polarity time scale⁶⁵. We calibrated the ages at⁵²⁴ certain depths in the succession using the ages of chron bound-⁵²⁵aries as tie points, and applying a linear interpolation between⁵²⁶ these known points (which assumes constant lava accumula-⁵²⁷tion rates). We found that this age model was broadly consis-⁵²⁸tent with existing radiometric ages (i.e., in most cases within⁵²⁹ uncertainty)³.

REE melting model

A simple batch melting equation was used in the construction⁵³⁰ of Figure 2e:

$$C_L/C_0 = \frac{1}{D + F - PF} \quad (3)$$

where C_0 is the initial concentration of some element in the⁵³¹ mantle source, C_L is its concentrations in the liquid, F is⁵³² the melt fraction, D is the average distribution coefficient for⁵³³ the mantle phases weighted by their respective mass fractions⁵³⁴

483 before the onset of melting, and P is the average distribu-526
 484 tion coefficient for the mantle phases weighted by their re-527
 485 spective contribution to the melt. We use a peridotite mantle528
 486 source with a mineralogical composition given by McKenzie529
 487 and O’Nions (1991)⁶⁹: (1) 57.8% olivine, 27.0% orthopyrox-530
 488 ene, 11.9% clinopyroxene and 3.3% spinel (spinel lherzolite);531
 489 and (2) 59.8% olivine, 21.1% orthopyroxene, 7.6% clinopy-532
 490 roxene and 11.5% garnet (garnet lherzolite). Partition coeffi-533
 491 cients (D) vary significantly with pressure, temperature and liq-534
 492 uid composition, and this limits the reliability of partial melting535
 493 models since the choice of values is always somewhat subject-536
 494 tive. Here we have attempted to be more objective by using537
 495 averages of all appropriate published D values (Supplementary538
 496 Table 6). We use the following mantle melting proportions for539
 497 spinel lherzolite (equation 4; ref.⁷⁰) and garnet lherzolite (equa-540
 498 tion 5; ref.⁷¹):

$$441 \text{ liquid} = -0.22 \text{ ol} + 0.38 \text{ opx} + 0.71 \text{ cpx} + 0.13 \text{ sp} \quad (4)_{444}$$

$$445 \text{ liquid} = 0.08 \text{ ol} - 0.19 \text{ opx} + 0.81 \text{ cpx} + 0.30 \text{ gt} \quad (5)_{448}$$

499 where ol is olivine, opx is orthopyroxene, sp is spinel, cpx is549
 500 clinopyroxene and gt is garnet.550

501 The concentrations of REE in our mantle source (Ce 1.236551
 502 ppm, Sm 0.346 ppm, Yb 0.432 ppm) are calculated from the552
 503 composition of primitive basalt from the Ontong Java Plateau553
 504 and are thought to be typical of mantle plumes⁷².554

505
 506 Please note that Supplementary Table 6 cites additional refer-556
 507 ences^{73,74,75,76,77,78,79,80}.557

508 *Modelling of CO₂ fluxes*

509 We perform a simple Monte Carlo simulation, sampling from560
 510 probability distributions for six uncertain parameters (Supple-561
 511 mentary Tables 7–8) to estimate the probable combined CO₂562
 512 emissions from incipient ridge volcanism and active large igne-563
 513 ous provinces (LIPs). This enables us to evaluate the relative564
 514 contributions to carbon release during the PETM due to ridge565
 515 production and LIP formation. We also calculate the effect of566
 516 incorporating carbonated SCLM melt in different amounts dur-567
 517 ing enhanced melting along the ridge axis. In our calculations,568
 518 we assumed a PETM duration of 170 kyr (ref.¹⁵), which is sup-569
 519 ported by recent astrochronological solutions¹¹.570

520 We use Beta distributions to represent uncertainty in the pa-571
 521 rameters (Supplementary Table 8). The Beta distribution is a572
 522 continuous distribution over a fixed interval [0, 1], but can be573
 523 rescaled to any desired range. It is defined by two shape pa-574
 524 rameters α and β , which can be estimated from the distribution575
 525 mean (μ) and variance (σ^2):

$$576 \alpha = \mu \left(\mu \frac{(1 - \mu)}{\sigma^2} - 1 \right) \quad (6)_{574}$$

$$577 \beta = \alpha \left(\frac{1}{\mu} - 1 \right) \quad (7)_{577}$$

where μ = the mean, σ = standard deviation, and σ^2 = the vari-
 ance. To estimate Beta distribution parameters, we use best esti-
 mates (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 μ and σ). For simplic-
 ity we assume that the standard deviation for a given variable is
 20% of the range, $\sigma_v = 0.2$ (max–min). The inputs to the cal-
 culations (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 CO₂ 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 reconstruc-
 tions (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 average⁸¹. Only a small fraction of the
 total amount of CO₂ 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 $\sim 18 \text{ km}^3 \text{ yr}^{-1}$) and the estimated CO₂ fluxes
 from the present-day global ridge system ($7 \times 10^{11} \text{ mol yr}^{-1}$; a
 middle value of ref.²⁰). This analysis indicates that about 10%
 of the CO₂ available in the ocean crust is degassed at ridges (see
 main text), corresponding to the upper 600 m. Thus, in our sim-
 ulations we used a Beta distribution with a mean value of 10%
 (i.e., 10% of the total available CO₂ is lost from the crust), and
 minimum and maximum values of 5% and 15%, respectively.
 For the pre-eruptive CO₂ content of basaltic magmas, we used
 a Beta distribution with a mean value of 0.5 wt%, and mini-
 mum and maximum values of 0.2 wt% and 2 wt%, considered
 reasonable for flood basalts¹⁹ and ocean crust at this time⁸².

To estimate the CO₂ fluxes from LIPs, we used existing erup-
 tive rate estimates for the NAIP⁹. We consider two LIP erup-
 tive rate scenarios: (S1) low flood basalt productivity (0.6 km^3
 yr^{-1}); and (S2) high productivity ($2.4 \text{ km}^3 \text{ yr}^{-1}$; 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 CO₂ 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
 CO₂ is lost to degassing¹⁹. Ridge and LIP CO₂ outgassing was
 calculated using equation 8:

$$578 W_{CO_{\text{Ocean}}} = \frac{12}{44} \frac{t_{\text{rift}}}{10^{12}} (O_{\text{Floss}} \cdot r_{\text{basaltprod}} \cdot \rho_{\text{basalt}} \cdot B_{\text{FractCO}_2}) \quad (8)$$

$$579 + (C_{\text{FractLIP}} \cdot r_{\text{LIPprod}} \cdot \rho_{\text{basalt}} \cdot B_{\text{FractCO}_2})$$

580 where $W_{CO_{\text{Ocean}}}$ = total weight of C released from the ocean crust
 581 and LIPs; t_{rift} is the duration of extension in years; O_{Floss} =
 582 fraction of CO₂ lost from the ocean crust; $r_{\text{basaltprod}}$ = volumet-
 583 ric rate of basalt production from the NAIP ridges (see equa-
 584 tion 1); ρ_{basalt} = density of basalt (3000 kg m^{-3}); B_{FractCO_2} =
 585 CO₂ content (weight percent) of basalt, expressed as a fraction;

578 $C_{FractLIP}$ = fraction of LIPs that is fully degassed of CO_2 ; and
 579 $r_{LIPprod}$ = volumetric rate of basalt production from LIPs. Note
 580 that a factor of 12/44 converts from mass of CO_2 to mass of C.

581 The combined carbon flux estimates show that outgassing
 582 from ridges and LIPs alone are insufficient to trigger PETM
 583 warming, unless they are augmented by a transient increase in
 584 CO_2 content (as we propose, via the incorporation of carbonated
 585 SCLM during mantle melting). To account for this apparent deficit,
 586 we estimated the potential influence of SCLM melting contributions,
 587 using scenarios S1 and S2 (above) as a baseline (i.e., the background
 588 volcanic carbon flux; Fig. 3). During continental extension (as in the
 589 early Cenozoic North Atlantic) the deep metasomatized mantle lithosphere
 590 is stretched, exhumed, and melted along the length of the newly-formed
 591 ridge³⁹. We estimated the total volume of carbonated SCLM (c-SCLM)
 592 from the length of the ridge segments active between 56–55 Ma (Fig. 1d),
 593 assuming a plausible range of thicknesses and melting widths for this
 594 layer (Supplementary Table 8).

596 The precise thickness of the c-SCLM of the North Atlantic
 597 craton at 55 Ma is poorly constrained and is likely to have been
 598 spatially heterogeneous, but is considered to lie in the range 10
 599 km (ref.³⁹) to ~30 km thick (based on chemical tomography of
 600 cratonic regions⁸³). This is broadly consistent with geochemical
 601 and tectono-magmatic models of the North Atlantic craton^{84,85}. We
 602 assume a mean c-SCLM thickness of 20 km, with minimum and maximum
 603 values of 5 and 25 km. We estimated the width of SCLM involved in
 604 melting, using petrological-thermomechanical models of lithospheric
 605 rifting processes³⁹. In cases where the lithosphere is about 200 km
 606 thick, the width of the melting zone is most likely to be of the order
 607 of 25 km (ref.³⁹). We therefore used a Beta distribution with a mean
 608 width of 25 km and range of 5 to 30 km. SCLM melting occurs on
 609 both sides of the stretching lithosphere, doubling the total contribution
 610 to CO_2 output. c-SCLM is expected to have a high CO_2 content, of
 611 the order 5–8 wt%, but potentially even higher³⁷. We therefore use
 612 a mean value of 5 wt%, minimum of 1 wt% and maximum of 10 wt%.
 613 The total contribution of SCLM melting to fluxes was estimated using
 614 equation 9:

$$W_{CSCLM} = \frac{l_{SCLM} \cdot w_{SCLM} \cdot h_{SCLM} \cdot \rho_{SCLM}}{10^{12}} \quad (9)$$

616 where W_{CSCLM} = total weight of carbon in the SCLM (Gt);
 617 l_{SCLM} = length of SCLM involved in melting along the North
 618 Atlantic mid-ocean ridge system; w_{SCLM} = width of the SCLM
 619 zone (km); h_{SCLM} = thickness of the SCLM (km), and ρ_{SCLM} =
 620 density of SCLM (lherzolite; 3200 kg m^{-3}). A factor of $1/10^{12}$
 621 converts weight from kg to Gt. Finally, we calculated the total
 622 weight of carbon (W_{TotalC}) produced from the ocean crust, LIPs
 623 and enhanced melting of the c-SCLM using equation 10:

$$W_{TotalC} = W_{COcean} + F_{CSCLM} W_{CSCLM} \quad (10)$$

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

626 Please note that additional references are cited in the Supplementary
 627 Figures and Tables^{86,87,88,89,90,91}.

Data availability

All data generated or analysed during this study are provided in the online version of this article and in Supplementary Tables 1–8.

Code availability

The numerical modelling codes associated with this paper are available from the corresponding author (Thomas.Gernon@noc.soton.ac.uk) upon reasonable request. The map in Fig. 1a was plotted with open software GMT (under a GNU Lesser General Public License), and the map in Fig. 1b was plotted with open software GPlates (licensed for distribution under a GNU General Public License).

References

45. R. D. Müller, M. Seton, S. Zahirovic, S. E. Williams, K. J. Matthews, N. M. Wright, G. E. Shephard, K. T. Maloney, N. Barnett-Moore, M. Hosenipour, D. J. Bower, and J. Cannon. Ocean basin evolution and global-scale plate reorganization events since Pangea breakup. *Annual Review of Earth and Planetary Sciences*, 44(1):107–138, 2016.
46. K. S. Karlsen, C. P. Conrad, and V. Magni. Deep water cycling and sea level change since the breakup of Pangea. *Geochemistry, Geophysics, Geosystems*, 20(6):2919–2935, 2019.
47. K. S. Karlsen, M. Domeier, C. Gaina, and C. P. Conrad. A tracer-based algorithm for automatic generation of seafloor age grids from plate tectonic reconstructions. *Computers & Geosciences*, 140:104508, 2020.
48. A. S. Merdith, S. E. Atkins, and M. G. Tetley. Tectonic controls on carbon and serpentinite storage in subducted upper oceanic lithosphere for the past 320 Ma. *Frontiers in Earth Science*, 7:332, 2019.
49. A. S. Merdith, P. G. del Real, I. Daniel, M. Andreani, N. M. Wright, and N. Coltice. Pulsated global hydrogen and methane flux at mid-ocean ridges driven by Pangea breakup. *Geochemistry, Geophysics, Geosystems*, 21(4):e2019GC008869, 2020.
50. J. Fitton, A. Saunders, L. Larsen, B. Hardarson, and M. Norry. Volcanic rocks from the southeast Greenland Margin at 63 °N: composition, petrogenesis, and mantle sources. *Proceedings of the Ocean Drilling Program: Scientific Results*, 152:331–350, 1998.
51. J. G. Fitton and M. Godard. Origin and evolution of magmas on the Ontong Java Plateau. *Geological Society, London, Special Publications*, 229(1):151–178, 2004.
52. K. Norrish and J. T. Hutton. An accurate X-ray spectrographic method for the analysis of a wide range of geological samples. *Geochimica et Cosmochimica Acta*, 33(4):431–453, 1969.
53. R. C. Reynolds. Matrix corrections in trace element analysis by X-ray fluorescence: Estimation of the mass absorption coefficient by Compton scattering. *American Mineralogist*, 48(9-10):1133–1143, 1963.
54. K. Govindaraju. 1994 compilation of working values and sample description for 383 geostandards. *Geostandards Newsletter*, 18(S1):1–158, 1994.
55. K. P. Jochum, H. M. Seufert, and M. F. Thirlwall. High-sensitivity Nb analysis by spark-source mass spectrometry (SSMS) and calibration of XRF Nb and Zr. *Chemical Geology*, 81(1):1–16, 1990.
56. N. Imai, S. Terashima, S. Itoh, and A. Ando. 1994 compilation of analytical data for minor and trace elements in seventeen GSJ geochemical reference samples, “igneous rock series”. *Geostandards Newsletter*, 19(2):135–213, 1995.
57. B. J. Murton, R. N. Taylor, and M. F. Thirlwall. Plume–ridge interaction: a geochemical perspective from the Reykjanes Ridge. *Journal of Petrology*, 43(11):1987–2012, 2002.

58. T. Tanaka, S. Togashi, H. Kamioka, H. Amakawa, H. Kagami, T. Hamamoto, M. Yuhara, Y. Orihashi, S. Yoneda, H. Shimizu, T. Kunimaru, K. Takahashi, T. Yanagi, T. Nakano, H. Fujimaki, R. Shinjo, Y. Asahara, M. Tanimizu, and C. Dragusanu. JNd-1: a neodymium isotopic reference in consistency with LaJolla neodymium. *Chemical Geology*, 168(3):279–281, 2000.
59. S. B. Jacobsen and G. J. Wasserburg. Sm-Nd isotopic evolution of chondrites. *Earth and Planetary Science Letters*, 50(1):139–155, 1980.
60. L. E. Munro, F. J. Longstaffe, and C. D. White. Effects of heating on the carbon and oxygen-isotope compositions of structural carbonate in bioapatite from modern deer bone. *Palaeogeography, Palaeoclimatology, Palaeoecology*, 266(3):142–150, 2008.
61. R. M. Macintyre and P. J. Hamilton. Isotopic geochemistry of lavas at DSDP Holes 81-553 and 81-555. In *DSDP Initial Reports*, chapter 30, pages 775–781. 1984.
62. D. G. Roberts, J. Backman, A. C. Morton, J. W. Murray, and J. B. Keene. Evolution of volcanic rifted margins: Synthesis of Leg 81 results on the west margin of Rockall Plateau. In *DSDP Initial Reports*, chapter 39, pages 883–911. 1984.
63. W. A. Berggren, D. V. Kent, C. C. Swisher, and M. P. Aubrey. A revised Cenozoic geochronology and chronostratigraphy. *Society for Sedimentary Geology Special Publication*, 54:129–212, 1995.
64. J. Lund. A late Paleocene non-marine microflora from the interbasaltic coals of the Faeroe Islands, North Atlantic. *Bulletin of the Geological Society of Denmark*, 37:181–203, 1988.
65. J. G. Ogg. *Chapter 5 - Geomagnetic Polarity Time Scale*, chapter 5, pages 85–113. Elsevier, Boston, 2012.
66. M. M. Hirschmann, P. R. Renne, and A. R. McBirney. $^{40}\text{Ar}/^{39}\text{Ar}$ Ar dating of the Skaergaard intrusion. *Earth and Planetary Science Letters*, 146(3):645–658, 1997.
67. J. K. Jakobsen, C. Tegner, C. K. Brooks, A. J. R. Kent, C. E. Leshner, T. F. D. Nielsen, and M. Wiedenbeck. Parental magma of the Skaergaard intrusion: constraints from melt inclusions in primitive troctolite blocks and FG-1 dykes. *Contributions to Mineralogy and Petrology*, 159(1):61–79, 2009.
68. R. Waagstein. Structure, composition and age of the Faeroe basalt plateau. *Geological Society, London, Special Publications*, 39(1):225–238, 1988.
69. D. McKenzie and R. K. O’Nions. Partial melt distributions from inversion of rare earth element concentrations. *Journal of Petrology*, 32(5):1021–1091, 7/16/2020 1991.
70. M. B. Baker and E. M. Stolper. Determining the composition of high-pressure mantle melts using diamond aggregates. *Geochimica et Cosmochimica Acta*, 58(13):2811–2827, 1994.
71. M. J. Walter. Melting of garnet peridotite and the origin of komatiite and depleted lithosphere. *Journal of Petrology*, 39(1):29–60, 1998.
72. A. R. Hastie, J. G. Fitton, A. C. Kerr, I. McDonald, A. Schwindrofska, and K. Hoernle. The composition of mantle plumes and the deep earth. *Earth and Planetary Science Letters*, 444:13–25, 2016.
73. S. R. Hart and T. Dunn. Experimental cpx/melt partitioning of 24 trace elements. *Contributions to Mineralogy and Petrology*, 113(1):1–8, 1993.
74. E. H. Hauri, T. P. Wagner, and T. L. Grove. Experimental and natural partitioning of Th, U, Pb and other trace elements between garnet, clinopyroxene and basaltic melts. *Chemical Geology*, 117(1):149–166, 1994.
75. K. T. M. Johnson. Experimental determination of partition coefficients for rare earth and high-field-strength elements between clinopyroxene, garnet, and basaltic melt at high pressures. *Contributions to Mineralogy and Petrology*, 133(1):60–68, 1998.
76. P. McDade, J. D. Blundy, and B. J. Wood. Trace element partitioning on the Tinquillo Lherzolite solidus at 1.5GPa. *Physics of the Earth and Planetary Interiors*, 139(1):129–147, 2003.
77. V. J. M. Salters, J. E. Longhi, and M. Bizimis. Near mantle solidus trace element partitioning at pressures up to 3.4 GPa. *Geochemistry, Geophysics, Geosystems*, 3(7):1–23, 2002.
78. T. Skulski, W. Minarik, and E. B. Watson. High-pressure experimental trace-element partitioning between clinopyroxene and basaltic melts. *Chemical Geology*, 117(1):127–147, 1994.
79. J. Tuff and S. A. Gibson. Trace-element partitioning between garnet, clinopyroxene and Fe-rich picritic melts at 3 to 7 GPa. *Contributions to Mineralogy and Petrology*, 153(4):369–387, 2007.
80. A. J. Irving. A review of experimental studies of crystal/liquid trace element partitioning. *Geochimica et Cosmochimica Acta*, 42(6):743–770, 1978.
81. M. R. Perfit and J. H. Steele. *Mid-Ocean Ridge Geochemistry and Petrology*, pages 815–825. Academic Press, Oxford, 2001.
82. R. D. Müller and A. Dutkiewicz. Oceanic crustal carbon cycle drives 26-million-year atmospheric carbon dioxide periodicities. *Science Advances*, 4(2):eaq0500, 2018.
83. S. Y. O’Reilly and W. L. Griffin. Imaging global chemical and thermal heterogeneity in the subcontinental lithospheric mantle with garnets and xenoliths: Geophysical implications. *Tectonophysics*, 416(1):289–309, 2006.
84. S. Tappe, S. F. Foley, A. Stracke, R. L. Romer, B. A. Kjarsgaard, L. M. Heaman, and N. Joyce. Craton reactivation on the Labrador Sea margins: $^{40}\text{Ar}/^{39}\text{Ar}$ age and Sr–Nd–Hf–Pb isotope constraints from alkaline and carbonatite intrusives. *Earth and Planetary Science Letters*, 256(3):433–454, 2007.
85. S. Tappe, R. L. Romer, A. Stracke, A. Steinfeldt, K. A. Smart, K. Muehlenbachs, and T. H. Torsvik. Sources and mobility of carbonate melts beneath cratons, with implications for deep carbon cycling, metasomatism and rift initiation. *Earth and Planetary Science Letters*, 466:152–167, 2017.
86. T. M. Gernon, B. G. J. Upton, R. Ugra, C. Yücel, R. N. Taylor, and H. Elliott. Complex subvolcanic magma plumbing system of an alkali basaltic maar-diatreme volcano (Elie Ness, Fife, Scotland). *Lithos*, 264:70–85, 2016.
87. M. J. Le Bas, R. W. Le Maitre, A. Streckeisen, and B. Zanettin. A chemical classification of volcanic rocks based on the total alkali-silica diagram. *Journal of Petrology*, 27(3):745–750, 1986.
88. L.M. Chambers. *Age and duration of the British Tertiary Igneous Province: implications for the development of the ancestral Iceland plume*. PhD thesis, University of Edinburgh, 2000.
89. J. A. Brodie and J. G. Fitton. Data report: Composition of basaltic lavas from the seaward-dipping reflector sequence recovered during Deep Sea Drilling Project Leg 81 (Hatton Bank). In *Proceedings of the Ocean Drilling Program, Scientific Results*, volume 152, pages 431–435, 1998.
90. J. G. Fitton, J. J. Mahoney, P. J. Wallace, and A. D. Saunders. Origin and evolution of the Ontong Java Plateau: introduction. *Geological Society, London, Special Publications*, 229(1):1–8, 2004.
91. W. F. McDonough and S. s. Sun. The composition of the Earth. *Chemical Geology*, 120(3):223–253, 1995.

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Author contributions

T.G. conceived the idea, led the study, interpreted the data and prepared the manuscript and figures. T.H. performed the modelling, with input from T.G. R.N.M. assisted with tectonic and geodynamic interpretation, and G.F., J.L., R.B. and

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

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Figures

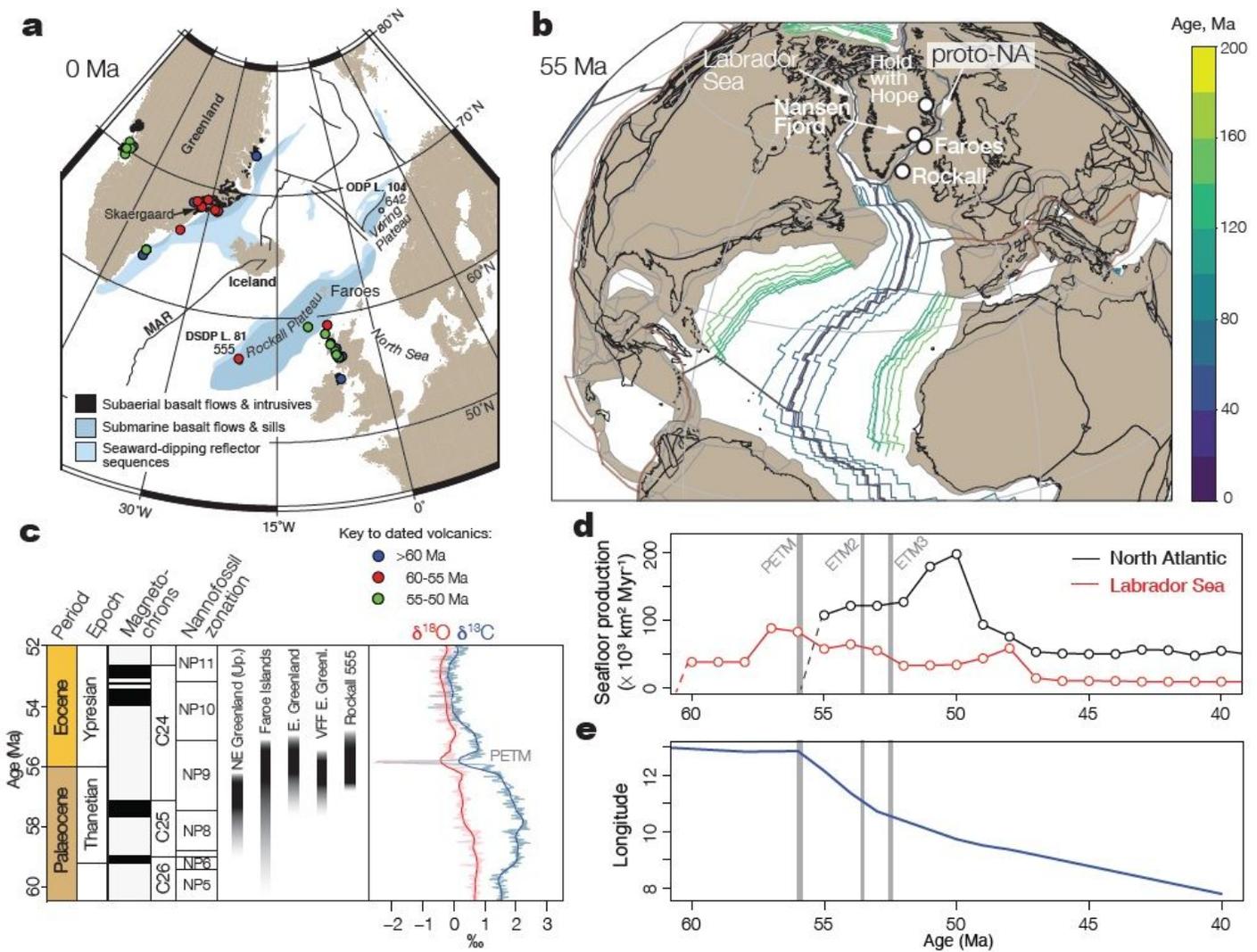


Figure 1

Early Cenozoic tectonic and magmatic evolution of the North Atlantic region 1 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 2. 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 1,3, magnetostratigraphy and nannofossil zonation 4,5,6, 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 8 indicating the onset of ocean crustal production in the North Atlantic and ridge push at 56 Ma.

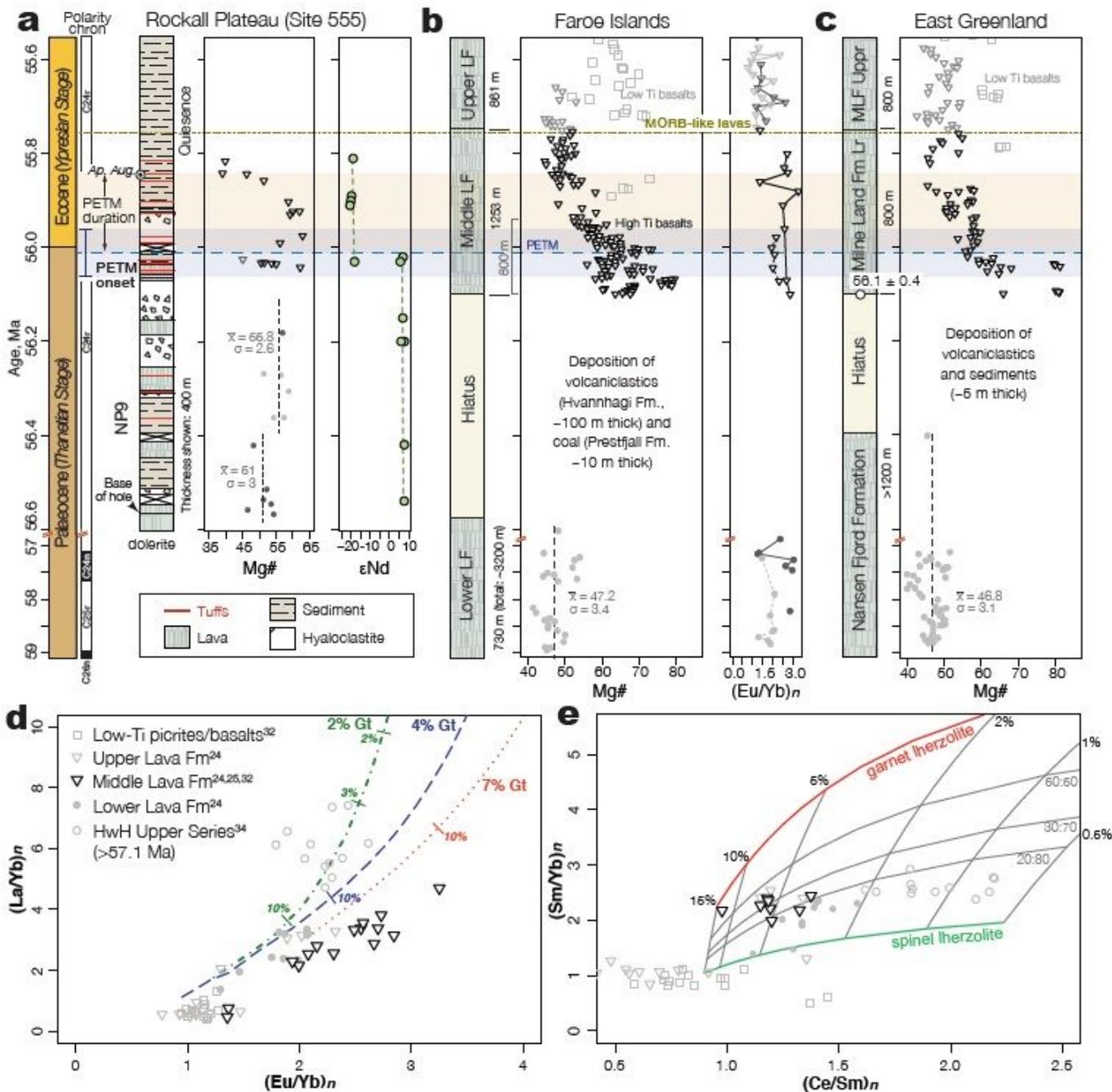


Figure 2

Palaeocene-Eocene volcanostratigraphy and geochemistry of the proto-North Atlantic ridge. a, Simplified log of the Rockall 'Phase 1' sequence 21 showing lithologies, Mg# (i.e., 100 x molecular MgO/(MgO + FeO), where FeO is assumed to be 0.9FeOT), and ϵ_{Nd} (Supplementary Fig. 1). b, Simplified log of the Faroes Basalt Formations 22, with Mg# and $(Eu/Yb)_n$ (chondrite-normalized 23); Mg# data are from 22 and $(Eu/Yb)_n$ are from 24,25; note the sharp transition to high Mg# (and enriched REE contents) at ca. 56.1 Ma, which is also observed in c, east Greenland (Milne Land Formation) 22. d, $(La/Yb)_n$ vs $(Eu/Yb)_n$ of the Faroes and Hold with Hope (HwH) lavas (chondrite-normalized 23) and modeled non-modal batch melting of a hercynitic mantle source, adopted from 26, showing different degrees of

melting of a garnet lherzolite (green, blue and red curves). e, $(\text{Sm}/\text{Yb})_n$ vs $(\text{Ce}/\text{Sm})_n$ and an REE melting model (Methods), showing percentage melt along the top and the relative proportions of garnet- and spinel-lherzolites from 100% gt-lherzolite (red curve) to 100% sp-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.

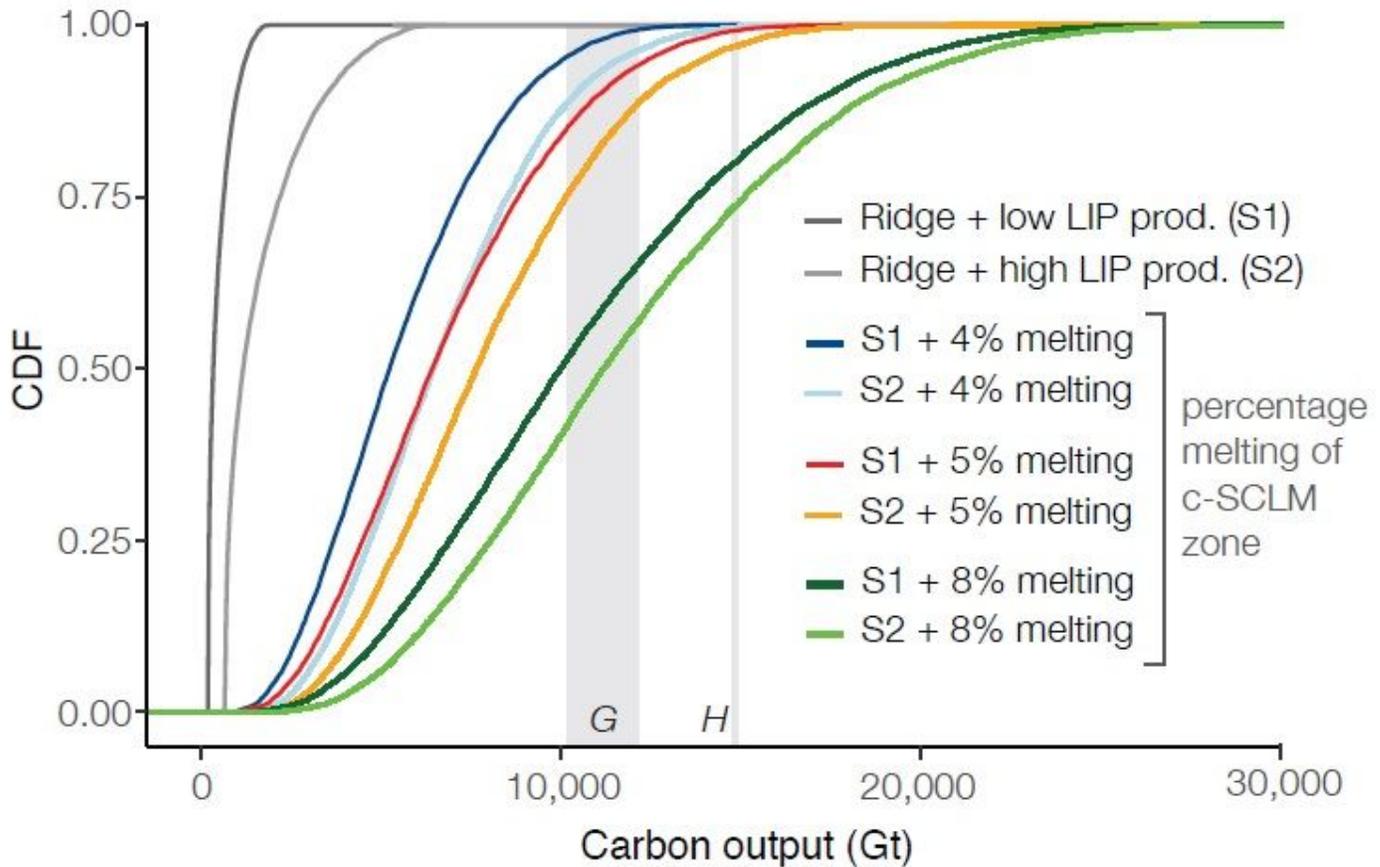


Figure 3

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 ($0.6 \text{ km}^3 \text{ yr}^{-1}$) and high ($2.4 \text{ km}^3 \text{ yr}^{-1}$) LIP eruption rate scenarios 9, respectively (Supplementary Fig. 3) (see Methods). The coloured lines show the effects of adding 4% to 8% carbonated (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. 14 (labelled G; 10,200–12,200 Gt C), and Haynes & Hönisch 16 (labelled H; 14,900 Gt C).

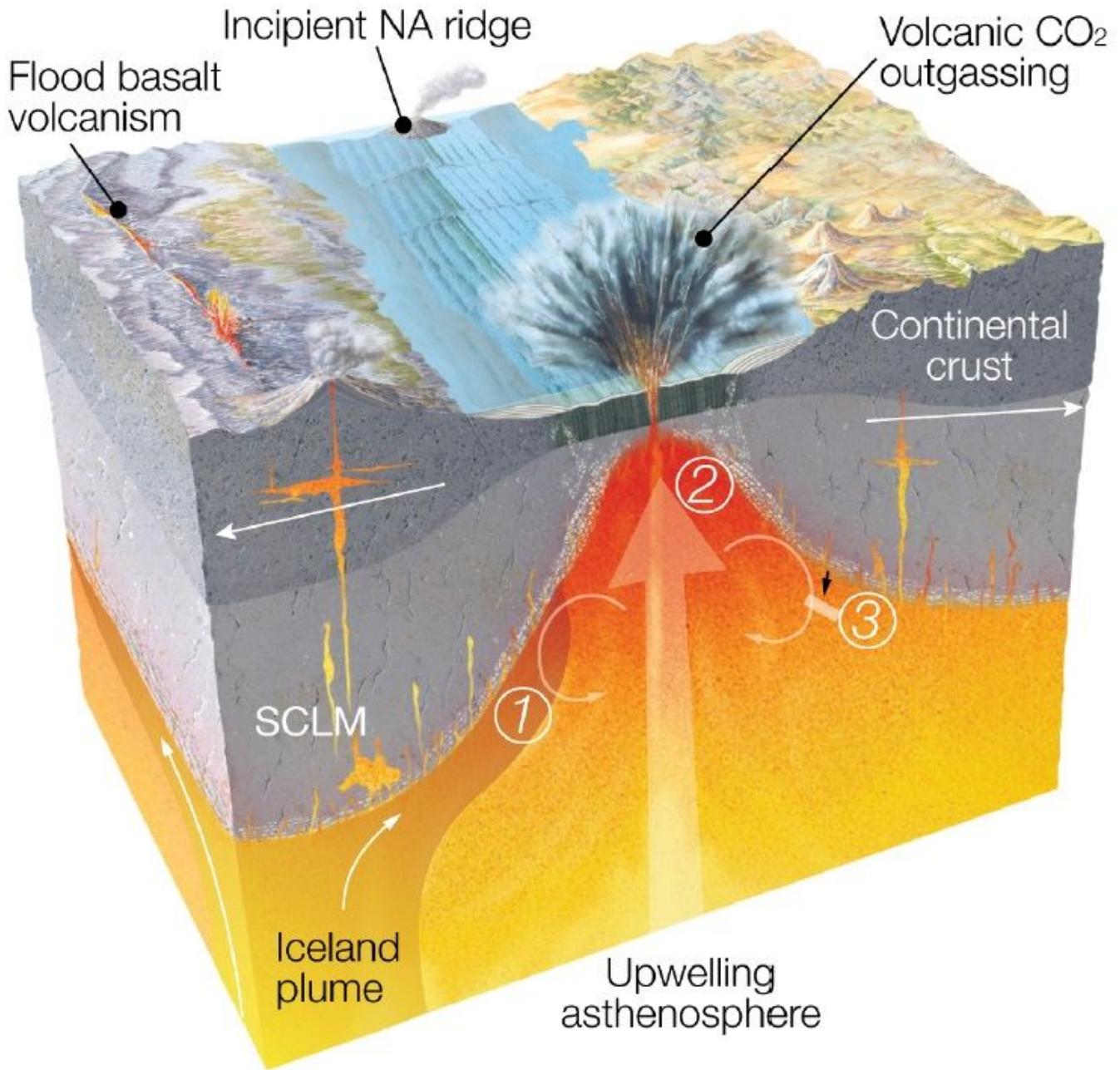


Figure 4

Volcanic carbon release in the North Atlantic during the PETM. Model stages: 1, progressive thermo-mechanical weakening of the sub-continental lithospheric mantle (SCLM) by the Iceland plume; 2, disruption of the ruptured metasomatized SCLM zone by incipient asthenospheric upwelling; 3, delamination of carbonated fragments of the deep SCLM during lithospheric stretching, which generate the enduring enriched components ('EM1') still present in the Icelandic mantle today 41.

Supplementary Files

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