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High temperatures in the terrestrial mid-latitudes during the early Paleogene

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The early Paleogene (56–48 Myr) provides valuable information about the Earth’s climate system in an equilibrium high $pCO_2$ world. High ocean temperatures have been reconstructed for this greenhouse period, but land temperature estimates have been cooler than expected. This mismatch between marine and terrestrial temperatures has been difficult to reconcile. Here we present terrestrial temperature estimates from a newly-calibrated $brGDGT$-based paleothermometer in ancient lignites (fossilized peat). Our results suggest early Paleogene mid-latitude (45-60 degrees paleolatitude) mean annual air temperatures of 23 – 29 °C (with an uncertainty of ± 4.7 °C), 5-10 °C higher than
The identification of archaeal biomarkers in these same lignites, heretofore observed only in thermophiles and hyperthermophilic settings, support these high temperature estimates. These mid-latitude terrestrial temperature estimates are consistent with reconstructed ocean temperatures and indicate that the terrestrial realm was much warmer during the early Paleogene than previously thought.

The early Paleogene is characterized by an extended period of high atmospheric carbon dioxide ($p$CO$_2$) levels$^{1,2}$. Quantification of temperatures during greenhouse climates is needed 1) because they can be used to evaluate climate model simulations at elevated $p$CO$_2^3$, 2) because temperature governs diverse components of climate dynamics (e.g. circulation patterns)$^4$ and feedback mechanisms within the Earth system (e.g. weathering)$^5$, and 3) because they influence biogeochemical processes (e.g. flux of methane from wetlands into the atmosphere)$^6$. Although potentially not continuously as hot as the relatively short-lived extreme greenhouse events such as the Paleocene Eocene Thermal Maximum (PETM), the extended greenhouse climate state of the early Paleogene is the focus here.

Over the last decade, considerable effort has been made to reconstruct the early Paleogene greenhouse climate with a variety of calcite-based, leaf physiognomic, and organic geochemical proxies. For example, sea surface temperatures (SSTs) have been reconstructed with the organic TEX$_{86}$ proxy$^7$, based on the distribution of isoprenoidal glycerol dialkyl glycerol tetraethers (isoGDGTs), lipids synthesized by Archaea, in marine sediments. Those records indicate SSTs significantly higher than modern, with SSTs from the SW Pacific at 60 °S paleolatitude above 30 °C$^8$. Similarly, calcite-based SST proxies such as the Mg/Ca
ratio or clumped isotopic composition of foraminiferal calcite indicate significantly elevated SSTs at all latitudes during the early Paleogene\textsuperscript{9}. Together the SST records indicate ocean temperatures significantly higher than modern with most estimates from between 60 °S and 50 °N above 22 °C (Fig. 1).

Some climate models, such as CCSM3, can (partly) reproduce these elevated temperatures using 16x modern-day $p$CO\textsubscript{2} levels\textsuperscript{10}, but such $p$CO\textsubscript{2} values are higher than proxy estimates for the early Paleogene\textsuperscript{1,2}. Other models, such as HadCM3L and ECHAM, generally cannot reproduce the warming at $p$CO\textsubscript{2} levels consistent with the marine proxy estimates\textsuperscript{3}. The apparent high SST reconstructions have therefore been attributed to proxy complications, such as a subsurface origin of the lipids used in the TEX\textsubscript{86} proxy\textsuperscript{11}, variations in early Paleogene seawater chemistry compared to modern that especially influences the calcite-based paleothermometers\textsuperscript{12}, and/or a seasonal (summer) bias in the marine proxies\textsuperscript{13,14}. However, recent model simulations have identified potential biases against polar warming in general circulation models that are tuned to modern conditions\textsuperscript{15,16}, associated with the representation of cloud properties, which may partly explain the model-data discrepancy at mid/high latitudes.

The available terrestrial temperature proxies, based mainly on leaf physiognomic temperature estimates and the MBT(‘)/CBT organic mineral soil temperature proxy, based on the distribution of bacterial branched (br)GDGTs, suggest that early Paleogene terrestrial temperatures in general were also higher than modern\textsuperscript{10,17,18}, but to a lesser degree than indicated by SST reconstructions. There are very few terrestrial temperature data from the (sub)tropics, but almost all estimates indicate mean air temperatures below 22 °C during the early Paleogene at all latitudes (Fig. 1a). These terrestrial temperature estimates are more consistent with climate model simulations\textsuperscript{10}, but considerably lower than SST estimates, presenting a
conundrum. In order to understand this greenhouse climate state, independent early
Paleogene temperature estimates are needed to test whether temperatures on land
were as high as suggested by marine proxies or as low as indicated by most climate
model simulations and many existing terrestrial proxies. For this purpose, here we use
the distribution of archaeal and bacterial lipids obtained from lignites (ancient peat) to
reconstruct temperatures in early Paleogene mid-latitude peatlands.

**GDGTs in modern peat**

A decade of research has demonstrated that in mineral soils the degree of methylation
of bacterial brGDGTs, calculated using the degree of methylation of (5-methyl)
branched tetraethers (MBT’(5me)) index, is correlated with mean annual air
temperature\(^{19,20}\). Although temperature is highly correlated with the degree of
methylation, the influence of other factors (e.g. nutrient content) is currently poorly
constrained among others due to the lack of culture studies. The MBT(’)/CBT mineral
soil temperature proxy has been applied to marine sediments to reconstruct early
Paleogene terrestrial temperatures\(^{21}\). However, the application of the mineral soil
calibration to other climatic archives (e.g. peat and by extension lignite) can be
problematic as these represent different environmental conditions than those
predominantly comprising the modern mineral soil calibration dataset. To address
this, a global peat-specific brGDGT temperature calibration that is based on MBT’\(^{5me}\)
in a diverse range (n = 470) of modern peats (MAAT\(_\text{peat}\)) was recently developed\(^{22}\).
This proxy has a calibration error of ± 4.7 °C and reaches saturation at 29.1 °C. It is
important to note that in peat settings, MAAT\(_\text{peat}\) is unlikely to record seasonal
temperatures, because in peats the brGDGT pool is dominated by bacterial production
at depth below the water table where seasonal temperature fluctuations are muted and
converge to mean annual air temperatures\textsuperscript{22}. As with all paleothermometers, we assume that the strong correlation between the degree of methylation of \textit{br}GDGTs and temperature observed in the modern calibration dataset\textsuperscript{22} was the same during the early Paleogene.

In addition to Bacteria (that can produce \textit{br}GDGTs), Archaea also live in peat, and their membrane lipids (\textit{iso}GDGTs) are similarly preserved in ancient peat and lignite. Here we examined the \textit{iso}GDGT distribution in our previously compiled global database of modern peat\textsuperscript{22}. For the first time, we report \textit{iso}GDGT-5 (as well as \textit{iso}GDGT-6 and -7) in modern mesophilic peats. So far \textit{iso}GDGTs with more than 4 cyclopentane rings have only been found in hot springs and cultures of (acido) hyperthermophiles\textsuperscript{23}. It has been suggested that the ability to synthesize \textit{iso}GDGT-5 to 8 is a unique adaption of extremophiles and does not occur in mesophilic settings\textsuperscript{23}. However, our work demonstrates that this biomarker is also present in ombrotrophic (acidic) tropical peats located between 20 °S and 20 °N latitude today. \textit{iso}GDGT-5 is only present in significant amounts (>1% of total \textit{iso}GDGT distribution with 1-5 cyclopentane rings) in tropical and ombrotrophic peats with a pH < 5.1 and MAAT > 19.5 °C (Fig. 2). It is absent in all peatlands with a pH > 5.1 or MAAT < 12 °C and present only in trace proportions (<1% of \textit{iso}GDGTs) in acidic peatlands with MAAT between 12°C and 19.5 °C. The highest proportion of \textit{iso}GDGT-5 in the modern database is 9% in an ombrotrophic Indonesian peat (modern MAAT 26.5 °C, pH 3). The distribution of these compounds in modern peats provides strong evidence that their occurrence (when greater than 1% of total \textit{iso}GDGTs with 1-5 cyclopentane rings) is diagnostic for peatlands with high temperatures (>19.5 °C) and low pH (<5.1). We suggest that the proportional abundance of \textit{iso}GDGT-5 (as well as
isoGDGT-6) likely increases with temperature when pH is held constant, although we have insufficient data to convert that into an empirical calibration.

Terrestrial temperatures from early Paleogene lignites

Here we use the relative abundance of the archaeal lipid isoGDGT-5 and degree of methylation of bacterial brGDGTs (MBT’5me) obtained from lignites and newly calibrated proxies using modern peats to reconstruct temperature in early Paleogene peatlands (see SI for details on age models). Ancient peats can be preserved in the form of immature lignites, also known as brown coals, after compaction under low burial pressure and temperatures (< 100 °C). We use lignites from Germany (Schöningen), UK (Cobham), New Zealand (Otaio), and several basins in western India (Barsingsar seam, Bikaner Basin; Kasnau Matasukh seam, Nagaur Basin; Matanomadh and Panandhro seams, Kachchh Basin; and Khadsaliya Clays, Saurashtra Basin). These lignites derive from peatlands influenced by marine incursions and hence reflect local temperature very near sea level.

As far as is possible, given the difficulties of precise dating in purely continental strata, samples deposited within hyperthermals have been avoided (see SI), such that these samples are expected to represent minimum temperature estimates of early Paleogene warmth. However, dating terrestrial sections is difficult and the precise age of all samples, but especially the Indian lignites, remains difficult to confirm, and it remains possible that more extreme climate states have been included.

All latitudes reported here are best estimates for paleolatitudes. Early Paleogene lignites reveal $MAAT_{\text{peat}}$ in Schöningen (~46 °N) varied between 22.5 and 28 °C ± 4.7 °C (n = 39, 0.87 < MBT’5me < 0.98) and in Cobham (~48 °N) between 23.5 and 26 °C ± 4.7 °C (n = 7, 0.90 < MBT’5me < 0.94) during the latest
Paleocene/earliest Eocene (Fig. 1a). At Otaio (~57 °S) MAAT$_{\text{peat}}$ in earliest Eocene lignites (i.e. directly following the PETM) varied between 27 and 29 °C ± 4.7 °C (n = 7, 0.91 < MBT$'_{5\text{me}}$ < 1), close to the upper limit of MAAT$_{\text{peat}}$. These mid-latitude temperature reconstructions for the early Paleogene (22 to 29 °C), are markedly warmer than present (2 to 15 °C), even when taking the calibration error of 4.7 °C into account (Fig. 1a). The Indian lignites (~0-5 °N) consist of a variety of lignites of early Paleogene age and are not as well-dated. MAAT$_{\text{peat}}$ in these lignite samples varied between 28 and 29 °C ± 4.7 °C (n = 9, 0.98 < MBT$'_{5\text{me}}$ < 1) and were close to the maximum value of the calibration, such that they might be minimum estimates.

All lignites are also associated with the occurrence of archaeal isoGDGTs with more than 4 cyclopentane moieties (Fig. 1), predominantly isoGDGT-5 but also isoGDGT-6 in some samples (see SI). It is unlikely that the presence of these unusual biomarkers is evidence for hyperthermophilic (e.g hot springs) conditions in all of these ancient peatlands. Deep biosphere production of GDGTs during burial at depth is unlikely to be a significant influence on our temperature records as lignite deposits are characterized by low amounts of intact polar lipid GDGTs$^2_{24}$, arguing against an active GDGT-producing microbial community in such settings.

In the early Paleogene lignites, the abundance of isoGDGT-5 is the highest, on average, in India in the palaeotropics; lower values occur between 45-60° paleolatitude (Fig. 1). The high proportions of isoGDGT-5 in early Paleogene lignites suggests that acidic peatlands with temperatures higher than 19.5 °C existed at paleolatitudes of 46-48 °N (Cobham and Schöningen) as well as 57 °S (Otaio).

Moreover, the proportion of isoGDGT-5 in Indian lignites is higher than those found in any modern peat. We suggest that the higher proportions in Indian lignites compared to the other Paleogene sites is not the result of a much lower pH, as there is
independent evidence that at least some of the latter were formed in ombrotrophic *Sphagnum* peats. Instead, it is likely that higher proportions of *iso*GDGT-5 in the Indian lignites indicates MAATs higher than presently found in the low-latitudes.

**Comparison with existing temperature reconstructions**

Collectively, the entire GDGT biomarker distribution yields two independent temperature estimates that originate from two different domains of life, suggesting that terrestrial peatland temperatures between 45-60° paleolatitude were significantly higher than modern during the early Paleogene period of elevated $p$CO$_2$, with values similar to those found at present only in tropical peatlands. Although the bacterial-based MAAT$_{\text{peat}}$ calibration is near its limit in the Indian lignites, high abundances of *iso*GDGT-5 provide evidence that tropical temperatures were also elevated relative to those of today, consistent with SST reconstructions.

The majority of existing multi-proxy terrestrial temperature data (e.g. foliar physiognomy, MBT'/CBT, etc.) suggests that continental temperatures in the mid-latitude Northern Hemisphere (40-60 °N) were below 22 °C during the early Paleogene (Fig. 1a). Some leaf physiognomic estimates from the NW America, based mainly on the Kowalski and Dilcher calibration and especially the CLAMP data, suggest temperatures within error to those found at present at these latitudes. Similarly, all paleosol-based temperature estimates, obtained using a range of geochemical methods, are close to or below modern-day temperatures at similar latitudes. This is difficult to reconcile given the multi-proxy evidence for significantly elevated $p$CO$_2$ levels during the early Paleogene. Such low temperatures in the mid-latitude Northern Hemisphere are also difficult to reconcile with terrestrial temperatures from the high-latitude Northern Hemisphere (> 60 °N).
that range between 14 and 20 °C\textsuperscript{28,29} and widespread evidence of subtropical flora\textsuperscript{29,30} and fauna\textsuperscript{31,32} in the (high) Arctic.

The MAAT\textsubscript{peat} estimates from the UK and Germany with average values ca. 25 and 27 ± 4.7 °C, respectively, indicate that mid-latitude terrestrial temperatures are at the high end of (or higher than) leaf physiognomic proxy estimates for comparable latitudes (Fig. 1a). However, these new data are consistent with summer temperature estimates in excess of 40 °C based on clumped isotopes of paleosol carbonates from the Bighorn Basin (~45 °N paleolatitude)\textsuperscript{33} and \(\delta^{18}O\)-based terrestrial temperatures from mammalian tooth enamel and fish (gar) scales from the southern USA (~30 to 40 °N) with estimates between 28 and 32 ± 5.5 °C\textsuperscript{34}. Similarly, the data from Schöningen are consistent with early Eocene temperatures of 22.5 ± 2.5 °C based on leaf margin analysis from the nearby Messel oil shale\textsuperscript{35}. These new terrestrial temperature estimates are also consistent with TEX\textsubscript{86}, Mg/Ca, and clumped isotope-based SST estimates between 19 and 32 °C from the mid-latitude Northern Hemisphere\textsuperscript{9,36} (Fig. 1b).

The published early Paleogene terrestrial temperature estimates from between 45 and 65 °S indicate values between ~10 and 20 °C, in general higher than modern values at these latitudes (Fig. 1a). MAAT\textsubscript{peat} estimates from New Zealand are ~ 5-10 °C higher than existing terrestrial temperature estimates for the region, with an average value of 28 ± 4.7 °C. However, some of the existing terrestrial temperature estimates were obtained from marine sediment cores in the Southern Ocean at ~60 °S, but record conditions further south at Wilkes Land (Antarctica) at ~70 °S. They indicate the presence of near-tropical forests on Antarctica\textsuperscript{37} and, together with plant microfossil evidence from the Tawanui section in N. Zealand that indicates the presence of thermophilic taxa directly before and after the PETM\textsuperscript{38}, they are
consistent with high MAAT$_{\text{peat}}$ values and presence of $iso$GDGT-5 in the Otaio lignite. Furthermore, MAAT$_{\text{peat}}$ is consistent with multi-proxy SST estimates from the mid/high latitude Southern Hemisphere that indicate values between 28 and 35 °C$^{8,9}$ (Fig. 1b).

It is likely that the MAAT$_{\text{peat}}$ estimates from India of 28-29 ± 4.7 °C represent minimum values, as also indicated by the higher than modern abundance of $iso$GDGT-5. This prevents a direct comparison with published low-latitude SST estimates. Even so, our estimates are slightly higher than terrestrial temperatures currently suggested for the early Paleogene of the Indian subcontinent$^{39}$, but within error of clumped isotope-based SSTs from the coast of India with values of 30-35 ± 2.5 °C$^{9}$.

The offset between some of the existing and MAAT$_{\text{peat}}$ terrestrial temperatures could partly be explained by a potential cold bias in temperatures based on leaf physiognomic and paleosol proxies$^{10,27}$, as well as uncertainty in paleo-elevation of several of the archives, especially those from N. America. We also note that MAAT$_{\text{peat}}$ estimates are higher than most previously published soil MBT'/CBT-based terrestrial temperature estimates from (proximal) marine sediments (Fig. 1a).

Although also based on the distribution of $br$GDGTs, MBT'/CBT-based temperatures from marine sediments could be biased by production in the water column or sediments$^{40}$. Marine sediments also represent an integrated temperature across a large catchment area, potentially including a contribution from high altitudes. In addition, recent analytical advances urge for caution in interpreting MBT'/CBT data as the original measurements could be biased by co-eluting compounds$^{19}$. As such, some of the original MBT'/CBT data might not reflect terrestrial temperatures at sea level, explaining the offset with our data.
These lignite-based data therefore reinvigorate the debate about early Paleogene temperatures: we find new evidence for high temperatures on land that are consistent with SST reconstructions, resolving the prior conundrum, but retaining the discrepancies between data and climate model simulations.

**Comparison with climate model simulations**

There are a number of climate models that have been used to simulate the early Paleogene climate, including CCSM3\(^{10,15,41}\), HadCM3L\(^{42}\), ECHAM5\(^{43}\), FAMOUS\(^{16}\), and GISS\(^{44}\). Although these climate models originally struggled to simulate warm climates like that of the early Paleogene, especially when using \(p\text{CO}_2\) estimates consistent with proxy-estimates\(^3\), more recently there has been progress. The latest set of climate model simulations for the early Paleogene (using CCSM3\(^{15}\) and FAMOUS\(^{16}\)) provide a better fit with proxy estimates of SSTs using \(p\text{CO}_2\) estimates that are consistent with proxy data after changing specific model parameters such as cloud properties, although they still struggle to reach the extent of warming indicated by SST proxies in the SW Pacific. Crucially, for the mid-latitude Northern Hemisphere (45-50 °N) the latest set of climate models fit the MAAT\(_\text{peat}\) temperature data, but are 5-10 °C warmer than most of the published mid-latitude temperature data (see Fig. 3b).

However, for the mid-latitude Southern Hemisphere (55-60 °S), the magnitude of warming simulated by all climate models is still less than indicated by MAAT\(_\text{peat}\) (Fig. 3a) and published SST estimates\(^8,45\). This could suggest that climate models are still missing crucial processes. However, it is important to highlight that virtually all mid/high-latitude Southern Hemisphere SST and terrestrial data (including the new MAAT\(_\text{peat}\) data from Otaio) come from the SW Pacific and Pacific sector of the
Southern Ocean. As such, the high temperatures so far found in the mid/high latitude Southern Hemisphere might reflect local conditions and not be fully representative of zonal averages\(^4\). Future terrestrial temperature estimates using early Paleogene lignites from for example S. America might be able to shed new light on whether these high temperatures were present throughout the mid/high latitude Southern Hemisphere.

These novel terrestrial temperature estimates have important climatic and biogeochemical implications. For example, studies across microbial to ecosystem scales have demonstrated that methanogenesis rates in peatlands and emission of methane to the atmosphere increase significantly with increasing temperature\(^6,47\). Combined with evidence that indicates that high \(p\mathrm{CO}_2\) would have stimulated primary productivity\(^48\), our temperature estimates further suggest that the methane flux for a given areal extent of peatland between 45-60 degree paleolatitude could have been much greater during the early Paleogene than at present. As methane is a potent greenhouse gas, our results support previous modeling work\(^48,49\) indicating the presence of an additional positive feedback mechanism associated with extensive warm mid-latitude peats in a high-CO\(_2\) world that could amplify warming to a greater degree than that estimated using existing or GCM-derived temperature estimates.

References


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Wilde and Walter Riegel (Schöningen) and Jerry Hooker (Cobham) for their extensive personal support during sample collection. Schöningen samples were collected with logistical and financial support that includes NERC grants numbers NE/J008656/1 to MEC and NE/J008591/1 to RDP and we thank ‘Helmstedter Revier of MIBRAG’ and previous owners for access to the mine which has now closed. Alfred McAlpine, AMEC, and Channel Tunnel Rail Link are thanked for providing access to the Cobham site. We thank A. Farnsworth for calculating the paleolatitudes for all locations.

Author contributions

BDAN, MEC, and RDP designed the project. BDAN analyzed all samples in the modern peat database for isoGDGTs and wrote the manuscript with contributions from all authors. MR analyzed the Indian and Otaio lignite samples for GDGTs, while GNI analyzed the Cobham and Schöningen lignite samples for GDGTs. BDAN, MEC and EMK developed the database of early Paleogene terrestrial palaeoclimate proxies. MEC (Cobham and Schöningen samples), EMK (Otaio samples) and PKS (Indian samples) provided age models and stratigraphic context of lignites. OL provided the modern tropical peat samples from Peru.

The authors declare no competing financial interests.

**Figure Captions**

**Fig. 1: early Paleogene temperature**

a) MAAT\textsubscript{peat} (stars) and abundance of $iso$GDGT-5 in early Paleogene lignites (bar chart) together with published temperatures using leaf physiognomy (green squares), MBT’/CBT proxy (dark circles), paleosol proxies (purple diamonds), and mammalian $\delta^{18}$O (violet crosses). b) MAAT\textsubscript{peat} and abundance of $iso$GDGT-5 with published TEX\textsubscript{86}/BAYSPAR-based (blue circles) and calcite–based SSTs (triangles) for the early Paleogene. Error bars on temperature data reflect combined spread in data (1$\sigma$) and calibration uncertainty (SI), while those for $iso$GDGT-5 reflect 1$\sigma$ from the average. All data and references are in the SI. Small grey circles and squares represents modern-day terrestrial and marine temperatures, respectively.

**Fig. 2: $iso$GDGT in modern peats**

Maximum relative abundance of $iso$GDGT-5 in modern peats plotted against in situ peat pH\textsuperscript{22} and mean annual air temperature\textsuperscript{22}. Vertical bars reflect range in pH reported for each peat. Shaded area indicates tropical ombrotrophic peats characterized by an $iso$GDGT-5 abundance > 1%.

**Fig. 3: data-model comparison for the early Paleogene**
Temperature anomaly between the early Paleogene and present at the paleolatitude of each location for all terrestrial temperature data from a) between 55 and 60 °S and b) from between 45 and 50 °N. Error bars reflect combined spread in data (1σ) and calibration uncertainty (see SI for details). Also shown is the zonal mean anomaly (early Paleogene minus pre-industrial) simulated by a range of climate models; 2xCO2 ECHAM543, 2xCO2 FAMOUS16, 4xCO2 GISS44, 5xCO2 CCSM3_K15, 6xCO2 HadCM3L42, 16xCO2 CCSM3_W41 and 16xCO2 CCSM3_H10 (see SI).
Methods

The biomarkers from the lignites from Schöningen were previously extracted\textsuperscript{18}. For this purpose approximately 0.5-10 g of sediment were extracted via Soxhlet apparatus for 24 hours using dichloromethane:methanol (DCM:MeOH; 2:1 v/v) to yield a total lipid extract (TLE). The TLE was initially separated over silica into neutral and fatty acid fractions using chloroform-saturated ammonia and chloroform:acetic acid (100:1 v/v), respectively. The neutral fraction was subsequently fractionated over alumina into apolar and polar (containing the GDGTs) fractions using Hexane:DCM (9:1 v/v) and DCM:MeOH (1:2 v/v), respectively. The biomarkers from the Cobham lignite were previously extracted\textsuperscript{50}. For this purpose samples were extracted by sonication with a sequence of increasingly polar solvents (four times with dichloromethane (DCM), four times with DCM/methanol (1:1 v/v) and three times with methanol). The total lipid extracts were separated into three fractions using a column packed with (activated) alumina by elution with hexane (apolar fraction), hexane/DCM (9:1 v/v; 3 ml) and DCM/methanol (1:2 v/v; 3 ml; polar fraction). Lignites from New Zealand were extracted for 24h in Soxhlet using DCM:MeOH, (2:1 v/v) and separated over alumina into apolar (hexane:DCM, 9:1 v/v) and polar (DCM:MeOH, 1:2 v/v) fractions. TLEs from Indian lignites were obtained via microwave extraction (Milestone Inc., CT, USA) using DCM:MeOH (9:1 v/v) for 10 minutes at 70°C. Aliquots of TLE were separated into hydrocarbon (hexane), aromatic (hexane:DCM, 1:1 v/v), and polar fractions (DCM:MeOH 3:1 v/v) over silica.

For all samples the polar fraction was dissolved in hexane/\textit{iso}-propanol (99:1, v/v) and passed through 0.45µm PTFE filters. Fractions were analyzed by high performance liquid chromatography/atmospheric pressure chemical ionisation – mass spectrometry (HPLC/APCI-MS). Instrument methods followed Hopmans et al.\textsuperscript{51}. 
Analyses were performed in selective ion monitoring (SIM) mode to increase sensitivity and reproducibility, and M+H⁺ (protonated molecular ion) GDGT peaks were integrated.

Mean annual air temperatures for the lignites were obtained using the degree of methylation of brGDGTs as reflected in the MBT'₅me index¹⁹ and MAATₚᵉᵃᵗ calibration²² (see SI for additional information).

\[
MBT'_{5Me} = \frac{(brGDGT - Ia + brGDGT - Ib + brGDGT - Ic)}{(brGDGT - Ia + brGDGT - Ib + brGDGT - Ic + brGDGT - IIa + brGDGT - IIb + brGDGT - IIc)}
\]

\[
MAATₚᵉᵃᵗ (°C) = 52.18 \times MBT'₅me - 23.05
\]

isoGDGT-5 was identified based on relative retention times, as well as co-injection with an acid hydrolyzed >95% pure culture of the thermoacidophile *Thermoplasma acidophilum* (Matreya) (see SI). The relative abundance of isoGDGT-5 was calculated using the respective peak areas of isoGDGTs with one, two, three, and five cyclopentane rings;

\[
(1) \text{ isoGDGT} - 5 \, (\%)
\]

\[
= 100 \times \frac{(\text{isoGDGT} - 5)}{(\text{isoGDGT} - 1) + (\text{isoGDGT} - 2) + (\text{isoGDGT} - 3) + (\text{isoGDGT} - 5)}
\]

IsoGDGT-4 was excluded from this ratio due to the co-elution with the [M+H]⁺ + 2 ion of crenarchaeol that also gives m/z 1294⁵².

**Data availability**

The authors declare that all data supporting the findings of this study are available within the article (and its supplementary information files and the Pangaea database).

All data are available in the supplements and in addition all modern peat GDGT data are available on the Pangaea database [https://doi.org/10.1594/PANGAEA.883765](https://doi.org/10.1594/PANGAEA.883765) as.
well. The compilation of all previously published terrestrial and marine temperature
data from the early Paleogene together with the original references is also available in
the supplements.

References

50 Pancost, R. D. et al. Increased terrestrial methane cycling at the Palaeocene-
51 Hopmans, E. C., Schouten, S. & Sinninghe Damsté, J. S. The effect of
improved chromatography on GDGT-based palaeoproxies. Org. Geochem. 93,
1-6 (2016).
52 Weijers, J. W. H., Schouten, S., van der Linden, M., van Geel, B. & Sinninghe
Damsté, J. S. Water table related variations in the abundance of intact archaeal
membrane lipids in a Swedish peat bog. FEMS Microbiol. Lett. 239, 51-56
(2004).
Terrestrial temperature anomaly (°C)

Latitude

-60 -59 -58 -57 -56 -55
0 5 10 15 20 25 30

a) b)

MAAT_{pale}
K&D calibr.
CLAMP
Other leaf physio.
MBT/CBT
Mammal δ^{18}O

16xCO_{2}; CCSM3_H
16xCO_{2}; CCSM3_W
6xCO_{2}; HadCM3L
5xCO_{2}; CCSM3_K
4xCO_{2}; GISS
2xCO_{2}; FAMOUS
2xCO_{2}; ECHAM5
Supplementary information to *High temperatures in the terrestrial mid-latitudes during the early Paleogene* by Naafs et al.

1. Description of lignites and age models

1.1 Schöningen lignite (Germany)

36 samples were collected from Seam 1 in the Schöningen Südfeld mine, northern Germany (51.13°N, 11.00°E) (Fig. S1). Samples no. 33 to 1 were obtained from the high-resolution sampling series of 2008 and 2012\(^{S1,2}\). Samples XXIII 4a to XXXIII 7b were obtained from subsequent low-resolution sampling\(^{S2}\). The lignites in this mine were deposited as peat in a low-lying coastal setting\(^{S3}\) with a paleolatitude of around 46 °N. The seam from which the samples are derived is ~2.7 m thick and is overlain and underlain by brackish to shallow marine, clastic sedimentary deposits\(^{S3,4}\).

The dinocyst zone D 5nb was recognized above the Main Seam in the nearby Emmerstedt area by Ahrendt et al.\(^{S5}\). If the Main seam is coeval at both sites this would indicate that Seam 1 at Schöningen is earliest Eocene. However, within marine Interbed 2, directly above Seam 1, there is a dramatic increase in the abundance of the dinocyst *Apectodinium*\(^{S3}\) which may represent the onset of the Paleocene-Eocene Thermal Maximum (PETM) as it does at other sites\(^{S6,7}\). However, none of the studied samples yielded a negative δ\(^{13}C\) excursion that would suggest it was deposited during the main body of the PETM\(^{S4}\). Therefore, Seam 1 is considered to be either very latest Paleocene or very earliest Eocene in age. During the subsequent early Eocene (Seam 3 upwards), there is a long-term temperature maximum recorded from both the lignites and nearshore marine interbeds, consistent with changes in the palynological assemblage\(^{S2,3}\). As this interval may include the Early Eocene Climatic Optimum (EECO)\(^{S1}\), this suggests that Seam 1 was deposited prior to the EECO.

Further details of sample positions and the lignite sequence at Schöningen can be found in the supplementary material to Robson et al.\(^{S1}\) and Inglis et al.\(^{S2}\).

1.2 Cobham lignite (UK)

A total of 7 samples were used from the Cobham Lignite Bed at Cobham, UK (51.40°N, 0.40°E). Samples were obtained from previous sampling events\(^{S8}\). This lignite was deposited in a low-lying freshwater setting at the southwest shore (very near sea-level) of the North Sea (~48 °N palaeolatitude)\(^{S9,10}\). The Cobham Lignite Bed
at Cobham comprises a thin clay layer (<3 cm) at the base, overlain by a laminated lignite (~55 cm thick). This is succeeded by another thin clay layer (<10 cm) and overlain by a blocky lignite (~130 cm).

The Cobham Lignite Bed at Cobham is underlain by the Upnor Formation, which, at a nearby site, is dated as latest Palaeocene by means of the occurrence of calcareous nannoplankton zone NP9 and magnetochron C25n in its lower part. The shallow-marine Woolwich Formation, which overlies the Cobham Lignite Bed at Cobham, contains the *Apectodinium* acme indicating that it is within the PETM. In addition, at Cobham a negative carbon isotope excursion (CIE) of ~ 1 ‰ is present near the top of the laminated lignite (54.4-55.3 cm) slightly below the middle clay layer, interpreted as being the negative CIE characteristic of the PETM. Here we used 7 samples from the lower laminated lignite below the inferred PETM CIE and thus of very latest Paleocene age.

1.3 Indian lignites

Lignites were collected from mines in several sites in the Rajasthan and Gujarat regions of western India (0-5 °N palaeolatitude). Paleogene-age subbituminous coals from the Meghalaya, Assam, and Nagaland regions of northeastern India were also analysed, but these lignites lacked GDGTs due to higher thermal maturity. All of these sections are associated with over- and/or underlying marine sediments, a characteristic consistent with deposition along the coastal margins of India. The elemental composition (relative concentration of C, H, O, N, and S) and TOC (total organic carbon) of the organic matter of Rajasthan and Gujarat lignites, in general, are suggestive of forest vegetation as the main source and peatification under topogenous conditions. This is further supported by the study of paleomires using petrography based information, using macerals as tools, which indicate deposition under tropical humid climatic conditions at a coastal setting with intermittent fluvial incursions.

Several lignites from the Kachchh Basin were analysed: one sample from the Matanomadh seam (present-day lat./long.: 23°30’05”N, 68°58’E) and two samples from the Panandhro seam (present lat./long.: 23°41’34”N, 68°46’24”E). The Naredi Formation, including these lignite seams, is largely constrained to the early to early middle Eocene on the basis of the age diagnostic foraminifera and pollen. Abundant dinoflagellate cysts in associated shales and mudstones and pollen
dominated by mangrove (*Nypa*) imply an occasional marine influence in a near-shore environment\(^S^{20}\).

In addition, 3 lignite samples from the Khadsaliya Clays of the Saurashtra Basin (present lat./long. 21°39'32"N, 72°12'08"E) were analysed. These lignites are considered early Eocene on the basis of pollen and fungal remains\(^S^{21,22}\). The Khadsaliya Clays comprise gray to greenish-gray clays, carbonaceous clay, and lignite deposited in a woody swamp\(^S^{23}\).

Lastly, 3 lignite samples from the Palana Formation lignites were analyzed; one from the Barsingsar seam, Bikaner basin (present lat./long. 27.84°01N, 73.20°04E); and two from Kasnau Matasukh seam, Nagaur Basin (present lat./long.: 27°06'25"N, 74°04’30”E). The age of the Palana Formation is not well constrained. The Palana Formation was initially assigned to the Eocene on the basis of correlation with lignites in Pakistan\(^S^{24}\) and broad age constraints derived from pollen\(^S^{25,26}\). However, planktonic foraminifera in the overlying Marh Formation have been suggested to be of late Paleocene-early Eocene age\(^S^{27,28}\). In addition, the more recently described osteoglossid and lepisosteid fish are consistent with a Paleocene age for the Palana Formation\(^S^{29}\). As such the Palana Formation is considered of late Paleocene age.

### 1.4 Otaio River section lignites (New Zealand)

The Paleocene to Eocene Broken River Formation overlain by the early Eocene Kauru Formation is exposed in the Otaio River section, near Otaio Gorge, eastern South Island, New Zealand. The Broken River Formation exposures include two lignite seams >1 m thick and several thinner lignite seams\(^S^{30}\). Palynological analyses\(^S^{31}\) and unpublished data indicate that the lower portion of the Otaio River section spans the PETM and the rest of the Broken River Formation exposed in the Otaio River section belongs to the New Zealand stages Waipawan to Mangaorapan (56.0 Ma to 48.9 Ma)\(^S^{32}\). In order to avoid possible overlap with the PETM, we used samples from only the upper lignites, i.e. early Eocene. The 6 samples analysed were taken from thin lignites separated by dark brown sandstones as well as from the c. 2m thick seam at the top of the Broken River Formation exposure in Otaio River. Palynological analyses indicate that the samples fall into the NZ MH1 pollen zone, except for the lowermost sample analysed here (OGp30) which is placed in the PM3b pollen zone.
2. Detection of isoGDGT-5 and -6 in peats and lignites

IsoGDGT-5 and -6 were identified based on 1) comparison of relative retention times (Fig. S2 and S3) with published data\textsuperscript{33}, 2) comparison of LC-MS chromatograms with those of a sample from Champagne pool, a thermal hot spring with a temperature of 75 °C and pH of 5.5 that contains isoGDGT-0 to -8\textsuperscript{34}, and an acid-hydrolysate of the extremophile *Thermoplasma acidophilum* (Matreya, catalog # 1303) (Fig. S4), which is known to produce isoGDGT-0 to -6, but not crenarchaeol\textsuperscript{35}, and 3) co-injection of a peat sample from Peru and the acid-hydrolysed extract of the extremophile *T. acidophilum* (Fig. S5).

3. Environmental controls on the isoGDGT distribution in modern peat

Decades of research, based on both culture experiments and natural archives such as marine sediments and thermal hot springs, have demonstrated that Archaea can alter the distribution of their isoGDGT membrane-spanning lipids in response to changes in environmental parameters such as temperature and pH\textsuperscript{36-42}. However, so far it is unknown whether the isoGDGT distribution in terrestrial settings such as peats varies according to environmental parameters. Below, we discuss the isoGDGT distribution in a wide range of modern peats to assess whether key-environmental parameters such as peat pH and mean air annual temperature have an impact on the isoGDGT pool in peats. The peat samples were obtained from a database as described in detail in Naafs et al.\textsuperscript{43,44}. In short, we analyzed >470 samples from 96 different peatlands from around the world for their GDGT distribution. The database consists of peats from a wide range of environments with a total span in mean annual air temperature (MAAT) from -8 to 27 °C and pH range from 3 to 8. pH data does not exist for all peats and isoGDGTs were below detection limit in a number of peat samples (predominantly in samples from the very top of peat).

3.1 pH dependence

In thermal hot springs, where isoGDGTs are produced by extremophiles, the isoGDGT distribution is influenced by environmental factors such as pH, with increasing cyclisation at lower pH and higher temperatures\textsuperscript{34,41,45}. It is largely unknown whether the isoGDGT distribution in mesophilic (terrestrial) settings is influenced by pH, although Xie et al.\textsuperscript{46} recently demonstrated that the isoGDGT
distributions of a number of Chinese and American mineral soils as well as
enrichments of terrestrial *Thaumarchaeota* grown over a narrow pH range (6.5 to 8)
were correlated with pH.

We found no significant correlation ($R^2<0.2$) between the relative abundance
of individual *iso*GDGTs with cyclopentane rings (both if crenarchaeol was included
and when not) and pH (Fig. S6). The only *iso*GDGT that had a clear correlation
($R^2=0.56$) with pH was *iso*GDGT-5.

We collected a range of samples from peatlands in the Peruvian Amazon.
These tropical peats (MAAT ~26 °C) are located less than 200 km apart, but span a
pH range from 6.1 to 3.8. The peats with pH < 5.1 contain *iso*GDGT-5, whereas those
with a pH > 5.1 do not (Fig. 2 of main manuscript). To explore this further, we
compared the relative abundance of *iso*GDGT-5 relative to the other *iso*GDGTs with
cyclopentane rings (5/(1+2+3+5)) to the calcium concentration of individual samples. *Iso*GDGT-4 was excluded from this ratio due to the co-elution with the [M+H]$^+$ + 2
ion of crenarchaeol that also gives m/z 1294.

Calcium concentrations in peats are a good indicator of nutrient content and
alkalinity (pH) in these peats. Calcium concentrations are low, typically less than
500 mg/kg dry peat, in nutrient-poor ombrotrophic bogs. River-influenced nutrient-
rich minerotrophic peats with pH > 5 are characterized by much higher calcium
concentrations, up to 17,000 mg/kg dry peat. When we plot the 5/(1+2+3+5) ratio
against calcium concentration for individual peat samples (Fig. S7), it is clear that
*iso*GDGT-5 is only present in samples with a low calcium content (< 2000 mg/kg,
mostly < 500 mg/kg dry peat) and hence low pH. The CBT$^{peat'}$-based pH calibration
for peats has a relatively large error of ±0.8 pH units and caution should be taken with
applying CBT$^{peat'}$ to reconstruct absolute pH-values. Even so, the CBT$^{peat'}$ based
pH values for these samples support the inferences derived from Ca ratios. *iso*GDGT-5 is only present in samples with CBT$^{peat'}$-based pH < 5 and predominantly in
samples with CBT$^{peat}$-based pH < 4, as seen in the global dataset (Fig. 2 of the main
manuscript).

In addition, a 750 cm long peat core from the Aucayacu peatland is
characterized by a shift in peat forming environment. Sediments spanning 9 to 5 ka
(below 400 cm) formed under minerotrophic conditions with high calcium
concentrations (high pH), transitioning to low calcium concentrations (low pH) in the
upper 400 cm spanning the late Holocene (last 5 kyr). This transition occurred as
the peat deposit grew higher, out of river influence and into ombrotropic conditions. 

isoGDGT-5 is only present in the ombrotrophic (low pH), upper 400 cm of the core and absent in the underlying minerotrophic (high pH) peat (Fig. S8). Together, the modern surface samples and downcore results indicate a clear pH dependence controlling the abundance of isoGDGT-5.

TEX$_{86}$ and the ring index (RI)$_{S36}$, established indices that reflect the degree of cyclisation of isoGDGTs, did not correlate with pH (Fig. S9).

$$\text{TEX}_{86} = \frac{(\text{isoGDGT}_2 + \text{isoGDGT}_3 + \text{cren.isomer.})}{(\text{isoGDGT}_1 + \text{isoGDGT}_2 + \text{isoGDGT}_3 + \text{cren.isomer.})}$$

Ring index

$$= \frac{(\text{isoGDGT}_1 + 2 \times \text{isoGDGT}_2 + 3 \times \text{isoGDGT}_3 + 4 \times (\text{cren.} + \text{cren.isomer}))}{(\text{isoGDGT}_0 + \text{isoGDGT}_1 + \text{isoGDGT}_2 + \text{isoGDGT}_3 + \text{cren.} + \text{cren.isomer})}$$

3.2 Temperature dependence

Although the relationship differs between settings, both in culture experiments of hyperthermophiles and incubation experiments of mesophiles$^{S36,40}$ as well as natural archives such as marine$^{S38}$ and lake sediments$^{S51}$ and hot springs$^{S34,41}$ the degree of cyclization of isoGDGTs, reflected in RI and/or TEX$_{86}$, is positively correlated with growth temperature. So far it is largely unknown whether the cyclization of isoGDGTs in terrestrial settings is correlated to growth temperature, although there is some recent evidence that suggests that isoGDGTs in mineral soil altitude transects from Tanzania and China differ according to temperature$^{S52,53}$. Our results demonstrate that individual isoGDGTs with 0-3 cyclopentane rings have either no or weak ($0.1 < R^2 < 0.2$) correlations with MAAT (Fig. S10). Also RI (with or without crenarchaeol) and TEX$_{86}$ have no clear correlation with MAAT (Fig. S11). The lack of correlation between the distribution of isoGDGTs and MAAT is likely because the isoGDGT pool is derived from a mixture of GDGT-producing archaeal communities that thrive in peats. In regular marine sediments, the majority of GDGTs are derived from (planktonic) marine Thaumarchaeota that modify their membrane lipids depending on temperature, reflected in the TEX$_{86}$ proxy. However the dominance of isoGDGT-0 and low abundance of crenarchaeol in almost all peat samples, and resulting consistently low ring index, suggests a dominance of methanogenic Euryarchaeota. Consistent with this, if ring indices are calculated, excluding crenarchaeol, they remain poorly correlated to temperature and pH.
For isoGDGT-5 there is currently not enough data to construct a temperature calibration, especially due to the additional influence of pH on the relative abundance of isoGDGT-5 (see section 3.1). However, isoGDGT-5 is absent in ombrotrophic peats from the mid and high latitudes with MAAT < 12 °C. The highest relative abundance of isoGDGT-5 occurs in tropical peats accumulating under highest MAAT, indicating a temperature influence on the relative abundance of isoGDGT-5 (Fig. S10).

A combined pH/temperature control on the distribution of isoGDGT-5 is supported by four decades of research that reveal a pH and growth temperature dependence on isoGDGTs in cultures of acidohyperthermophilic Archaea$^{36}$ and mesocosm experiments of marine Thaumarchaeota$^{40}$, as well as the observed correlation between the degree of cyclization and temperature and/or pH in natural environments such as hot springs$^{34}$ and the open ocean$^{38}$. Amongst cultured organisms, Euryarchaeota belonging to the order Thermoplasmatales as well as Crenarchaeota of the orders Thermoproteales and Sulfolobales are the only known source organisms of isoGDGT-5 to -8$^{42}$; therefore, it is possible that (uncultured mesophilic) relatives of these specific orders are responsible for the presence of isoGDGT-5 to -7 in our modern ombrotrophic tropical peats and early Paleogene lignites.

4. Environmental controls on the brGDGT distribution in modern peat

brGDGTs are membrane-spanning lipids produced by bacteria, likely acidobacteria$^{54-56}$. A decade of research has demonstrated that in mineral soils and lakes the degree of methylation of bacterial brGDGTs depends on temperature$^{57-60}$. We recently expanded this by developing a global peat-specific brGDGT temperature calibration that is based on the degree of methylation of brGDGTs, reflected in the MBT$^\text{5me}$ index$^{57}$, in 470 samples from 96 different of modern peats: MAAT$_{\text{peat}}$$^{43}$. Importantly, the brGDGT data for this peat calibration dataset was generated using the latest HPLC-MS methods$^{61}$ that separate the recently discovered 5- and 6-methyl brGDGTs$^{62}$.

$$\text{MBT}_5^{\text{5me}} = \frac{(I_a + I_b + I_c)}{(I_a + I_b + I_c + IIa + IIb + IIc + IIIa)}$$

$$\text{MAAT}_{\text{peat}} \ (^\circ \text{C}) = 52.18 \times \text{MBT}_5^{\text{5me}} - 23.05 \quad (n = 96, \quad R^2 = 0.76, \quad \text{RMSE} = 4.7 \ ^\circ \text{C})$$
In addition, the degree of cyclization of brGDGTs in mineral soils can be used to reconstruct pH\textsuperscript{57,58}. We recently demonstrated that also in peat the degree of cyclization of brGDGTs, expressed in the CBT\textsubscript{peat} index, is correlated with pH\textsuperscript{543}, although the correlation is weaker compared to that seen in mineral soils.

\[ \text{CBT}_{\text{peat}} = \log \left( \frac{\text{Ib} + \text{IIa} + \text{Ib} + \text{IIb} + \text{IIb}'}{\text{Ia} + \text{IIa} + \text{IIa}'} \right) \]

\[ \text{pH} = 2.49 \times \text{CBT}_{\text{peat}} + 8.07 \quad (n = 51, \quad R^2 = 0.58, \quad \text{RMSE} = 0.8) \]

As lignites are formed from compaction of peat under low burial pressure and temperatures, we apply this peat-specific calibration to reconstruct terrestrial temperatures during the early Paleogene. Inherent to this approach is the assumption that the relationship between MBT\textsuperscript{5me} and temperature was the same during the early Paleogene as at present.

GDGTs can be influenced by thermal maturation. Schouten et al.\textsuperscript{542,63} showed that iso- and brGDGTs are similarly influenced by thermal degradation as GDGTs disappear at hydrous pyrolysis temperatures > 260 °C. Consistent with these experiments, GDGTs appear to be absent in thermally mature coal\textsuperscript{564}. In addition, thermal maturation of GDGTs between ~220 and 260 °C was shown to influence their distribution, with a decrease in the degree of methylation and cyclization\textsuperscript{542,63}. Thus, thermal maturation can not explain the high temperatures we reconstruct for the early Paleogene using lignites as 1) lignites are formed a low burial temperatures (<100 °C) where GDGTs are not influenced, and 2) if thermal degradation would have influenced the brGDGTs in our lignites, this would have lowered MBT\textsuperscript{5me} and hence resulted in low MAAT\textsubscript{peat}.

5. GDGT distribution early Paleogene lignites

As explained in the previous section, we assume that the relationship observed in modern peat between MBT\textsuperscript{5me} and temperature\textsuperscript{543} was the same during the early Paleogene. This assumption is supported by the observation that the broader GDGT distribution in our lignites, of which the majority formed between 45 and 60 degrees latitude during the early Paleogene, is very similar to modern-day distribution of GDGTs in tropical peats. The lignite and tropical modern-day peat are characterized by a high abundance of isoGDGTs with cyclopentane rings (including isoGDGT-5), H-isoGDGTs\textsuperscript{544} (characterized by a covalent bond between the two alkyl chains\textsuperscript{565}), and dominance of brGDGT-Ia over the other brGDGTs. On the other hand, the
GDGT distribution in our lignites looks different compared to a modern-day mid-
latitude peat (Fig. S12). Modern-day mid-latitude peats lack significant amounts of
isoGDGTs with cyclopentane rings, do not contain isoGDGT-5 or H-isoGDGTs, and
penta- and hexamethylated brGDGT are abundant.

Sinninghe Damsté recently used a ternary plot of the brGDGT distribution
in marine sediments and argued that samples that plot off the brGDGT distribution
seen in the modern mineral soil database contain a contribution of in situ brGDGT
production and do not exclusively contain mineral soil-derived terrestrial brGDGTs.
Following this approach, if the GDGT distribution of our early Paleogene lignites was
not produced in peats, the lignite data should plot outside of distribution of brGDGTs
in the modern peat database. However, when we compare the brGDGT distribution in
our early Paleogene lignites to that of modern peats using ternary plots (Fig. S13),
it is clear that the brGDGT distribution of early Paleogene lignites looks very similar
to that in modern peatlands. We then extended this approach by comparing the
isoGDGT distribution in our early Paleogene lignites with that seen in modern peats
and marine core-top sediments (Fig. S14). The isoGDGT distribution in our early
Paleogene lignites looks very similar to that seen in modern-day peats with a very low
proportion of crenarchaeol and looks very different from the isoGDGT distribution of
for example marine sediments. These results highlight that not only MBT5me (and
hence MAATpeat) and the abundance of isoGDGT-5 in our early Paleogene lignites
are similar to modern (tropical) peats, but that the broader GDGT distribution of our
early Paleogene lignites is comparable to a modern-day (tropical) peat.

The only difference is the abundance of isoGDGT-5 encountered in the Indian
lignites, which is higher than found in any modern peat, even in modern tropical peats
(MAAT ~ 26.5 °C) with pH ~ 3. As pH of 3 is the most acidic peat environment
known, the higher abundance of isoGDGT-5 found in the Indian lignites is at least
party related to temperatures higher than MAAT > 26.5 °C, inline with our MAATpeat
temperature estimates. In addition, it is unlikely that the high abundance of isoGDGT-5
in the Indian lignites (compared to the mid-latitude lignites) is the result of a much
lower pH. For example there is independent evidence that at least some of the mid-
latitude lignites were formed in ombrotrophic (low pH) Sphagnum peats and
CBTpeat’ is similar for all lignites.

6. Calculation of paleolatitudes
To be consistent the Ypresian paleolatitudes for all published terrestrial (and marine) sites as well as the lignites were (re)calculated using the models explained in S68. These paleolatitudes might differ slightly from those reported in the original publications. The uncertainty in the paleolatitude calculations for each site is not known, but can be up to several degrees paleolatitude.

7. Compilation of published early Paleogene terrestrial temperatures

We compiled terrestrial temperature data based on a range of proxy methods as plotted in figure 1. The majority of data is obtained using leaf physiognomy from the early Paleogene (late Paleocene and early Eocene) and derived mainly from the Huber and Caballero\textsuperscript{569} and Yang et al.\textsuperscript{570} compilations (see data file). There are different leaf physiognomy methods and we grouped them into three groups 1) data obtained using the Kowalski and Ditcher (K&D) leaf margin analysis calibration\textsuperscript{571}, 2) data obtained using Climate Leaf Analysis Multivariate Program (CLAMP), and 3) other leaf physiognomy data (e.g. using alternative leaf margin analysis calibrations\textsuperscript{572}). Estimates based on nearest living relatives data from plants (e.g. coexistence approach, bioclimatic analysis, etc) were omitted from figure 1 and 3 because of their reliance on correct identification of the nearest living relative. For comparison, figures S15 and S16 include this nearest living relative temperature data. In addition, we omitted a number of data points from the various compilations either because the data was confirmed to be middle Eocene in age (Axel Heidelberg, Geiseltal, Puryear-Buchanan, Kisinger Lakes, Chermurnaut Bay, Fossil Hill Flora - King George Island, and James Ross Basin), represented the PETM (Dragon Glacier - King George Island, Hubble Bubble – Bighorn Basin), the age of the data was poorly constrained (Mahenge and Raichikha), or because the altitude correction applied was uncertain (China Gulch, Camanche Bridge, Pentz, Cherokee Site 1, Fiona Hill, Council Hill, Iowa Hill, You Bet 2, Chalk Bluffs – E., Scotts Flat, Gold Bug, Hidden Gold Camp, Woolsey Flat, Mountain Boy, and Pine Grove 1). From Yang et al.\textsuperscript{570} we used the gridded data adjusted. Where available we show MAAT obtained using different calibrations to show the full uncertainty regarding leaf physiognomy based MAATs. For Climate Leaf Analysis Multivariate Program (CLAMP) data\textsuperscript{570} we use an uncertainty of \( \pm 2 \) °C (http://clamp.ibcas.ac.cn/CLAMP_Uncertainties.html). We want to highlight that use of the Kowalski and Ditcher (K&D) calibration used in Huber and Caballero\textsuperscript{569} often
does lead to higher MAAT estimates compared to other calibrations (e.g. CLAMP), but it is based on a very limited dataset.

All the previously published MBT/CBT-based mineral soil-derived MAAT\textsuperscript{31,73-77}, based on the distribution of \textit{br}GDGTs in (proximal) marine sediments, were revised using the updated MBT'/CBT calibration\textsuperscript{78}. The errors shown in figure 1 for the MBT'/CBT based data were obtained by adding the 5 °C calibration error of the MBT'/CBT calibration\textsuperscript{78} to the one standard deviation of the MBT'/CBT data for each site. For MAAT\textsubscript{peat} the error bars were calculated the same way, but using a calibration error of 4.7 °C\textsuperscript{43}. Only data spanning the late Paleocene and early Eocene (57-48 Myr) was used (see data file). Where the PETM was recognized; data from the PETM was excluded.

We also included temperature data from early Paleogene paleosols from Argentina\textsuperscript{79} and the USA\textsuperscript{80} as well as early Paleogene δ\textsubscript{18}O-based terrestrial temperatures from mammalian tooth enamel and fish (gar) scales, all from the Northern Hemisphere\textsuperscript{81,82}.

8. Compilation of published early Paleogene sea surface temperatures

To compare our early Paleogene terrestrial temperature data with sea surface temperature (SST) data, we compiled all available published data based on the organic geochemical TEX\textsubscript{86} palaeothermometer as well as calcite-based SSTs using Mg/Ca and δ\textsubscript{18}O of pristine planktonic foraminifera and clumped isotopes (see data file). TEX\textsubscript{86}-based SSTs were calculated using the BAYSPAR deep time analog approach\textsuperscript{67,83}. Error bars on TEX\textsubscript{86}-based SST in figure 1 represent the 1σ confidence interval. For the calcite-based proxies the errors were calculated by combining the calibration error and the one standard deviation of the data for each site under different assumptions of early Paleogene seawater composition; -0.64 < δ\textsubscript{18}O\textsubscript{sw} (VSMOW) < -0.21\textsuperscript{84} and 1.5 < (Mg/Ca)\textsubscript{sw} < 5\textsuperscript{85}. Only data spanning the late Paleocene and early Eocene (57-48 Myr) was used (see data file). Where the PETM was recognized SST data from the PETM was excluded.

9. Data model comparison

The model-data comparison shown in Figure 3 is carried out using identical methods to those outlined in Lunt et al.\textsuperscript{84}. In brief, the early Paleogene zonal mean near-surface (~2m) continental air temperature is calculated for each of 7 models using
different $pCO_2$ concentrations; 2x$CO_2$ ECHAM5\textsuperscript{586}, 2x$CO_2$ FAMOUS\textsuperscript{587}, 4x$CO_2$
GISS\textsuperscript{588}, 5x$CO_2$ CCSM3_K\textsuperscript{589}, 6x$CO_2$ HadCM3L\textsuperscript{590}, 16x$CO_2$ CCSM3_WS\textsuperscript{591} and 16x$CO_2$
CCSM3_HS\textsuperscript{569}. The prescribed Eocene paleogeography also varies across the
simulations as shown in the relevant references cited above.

An equivalent temperature (but global rather than continental), from an
equivalent preindustrial simulation from each model is also calculated, and the
difference, early Paleogene minus pre-industrial, is shown as coloured lines in Figure
3. In the nomenclature of Lunt et al.\textsuperscript{584}, this is $[\text{LAT}_{ep} - \text{GAT}_p]$. On top of these
modelled zonal mean anomalies, our compilation of proxy early Paleogene terrestrial
temperatures is plotted, including our new MAAT_{peat} estimates, and including
published estimates of uncertainties. These proxy temperatures are plotted as
anomalies relative to the zonal mean of observed modern global (not exclusively
terrestrial) near-surface air temperatures, (NCEP\textsuperscript{592}), for the period 1981–2010. As
such, the proxy data represent temperature anomalies at a single site, whereas the
modelled results are zonal means.

Supplementary references

S1 Robson, B. E. et al. Early Paleogene wildfires in peat-forming
environments at Schöningen, Germany, Palaeogeogr. Palaeoclimatol.
S2 Inglis, G. N. et al. Mid-latitude continental temperatures through the early
S3 Riegel, W., Wilde, V. & Lenz, O. K. The early Eocene of Schöningen (N-
S4 Inglis, G. N. et al. Ecological and biogeochemical change in an early
Paleogene peat-forming environment: Linking biomarkers and
palynology, Palaeogeogr. Palaeoclimatol. Palaeoecol. \textbf{438}, 245-255,
S5 Ahrendt, H., Köthe, A., Lietzow, A., Marhein, D. & Ritzkowski, S.
Lithostratigraphic, biostratigraphic and radiometric dating of the Early
Eocene at Helmstedt (Lower Saxony), Z. Dtsch. Geol. Ges. \textbf{146}, 450-457,
S6 Crouch, E. M. et al. The Apectodinium acme and terrestrial discharge
during the Paleocene–Eocene thermal maximum: new palynological,
geochemical and calcareous nannoplankton observations at Tawanui,
New Zealand, Palaeogeogr. Palaeoclimatol. Palaeoecol. \textbf{194}, 387-403,
(2003).
S7 Sluijs, A. et al. Environmental precursors to rapid light carbon injection at
S8 Pancost, R. D. et al. Increased terrestrial methane cycling at the


Supplementary figure captions

Figure S1; Present-day location of the lignites used in this study.

Figure S2; HPLC-APCI-MS base peak chromatogram (top) and mass chromatograms of a tropical peat sample from Peru (the Aucayacu peatland, 330 cm depth). Numbers indicate number of cyclopentane moieties in the isoGDGTs, while roman numbers highlight the different brGDGTs. Cren = crenarchaeol and reg.iso= crenarchaeol regioisomer. In H-isoGDGTs the two biphytane chains are covalently bound by a carbon-carbon bond\textsuperscript{665}.

Figure S3; HPLC-APCI-MS base peak chromatogram (top) and mass chromatograms of an early Paleogene lignite sample from Cobham (CL70, 11.95 cm). Numbers indicate number of cyclopentane moieties in the isoGDGTs, while roman numbers highlight the different brGDGTs. Cren = crenarchaeol and reg.iso= crenarchaeol regioisomer. In H-GDGTs the two biphytane chains are covalently bound by a carbon-carbon bond.

Figure S4; HPLC-APCI-MS base peak chromatograms of A) a tropical peat sample from Peru (the Aucayacu peatland, 330 cm depth), B) sample from the Champagne pool hot spring, and C) acid-hydrolized extract of the extremophile Thermoplasma acidophilum.

Figure S5; HPLC-APCI-MS base peak chromatograms of A) a tropical peat sample from Peru (the Aucayacu peatland, 330 cm depth) and B) co-injection of the tropical...
peat sample with the acid-hydrolized extract of the extremophile *Thermoplasma acidophilum* that contains isoGDGT-5 but not crenarchaeol.

Figure S6; Fractional abundance of the individual isoGDGTs versus peat pH.

Horizontal bars reflect range of peat pH\(^{S43}\), while vertical bars represent 1σ from the average fractional abundance and are based on the analysis of multiple samples from the same peatland. Fractional abundances < 0.001 are not shown.

Figure S7; Relative abundance of isoGDGT-5 (%) versus A) calcium content (a measure of pH) for individual samples in a range of tropical peatlands from Peru that all experience the same climate. (Ca content from\(^{S48,49}\)) and B) CBT\(_{\text{peat}}\)-based pH. Note that Ca data is not available for every sample.

Figure S8; Downcore relative abundance of isoGDGT-5 (%), orange and calcium content (mg/kg, blue) in the 750 cm long peat core from the Aucayacu peatland in Peru that spans the last 9 kyr. Pie charts reflect the relative distribution of isoGDGTs in the top and bottom of the peat. (Radiocarbon ages from\(^{S50}\))

Figure S9; A) Ring index and B) TEX\(_{86}\) versus peat pH. Horizontal bars reflect range of peat pH\(^{S43}\), while vertical error bars represent 1σ from the average and are based on the analysis of multiple samples from the same peatland.

Figure S10; Fractional abundance of the individual isoGDGTs versus overlying mean annual air temperature. Vertical error bars represent 1σ from the average fractional abundance and are based on the analysis of multiple samples from the same peatland. Samples with a fractional abundance < 0.001 are not shown.

Figure S11; A) Ring index and B) TEX\(_{86}\) versus mean annual air temperature. Vertical error bars represent 1σ from the average and are based on the analysis of multiple samples from the same peatland.

Figure S12; HPLC-APCI-MS base peak chromatograms highlight the iso- and brGDGT distribution in A) early Paleogene lignite from UK (Cobham CL70, 11.95 cm), B) modern mid-latitude peat samples from Germany (Bissendorfer Moor, 18 cm
depth), and C) modern tropical peat sample from Peru (the Aucayacu peatland, 330 cm depth). Modern MAAT Bissendorfer Moor and Aucayacu are 8.9 °C and 26 °C, while pH for these peats is 4 and 3.7, respectively.

Figure S13; Ternary plot of the \( br \)GDGT-distribution in the modern peat database\(^{S43}\) and all early Paleogene lignites used in this study. Plot shows the relative abundance of the tetra- (\( br \)GDGT-Ia, -Ib, and Ic), penta- (\( br \)GDGT-IIa, -IIa’, -IIb, -IIb’, -IIc, and -IIc’), and hexamethylated \( br \)GDGTs (\( br \)GDGT-IIIa, -IIIa’, -IIIb, -IIIb’, -IIIc, and -IIIc’).

Figure S14; Ternary plot of the \( iso \)GDGT-distribution in the modern peat database, marine core-top sediments\(^{S67}\), and all early Paleogene lignites used in this study. Plot shows the relative abundance of the \( iso \)GDGT with no rings (\( iso \)GDGT-0), \( iso \)GDGTs with 1 to 3 cyclopentane rings (\( iso \)GDGT-1, -2, and -3), and \( iso \)GDGT with a cyclohexane ring (crenarchaeol).

Figure S15; Same as figure 1 of the main manuscript, but including estimates based on nearest living relatives data (e.g. coexistence approach, bioclimatic analysis, etc.).

Leaf physiognomy methods: K&D - Kowalski and Ditcher leaf margin analysis calibration\(^{S70}\); CLAMP - Climate Leaf Analysis Multivariate Program\([Yang, 2011 #1991]\); other leaf physiognomic - for example using alternative leaf margin analysis calibrations\(^{S71}\). MAAT – mean annual air temperature.

Figure S16; Same as figure 3 of the main manuscript, but including estimates based on nearest living relatives data (e.g. coexistence approach, bioclimatic analysis, etc.).

For abbreviations see Figure S15.

Figure S17; Global temperature anomaly between the early Paleogene and present for all available terrestrial temperature data at the paleolatitude of each location together with the zonal mean anomaly simulated by a range of climate models; 2xCO\(_2\) ECHAM5\(^{S86}\), 2xCO\(_2\) FAMOUS\(^{S87}\), 4xCO\(_2\) GISS\(^{S88}\), 5xCO\(_2\) CCSM3\(_K\)\(^{S89}\), 6xCO\(_2\) HadCM3L\(^{S90}\), 16xCO\(_2\) CCSM3\(_W\)\(^{S91}\) and 16xCO\(_2\) CCSM3\(_H\)\(^{S69}\).
Figure S2

Tropical peat sample
(The Aucayacu peatland, 330 cm depth)

Retention time (min.)

Intensity

H-GDGT-0

m/z 1302

H-GDGT-0

m/z 1300

m/z 1298

m/z 1296

m/z 1294

m/z 1292

m/z 1290

Retention time (min.)
early Paleogene sample Cobham lignite (CL70, 11.95 cm)
Figure S4

Retention time (min.)

Intensity

A) Aucayacu tropical peatland
B) Champagne pool hot spring
C) Extremophile *T. acidophilum*

4+cren
Figure S5

B) AU-330 + \( T.\text{acidophilum} \)

\( \text{GDGT-5} \)

Retention time

m/z 1292

A) AU-330

\( \text{GDGT-5} \)

Retention time

m/z 1292
Figure S6

- **isoGDGT-0**: 
  - $R^2 = 0.00$ 
  - $n = 51$

- **isoGDGT-1**: 
  - $R^2 = 0.08$ 
  - $n = 51$

- **isoGDGT-2**: 
  - $R^2 = 0.10$ 
  - $n = 51$

- **isoGDGT-3**: 
  - $R^2 = 0.14$ 
  - $n = 51$

- **Crenarchaeol**: 
  - $R^2 = 0.12$ 
  - $n = 46$

- **isoGDGT-5**: 
  - $R^2 = 0.78$ 
  - $n = 12$
Figure S7

A) Relative abundance of isoGDGT-5 (%) vs. Calcium (mg/kg).

B) Relative abundance of isoGDGT-5 (%) vs. CBT

peat-based pH.
Figure S8

- **Relative abun. isoGDGT-5 (%)**
  - Depth (cm) vs. Depth (cm)
  - Calcium (mg/kg)

- **Pie charts**
  - Crenarchaeol isom.
  - GDGT-0
  - GDGT-1
  - GDGT-2
  - GDGT-3
  - GDGT-5
  - GDGT-6

- Key:
  - %isoGDGT-5
  - [Ca]

- Time markers:
  - 2025 yr
  - 3010 yr
  - 4035 yr
  - 5950 yr
  - 8110 yr
  - 8870 yr

- **Figure details**:
  - GDGT-0, GDGT-1, GDGT-2, GDGT-3, GDGT-5, GDGT-6
  - Crenarchaeol, Crenarchaeol isom.
Figure S9

**Ring index**

A) $R^2 = 0.01$
$n = 51$

**TEX$_{86}$**

B) $R^2 = 0.02$
$n = 51$
Figure S10

**Mean annual air temperature (°C)**

<table>
<thead>
<tr>
<th>IsoGDGT</th>
<th>R²</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>isoGDGT-0</td>
<td>0.07</td>
<td>81</td>
</tr>
<tr>
<td>isoGDGT-1</td>
<td>0.06</td>
<td>81</td>
</tr>
<tr>
<td>isoGDGT-2</td>
<td>0.03</td>
<td>81</td>
</tr>
<tr>
<td>isoGDGT-3</td>
<td>0.16</td>
<td>81</td>
</tr>
<tr>
<td>Crenarchaeol</td>
<td>0.00</td>
<td>69</td>
</tr>
<tr>
<td>isoGDGT-5</td>
<td>0.16</td>
<td>16</td>
</tr>
</tbody>
</table>
Figure S11

A) Ring index

R² = 0.05
n = 81

B) TEX_{86}

R² = 0.15
n = 81
Figure S12

Retention time
0 1 2 3 4 +
Cren. 5 H-0
Ia
IIa
isoGDGTs
H-isoGDGTs
brGDGTs
0 1 2 3 4 +
Cren.
5
IIa
A) Ib Ic
IIIa
B) C)
Intensity
Ib Ic
Figure S14

Proportion isoGDGT-0 vs. Proportion isoGDGT-1 to -3 vs. Proportion crenarchaeol triangle plot.

- Modern peat
- Indian lignite
- Cobham lignite
- Schöningen lignite
- Otaio lignite
- Marine core top sediments

Data points for different samples are color-coded accordingly.
Figure S16

A) Terrestrial temperature anomaly (°C) as a function of latitude for different CO2 doubling scenarios. The error bars indicate the range of modeled temperatures.

B) Similar to A, but focusing on a different region. The markers and lines represent various models and scenarios.
Figure S17

Terrestrial Temperature Anomaly

- 16xCO$_2$: CCSM3_H
- 16xCO$_2$: CCSM3_W
- 6xCO$_2$: HadCM3L
- 5xCO$_2$: CCSM3_K
- 4xCO$_2$: GISS
- 2xCO$_2$: FAMOUS
- 2xCO$_2$: ECHAM5

Legend:
- MAAT$_{\text{peat}}$
- K&D calibr.
- CLAMP
- Other leaf physiognomic
- MBT'/CBT
- Paleosol
- Mammal $\delta^{18}$O
- Nearest living relative