



- **Mastrichtian-Rupelian** paleoclimates in the southwest Pacific a critical evaluation of biomarker paleothermometry and dinoflagellate cyst Z 3 paleoecology at Ocean Drilling Program Site 1172 4 Peter K. Bijl^{1*}, Joost Frieling^{1,2}, Margot J. Cramwinckel^{1,3}, Christine Boschman¹, Appy 5 Sluijs¹, Francien Peterse¹ 6 7 8 ¹Department of Earth Sciences, Utrecht University, Utrecht, the Netherlands. 9 ²now at: Department of earth Sciences, University of Oxford, South Parks Road OX1 10 3AN, UK 11 ³now at: School of Ocean and Earth Science, National Oceanography Centre 12 Southampton, University of Southampton, Southampton, UK 13 14 * corresponding author. email: p.k.bijl@uu.nl 15 16 Abstract Sea surface temperature (SST) reconstructions based on isoprenoid glycerol dialkyl 17 18 glycerol tetraether (isoGDGT) distributions from the Eocene southwest (sw) Pacific 19 Ocean are unequivocally warmer than can be reconciled with state-of-the-art fully 20 coupled climate models. However, the SST signal preserved in sedimentary archives 21 can be affected by contributions of additional isoGDGT sources. Methods now exist to 22 identify and possibly correct for overprinting effects on the isoGDGT distribution in 23 marine sediments. We here use the current proxy insights to assess the reliability of 24 the isoGDGT-based SST signal in 69 newly analysed and 242 re-analysed sediments ODP Site 1172 (East Tasman Plateau, Australia) following state-of-the-art 25 26 chromatographic techniques, in context of paleo-environmental and 27 paleoclimatologic reconstructions based on dinoflagellate cysts. The resulting ~130 28 kyr-resolution Maastrichtian-Oligocene TEX₈₆-based SST record confirms previous 29 conclusions of anomalous warmth in the early Eocene sw Pacific and remarkably cool 30 conditions during the mid-Paleocene. Dinocyst diversity and assemblages show a 31 strong response to the local SST evolution, supporting the robustness of the TEX₈₆
- 32 record.





Soil-derived branched GDGTs stored in the same sediments are used to reconstruct 33 34 mean annual air temperature (MAAT) of the nearby land using the MBT'_{5me} proxy. 35 MAAT is consistently lower than SST during the early Eocene, independent of the 36 calibration chosen. General trends in SST and MAAT are similar, except for: 1) an 37 enigmatic absence of MAAT rise during the Paleocene-Eocene Thermal Maximum and 38 Middle Eocene Climatic Optimum, and 2) a subdued middle-late Eocene MAAT 39 cooling relative to SST. Both dinocysts and GDGT signals suggest a mid-shelf 40 depositional environment with strong river-runoff during the Paleocene-early 41 Eocene, progressively becoming more marine thereafter. This trend reflects gradual 42 drying and more pronounced wet/dry seasons in the northward drifting Australian 43 hinterland, which may also explain the subdued middle Eocene MAAT cooling relative 44 to that of SST. The overall correlation between dinocyst assemblages, marine 45 biodiversity and SST changes suggests that temperature exerted a strong influence on the surface-water ecosystem, probably in part through sea level changes caused by AC steric effects. Finally, we find support for a potential temperature control on 41 48 compositional changes of branched glycerol monoalkyl glycerol tetraethers 49 (brGMGTs) in marine sediments. It is encouraging that a critical evaluation of the 50 GDGT signals confirms the vast majority of the generated data is reliable. However, this also implies the high TEX₈₆-based SSTs for the Eocene sw Pacific, and the 51 52 systematic offset between absolute TEX₈₆-based SST and MBT'_{5me}-based MAAT 53 estimates remain <u>maxplained.</u> 54





55 **1. Introduction**

| 57Reconstructions of deep-sea (Westerhold et al., 2020) and sea surface58temperature (Bijl et al., 2009; 2013a; Cramwinckel et al., 2018; Frieling et al., 2014;59Hollis et al., 2014; 2019; Inglis et al., 2015, 2020) O'Brien et al., 2017; Evans et al.,602018; O'Comor et al., 2019; Sluijs et al., 2020) have revealed overall cool climate in61the Maastrichtian and Paleocene, long-term warming towards the early Eocene62Climatic Optimum (EECO; 53.4 - 49.2 Ma), and subsequent cooling during the middle63and late Eocene (48.6 - 33.6 Ma). The EECO stands out as particularly warm, with ice-64free polar regions (Bijl et al., 2013; Une es et al., 2017). Pross et al., 2012; Frieling et63al., 2014). Certain southern high-laticude regions retain warm-temperate conditions64into the late Eocene65into the late Eocene66transition (Salamy and Zachos, 1999; Bohaty et al., 2012), even into the early67Oligocene (Hartman et al., 2018; Passchier et al., 2013; O'Brien et al., 2020).78Variations in atmospheric CO2 concentrations (Beerling and Royer, 2011; Anagnostou79year climatic trends (Cramwinckel et al., 2018). While equatorial proxy-based sea71strace temperatures (SSTs) and deep-sea temperatures, assumed to reflect high-73kitude SSTs, show good correspondence with numerical model simulations under74bose from model simulations (Bijl et al., 2009; Cramwinckel et al., 2018; Hollis et al.,75Focene boundary conditions and with varying CO2 forcing (Cramwinckel et al., 2014).76appecifically, emerical models are c | 56 | 1.1 The Paleogene Southwest Pacific Ocean |
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| Poil2; Lunt et al., 2021), despite proposed zonal heterogeneity (Douglas et al., 2014). Specifically, commerical models are currently unable to simulate a paleoclimate in which the annual SST difference between the equatorial Atlantic Ocean (Cramwinckel et al. 2018) and the sw Pacific Ocean (Bijl et al., 2013a; Hollis et al., 2012) is as small as the proxy data suggests. Without this model-data mismatch resolved, it remains unclear to what extent numerical models properly simulate polar amplification of Paleogene climates and if the current proxies properly reflect high-latitude | 76 | proxy-based SST reconstructions of the southwest (sw) Pacific remain warmer than |
| 5pecifically, merical models are currently unable to simulate a paleoclimate in which the annual SST difference between the equatorial Atlantic Ocean (Cramwinckel et al. 2018) and the sw Pacific Ocean (Bijl et al., 2013a; Hollis et al., 2012) is as small as the proxy data suggests. Without this model-data mismatch resolved, it remains unclear to what extent numerical models properly simulate polar amplification of Paleogene climates and if the current proxies properly reflect high-latitude | 77 | those from model simulations (Bijl et al., 2009; Cramwinckel et al., 2018; Hollis et al., |
| which the annual SST difference between the equatorial Atlantic Ocean (Cramwinckel et al. 2018) and the sw Pacific Ocean (Bijl et al., 2013a; Hollis et al., 2012) is as small as the proxy data suggests. Without this model-data mismatch resolved, it remains unclear to what extent numerical models properly simulate polar amplification of Paleogene climates and if the current proxies properly reflect high latitude | 78 | 2012; Lunt et al., 2021), despite proposed zonal heterogeneity (Douglas et al., 2014). |
| et al. 2018) and the sw Pacific Ocean (Bijl et al., 2013a; Hollis et al., 2012) is as small as the proxy data suggests. Without this model-data mismatch resolved, it remains unclear to what extent numerical models properly simulate polar amplification of Paleogene climates and if the current proxies properly reflect high-latitude | 79 | specifically, |
| as the proxy data suggests. Without this model-data mismatch resolved, it remains unclear to what extent numerical models properly simulate polar amplification of Paleogene climates and if the current proxies properly reflect high-latitude | 80 | which the annual SST difference between the equatorial Atlantic Ocean (Cramwinckel |
| unclear to what extent numerical models properly simulate polar amplification of Paleogene climates and if the current proxies properly reflect high-latitude | 81 | et al. 2018) and the sw Pacific Ocean (Bijl et al., 2013a; Hollis et al., 2012) is as small |
| 84 Paleogene climates and if the current proxies properly reflect high-latitude | 82 | as the proxy data suggests. Without this model-data mismatch resolved, it remains |
| | 83 | unclear to what extent numerical models properly simulate polar amplification of |
| temperatures under greenhouse conditions (Lunt et al., 2012). | 84 | Paleogene climates and if the current proxies properly reflect high latitude |
| | 85 | temperatures under greenhouse conditions (Lunt et al., 2012). |





| 86 | Proxyevidence for warmth in the Eocene sw Pacific region derives from a |
|-----|---|
| 87 | suite of organic and calcite-based proxies. In terms of the latter, oxygen isotope ratios |
| 88 | (δ^{18} O) and trace element ratios (Mg/Ca) from well-preserved planktonic foraminifera |
| 89 | indicate warm temperatures from Eocene sections in New Zealand (Creech et al., |
| 90 | Hines et al., 2017; Hollis et al., 2009; 2012). These proxies require assumptions |
| 91 | regarding seawater chemistry (Mg/Ca, δ^{18} O seawater, pH) that carry significant |
| 92 | <mark>empertainty</mark> (e.g., <mark>Mandon et al., 2020, Evans et al. 2018</mark>). The application of clumped |
| 93 | isotope paleothermometry has great potential to partially alleviate such concerns , as |
| 94 | evident from work on Seymour Island (Douglas et al., 2014). Pollen-based vegetz 🔂 |
| 95 | reconstructions from New Zealand, the Tasman region and Wilkes Land (Carpenter et |
| 96 | al., 2012; Contreras et al., 2013; 2014; Huurdeman et al., 2020; Pross et al., 2012), |
| 97 | however, confirm warm conditions, and arguably deliver the best constraints on |
| 98 | winter temperatures. This is because of fundamental physiological restrictions in |
| 99 | their individual tolerances (e.g., Reichgelt et al., 2018), whereas mean annual air |
| 100 | temperature (MAAT) reconstructions from pollen assemblages are complicated |
| 101 | because AT exerts much less control on the standing vegetation than seasonal |
| 102 | temperature and hydrological extremes. Reconstructions of the warm Eocene |
| 103 | primarily relied on organic geochemical proxies, notably TEX ₈₆ (Bijl et al., 2009; |
| 104 | 2013a; Cramwinckel et al., 2018; 2020; Crouch et al., 2020; Hollis et al., 2009; 2012; |
| 105 | Sluijs et al., 2006; 2009; 2011). These absolute SST estimates for the sw Pacific are |
| 106 | closer to those from the equatorial Atlantic as they are to the deep-sea (Cramwinckel |
| 107 | et al., 2018), which is surprising given that the South Pacific was presumably the |
| 108 | dominant region of deep-water formation during the Eocene (Huber and Thomas, |
| 109 | 2010; Thomas et al., 2003; 2014). |
| 110 | |
| 111 | 1.2 GDGT paleothermometry |
| 112 | TEX ₈₆ utilizes the correspondence of higher abundances of cyclopentane rings |
| 113 | in sedimentary archaeal membrane lipids termed isoprenoid glycerol dialkyl glycerol |
| 114 | tetraethers (isoGDGTs) with higher SST of the overlying surface water (Schouten et |
| 115 | al., 2002). This relation is attributed to a viscoelastic adaptation of the membrane of |
| 116 | pelagic Thaumarcheota, the dominant source organisms of isoGDGTs, to temperature |
| 117 | (Schouten et al., 2002; 2013). For some periods in geological deep time, including the |
| | |





| 118 | Paleocene and Eocene, TEX $_{86}$ calibrations based on GDGTs in core top sediments need |
|--|---|
| 119 | to be extrapolated above the modern SST range (\sim 30°C) to estimate SST. The |
| 120 | linearity of the relation at and beyond the high-end of the core-top calibration is |
| 121 | poorly known , leading to very high uncertainty in SST estimates at the warm 👼 of |
| 122 | the calibration (Hollis et al., 2019). However, as the absolute TEX ₈₆ values of many |
| $\begin{array}{c} 122\\ =\\ 123\\ \end{array}$ | Eocene sediments exceed those observed for modern core-tops, even the most |
| 124 | conservative calibrations yield SSTs >30°C in the warmest intervals. Increasingly, |
| 125 | such temperatures are corroborated by estimates from other SST proxies (e.g., Zachos |
| 126 | et al, 2006; Frieling et al., 2017; Evans et al., 2018). |
| 127 | Along with calibration uncertainties, a number of confounding factors have |
| 128 | been identified since first publication of the isoGDGT-based SST records from the sw |
| 129 | Pacific (Hollis et al., 2009; 2012; Bijl et al., 2009; 2013), relating to a suite of pre-, syn- |
| 130 | , and post-depositional processes that might alter the pelagic isoGDGT signal in |
| 131 | marine sediments. IsoGDGT contributions from methanogenic (Blaga et al., 2009) and |
| 132 | methanotrophic (Weijers et al., 2011; Zhang et al., 2011) archaea, deep-dwelling |
| 133 | archaea (Taylor et al., 2013) and terrestrial sources (Hopmans et al., 2004; Weijers et |
| 134 | al., 2006) to the sedimentary isoGDGT pool have been determined . These factors can |
| 135 | be recognized in GDGT distributions, leading to better interpretation of TEX $_{86}$ based |
| 136 | SST reconstructions. However, the influence of growth phase (Elling et al., 2014), and |
| 137 | environmental ammonium and oxygen concentrations (Qin et al., 2015; Hurley et al., |
| 138 | 2016) on sedimentary isoGDGT distributions are as yet poorly constrained. |
| 139 | Branched GDGTs (brGDGTs) produced by soil bacteria provide |
| 140 | reconstructions of mean annual air temperature (MAAT) using the MBT' _{5me} index (De |
| 141 | Jonge et al., 2014a; Naafs et al., 2019; Peterse et al., 2012; Weijers et al., 2007; Dearing |
| 142 | Crampton-Flood et al., 2020). Based on brGDGTs supplied to marine sediments, the |
| 143 | MAAT evolution of Australian-New Zealand continents followed the trends in deep- |
| 144 | and surface ocean temperature remarkably well, but with much lower absolute |
| 145 | values than SST (Bijl et al., 2013a; Pancost et al., 2013). Albeit to a lesser extent than |
| 146 | SST, southern high-latitude MAAT reconstructions remain warmer than model |
| 147 | simulations (IIII) ber and Caballero, 2011) as well, even when simulations replicate |
| 148 | equatorial surface and global deep ocean temperatures (Cramwinckel et al., 2018). |
| 149 | With improved analytical techniques (Hopmans et al., 2016), brGDGT isomers with a |
| | |

5





| 150 | methylation on the 5- or the 6-position of the alkyl chain can now be separated and |
|-----|--|
| 151 | quantified (De Jonge et al., 2013). With this separation, the pH co-dependence of the |
| 152 | brGDGT signal can be removed, isolating the temperature relation (De Jonge et al., |
| 153 | 2014a; Naafs et al., 2017a). The separation of brGDGT isomers also allows <mark>to</mark> |
| 154 | recognize and correct for potential contributions of aquatic brGDGTs to the soil- |
| 155 | derived brGDGT signal stored in marine sediments that complicate the use of |
| 156 | brGDGTs as continental paleothermometer in continental margin sediments (De |
| 157 | Jonge et al., 2014b; 2015; Dearing Crampton-Flood et al., 2018; Peterse et al., 2009; |
| 158 | Sinninghe Damsté, 2016; Tierney and Russell, 2009; Zell et al., 2013). |
| 159 | Lipids from a related biomarker family, the branched glycerol monoalkyl |
| 160 | glycerol tetraethers (brGMGTs), were identified in the marine realm in core-top |
| 161 | sediments (Liu et al., 2012), oxygen minimum zones (Xie et al. 2014), and later in peat |
| 162 | (Naafs et al 2018a) and East African lake sediments (Baxter et al 2019), the latter |
| 163 | identifying 7 individual brGMGTs. The brGMGTs contain a covalent bond connecting |
| 164 | the two alkyl chains. The abundance of brGMGTs relative to that of brGDGTs, as well |
| 165 | as the relative distribution of brGMGT isomers seems to vary with temperature in |
| 166 | some degree (Baxter et al., 2019; Naafs et al., 2018a; Tang et al., 2021), although this |
| 167 | is thus far only based on empirical relationships. The exact sources of these |
| 168 | compounds, and consistency of such signals in various terrestrial, lacustrine and |
| 169 | marine realms, are as yet not fully understood. The degree of methylation of a specific |
| 170 | subset of brGMGT compounds (Naafs et al., 2018a, Sluijs et al. 2020) is however |
| 171 | similar to those in brGDGTs for which membrane stability regulation is proposed as |
| 172 | underlying mechanism (Weijers et al., 2007). Paleogene marine sediments from the |
| 173 | Arctic Ocean do contain abundant brGMGTs (Sluijs et al., 2020) , and t hese seem to be |
| 174 | produced in the marine system with a strong oxygen minimum zone , and substantial |
| 175 | terrestrial input. While the degree of methylation of acyclic brGMGTs (HMBT $_{ m acyclic}$) |
| 176 | does show similar trends to TEX ₈₆ in the Paleogene Arctic record, $\frac{1}{2}$ MGTI, which |
| 177 | was tentatively calibrated to temperature using a suite of tropical takes (Baxter et al., |
| 178 | 2019) does not (Sluijs et al., 2020). The response of brGMGTs to environmental and |
| 179 | climatic changes, as well as their sourc ing in the marine realm, is clearly diverse, |
| 180 | complex and, as yet, poorly understood. |
| 181 | |

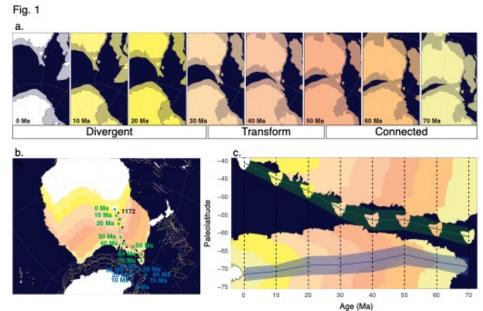




| 182 | 1.3 Revisiting GDGTs, and investigating GMGTs at Site 1172 |
|-----|--|
| 183 | From the new insights about the GDGT-based proxies, it is clear that assessing |
| 184 | the reliability of previously published GDGT-based temperature reconstructions |
| 185 | requires a revisit and a detailed constraints on past environmental, climatological and |
| 186 | depositional conditions. Moreover, the presence and proxy potential of brGMGTs the |
| 187 | early Paleogene sw Pacific has not yet been assessed. To this end, we have revisited ${f a}$ |
| 188 | sedimentary record from the sw Pacific Ocean: O DP Site 1172 on the East Tasman |
| 189 | Plateau (ETP), which contains an almost complete succession of late Cretaceous to |
| 190 | early Oligocene sediments (Bijl et al., 2013b; 2014; Brinkhuis et al., 2003; |
| 191 | Schellenberg et al., 2004; Stickley et al., 2004a). We have newly analysed isoGDGTs, |
| 192 | brGDGTs, and brGMGTs from the Maastrichtian and Paleocene section of the record, |
| 193 | and re-analysed previously published sample sets using the chromatography method |
| 194 | with improved compound separation (Hopmans et al., 2016). We critically evaluate |
| 195 | the biomarker results using established indicators for pre-, syn- or post-depositional |
| 196 | overprint of the primary sea surface and air temperature signals. In addition, we use |
| 197 | dinoflagellate cyst assemblages and terrestrial palynomorph abundance as recorders |
| 198 | of paleoenvironmental change on the continental shelf, for constraints on |
| 199 | depositional, environmental and hydrological changes, in order to aid interpretations |
| 200 | of the GDGT indices. We also evaluate the nature, source and possible temperature |
| 201 | affinity of the brGMGTs in our record. After this critical (re-)evaluation, we interpret |
| 202 | the Maastrichtian to early Oligocene sea surface and air temperature, and |
| 203 | paleoenvironmental evolution of the sw Pacific region. |
| | |







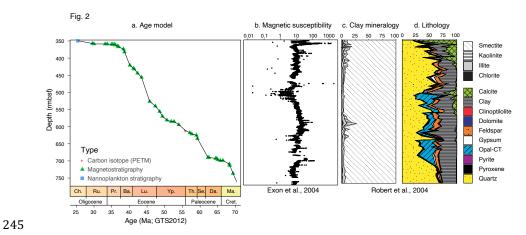
204 205 Figure 1. Absolute plate tectonic changes around Tasmania during the Maastrichtian 206 to recent. a. **TG** opening in 8 time slices, from **70** Ma to present. Relative tectonic 207 motion between Australia and Antarctica is limited until about 53 Ma, transfer until 208 34 Ma and divergent from 34 Ma onwards. b. Like a, but on an orthographic 209 projection (compiled with Gplates, using paleomagnetic reference frame (Torsvik et 210 al., 2012) from Seton et al. (2012)). Green and blue lines and dots represent the 211 pathways of Tasmania and Cape Adare, respectively, black dashed line and dots 212 represent the paleo-position of Site 1172. The absolute paleolatitude of Cape Adare, 213 Antarctica (blue line, blue shading = uncertainty), as indicative of the plate tectonic 214 motion around the pathway of the Tasman Current, and of Tasmania (green line, 215 green shading = uncertainty), as indicative of the plate tectonic motion of the source 216 area of the terrestrial organic matter, in 10 Myr time steps from 70–0 Ma. obtained 217 from paleolatitude.org (Van Hinsbergen et al., 2015). Plate contours represent 218 paleolatitude of present-day shorelines, for orientation (obtained from Gplates). Note that in visualizations b and c, submerged continental crust is not shown, but does 219 220 limit TG opening. 221 222





223 2. Material

| 224 | 2.1 Site locality and tectonic evolution |
|-----|--|
| 225 | Since the Cenomanian, the continental complex including Australia, Tasmania, |
| 226 | ETP and the South Tasman Rise (STR) tectonically moved as one continental plate |
| 227 | (Müller et al., 2000) – here referred to as Australia. True polar wander, although |
| 228 | relatively poorly constrained, the caused absolute plate motions relative to the spin |
| 229 | axis of the Earth (Torsvik et al., 2012; Van Hinsbergen et al., 2015). On a |
| 230 | paleomagnetic reference frame, Antarctica and Australia rotated northwestward by |
| 231 | 3° of latitude and over 3° longitude between 70 and 50 Ma (Fig. 1), with only little |
| 232 | transform displacement between them. From 50 Ma onwards, the tectonic drift |
| 233 | orientation of Antarctica shifted slightly more southwards than that of Australia, |
| 234 | causing left transform motion between notably west of Cape Adare and west |
| 235 | Tasmania (Fig. 1; Williams et al., 2019). Although this divergence effectively opened |
| 236 | the Tasmanian Gateway (TG) to surface flow of the Antarctic Counter Current close to |
| 237 | the early-middle Eocene boundary (Bijl et al., 2013a), probably through subsidence of |
| 238 | the Antarctic continental margin and STR, rapid northward movement of Australia |
| 239 | and southward movement of East Antarctica did not start before about 40 Ma (Fig. 1; |
| 240 | Cande and Stock, 2004; Seton et al., 2012). Yet, a connection between Australian and |
| 241 | Antarctic continental blocks persisted until 34Ma when transform motion between |
| 242 | STR and Wilkes Land changed into spreading and ocean crust formation (Cande and |
| 243 | Stock, 2004; Seton et al., 2012). |
| 244 | |







| 246 | |
|-----|--|
| 246 | Figure 2. a. Age tie points used to construct the age model for ODP Site 1172, based on |
| 247 | carbon isotope (Bijl et al., 2010; Sluijs et al., 2011), magnetostratigraphic (Bijl et al., |
| 248 | 2010; Houben et al., 2019; Stickley et al., 2004a), and nannofossil (Houben et al., |
| 249 | 2019) age constraints. b. Shipboard magnetic susceptibility from Exon et al. (2001). c. |
| 250 | Shore-based clay mineralogy and d. smear slide-based lithological observations from |
| 251 | Robert (2004). |
| 252 | |
| 253 | |
| 254 | 2.2 Lithology |
| 255 | A 760 meter thick sediment sequence was recovered at ODP Site 1172, on the |
| 256 | western part of the ETP, east of Tasmania, Australia (Exon et al., 2001). We studied |
| 257 | the interval from 760 meters below sea floor (mbsf) to about 350 mbsf in a composite |
| 258 | (Röhl et al., 2004a) of Hole A and D. The carbonate ooze from 350 mbsf upwards did |
| 259 | not yield any GDGTs. The studied succession consists broadly of green-grey silty |
| 260 | claystones from 760 to ${\sim}505$ mbsf, grading into dark grey to black silty claystones |
| 261 | from 636 – 625 mbsf, then a return of the greenish grey silty claystones, which is |
| 262 | interrupted by a more lithified unit at 610 mbsf (Exon et al., 2001; Röhl et al., 2004a, |
| 263 | b; Schellenberg et al., 2004; Sluijs et al., 2011). The silty claystones turn gradually and |
| 264 | cyclically lighter from 570 mbsf upwards until ${\sim}361$ mbsf, with gradually more |
| 265 | calcium carbonate and diatoms p reserved (Röhl et al., 2004a). The clay mineralogy is |
| 266 | dominated by smectite but contains progressively more kaolinite above ${\sim}500~{ m mbsf}$ |
| 267 | (Fig. 2; Exon et al., 2001; Robert, 2004). Grey silty claystones give way to a green |
| 268 | glauconitic horizon between 360.1 and 357.3 mbfs (Exon et al., 2001). Above this |
| 269 | greensand, carbonate oozes continue further upwards. The record is quasi- |
| 270 | continuously bioturbated by zoophycos (Exon et al., 2001). We follow the depth |
| 271 | corrections published in (Sluijs et al., 2011) for Cores 12R–31R, based on the |
| 272 | correlation of core-log and downhole log magnetic susceptibility. |
| 273 | |
| 274 | 2.3 Age model |
| 275 | For age tie points (Table S1; Fig. 2), we use the identification of the PETM |
| 276 | (Sluijs et al., 2011) and Middle Eocene Climatic Optimum (MECO; Bijl et al., 2010) and |
| 277 | robust biostratigraphical constraints from the Eocene–Oligocene Boundary interval |
| | |





| 278 | (Houben et al., 2019; Sluijs et al., 2003). In the intervals in between, the age model |
|--------------------------|---|
| 279 | relies on magnetostratigraphy, which in some intervals suffers from a strong normal |
| 280 | overprint (Dallanave et al., 2016), calibrated using nannofossil_diatom–and dinocyst |
| 281 | biostratigraphy (Bijl et al., 2013b; Stickley et al., 2004). Despite the overprint, there |
| 282 | does seem to be a poleomagnetic signal preserved in the inclination data (Fuller and |
| 283 | Touchard, 2004), and biostratigraphic constraints are broadly consistent with nearby |
| 284 | sites (Bijl et al., 2013b; Dallanave et al., 2016). |
| 285 | The nature of the sediments and depositional setting (see below) implies that |
| 286 | small hiatuses may exist between the age tie points (Röhl et al., 2004a). Indeed, |
| 287 | were already identified across the Cretaceous–Paleogene Boundary |
| 288 | (scnellenberg et al., 2004), in the mid-Paleocene (Bijl et al., 2013b; Hollis et al., 2014), |
| 289 | early Eocene (Bijl et al., 2013b) and in the middle Eocene (Röhl et al., 2004a), |
| 290 | corresponding to increases in magnetic susceptibility (Fig. 2). The section across the |
| 291 | Eocene–Oligocene transition is strongly condensed (Houben et al., 2019; Stickley et |
| 292 | al., 2004a, b). |
| 293 | |
| 294 | 2.4 Depositional setting |
| 295 | From the lithology (Robert, 2004) and palynological content (Brinkhuis et al., |
| 296 | 2003), the Maastrichtian–Eocene part of the record was interpreted to represent a |
| 297 | shallow-marine, mid-continental shelf depositional setting, with gradual deepening in |
| 298 | the middle Eocene based on an increase in calcium carbonate content (Fig. 2; Röhl et |
| 299 | al., 2004a). The late Eocene greensands have been initially interpreted as evidencing |
| 300 | strong deepening and current inception as a result of widening of the TG (Stickley et |
| 301 | |
| 202 | al., 2004b). Later studies have related the greensands to invigorated ocean circulation |
| 302 | al., 2004b). Later studies have related the greensands to invigorated ocean circulation and winnowing (Houben et al., 2019), and not necessarily deepening of the site . |
| | |
| 302 203 = 304 | and winnowing (Houben et al., 2019), and not necessarily deepening of the site . |
| 202 | and winnowing (Houben et al., 2019), and not necessarily deepening of the site . Volcanic activity of the Balleny plume in the late Eocene (Hill and Moore, 2001) might |
| 203 = 304 | and winnowing (Houben et al., 2019), and not necessarily deepening of the site . Volcanic activity of the Balleny plume in the late Eocene (Hill and Moore, 2001) might have played a profound role in the bathymetric changes of the ETP, in addition to the |
| 203 = 304 305 | and winnowing (Houben et al., 2019), and not necessarily deepening of the site . Volcanic activity of the Balleny plume in the late Eocene (Hill and Moore, 2001) might have played a profound role in the bathymetric changes of the ETP, in addition to the tectonic stresses that act on diverging plates in the TG area. |
| 202 304 305 306 | and winnowing (Houben et al., 2019), and not necessarily deepening of the site . Volcanic activity of the Balleny plume in the late Eocene (Hill and Moore, 2001) might have played a profound role in the bathymetric changes of the ETP, in addition to the tectonic stresses that act on diverging plates in the TG area. The regional tectonic evolution has implications for interpreting the marine |





| 210 | western boundary current of the proto-Ross gyre, bathed the plateau with Antarctic- |
|---------|--|
|) 11 | derived surface waters (Bijl et al., 2011; 2013b; Huber et al., 2004; Sijp et al., 2014; |
| 312 | 2016). Palynological evidence confirms that the Proto-Ross Gyre influence persisted |
| 313 | at the ETP until the late Eocene (Bijl et al., 2011; Warnaar et al., 2009). This means |
| 314 | that despite northward tectonic drift, the same strong western boundary current |
| 315 | bathed the site during the Maastrichtian to early Eocene (Sijp et al., 2016), with |
| 316 | perhaps some intermittent influence of East Australian Current waters from the north |
| 317 | (Bijl et al., 2010; Cramwinckel et al., 2020). This ended when the proto-Leeuwin |
| 318 | Current started to flow through the progressively widening TG (Fig. 1), bringing the |
| 319 | ETP under the influence of more northerly sourced surface waters (Houben et al., |
| 320 | 2019; Stickley et al., 2004b). |
| 321 | The source area for the terrestrial organic matter (OM) and detrital input was |
| 322 | likely Tasmania. Persistent terrigenous input (Brinkhuis et al., 2003) arguably |
| 323 | requires a large terrestrial catchment area, and the ETP seems too small (~50,000 |
| 324 | km²) to have had vast areas above sea level. Moreover, Paleocene–Eocene terrestrial |
| 325 | palynomorph assemblages contain common Permian–Triassic elements (Contreras et |
| 326 | al., 2014); the Permian–Triassic upper Parmeener group contains thick terrestrial |
| 327 | (coal) deposits and comprises the surface lithology of most of eastern Tasmania |
| 328 | today. Although that same formation might be present in the ETP subsurface as well, |
| 329 | it was probably covered with sediment throughout the Cenozoic (Hill and Moore, |
| 330 | 2001). Rivers flowing from southeast Australia drained into the Gippsland and Bass |
| 331 | Basins, and that terrigenous material is unlikely to have reached the ETP. Seismic |
| 332 | information from the East Tasman Saddle, connecting the Tasmanian Margin to the |
| 333 | ETP, does not suggest there was a deep basin in between (Hill and Exon, 2004). |
| 334 | Therefore, Tasmanian-sourced detrital material could reach the ETP. The ETP was |
| 335 | close enough to the Antarctic margin during the Maastrichtian–early Eocene to have |
| 336 | received perhaps a minor component of Antarctic-sourced terrestrial OM input, in |
| 337 | addition to the dominant Tasmanian source. The regional palynology (Macphail, |
| 338 | 2000; 2002; Carpenter et al., 2012; Pross et al., 2012; Contreras et al., 2013; 2014; |
| 339 | Truswell, 1997), the abundance of peatlands (Holdgate et al., 2009), and the felsic |
| 340 | lithology (Moore, Betts, and Hall, 2013) suggests the hinterland catchment contained |
| 341 | acidic, wet soils and peats. |
| | |





| 342 | |
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| 343 | 2.5 Samples |
| 344 | For this study, we used lipid extracts that have been analysed for GDGTs |
| 345 | previously (Bijl et al., 2009; Hollis et al., 2014; Houben et al., 2019; Sluijs et al., 2011). |
| 346 | We augmented these with 69 new extracts of sediments from the Maastrichtian and |
| 347 | the Paleocene, to extend and improve the temporal resolution of the record. |
| 348 | Unfortunately, not all archived samples from the PETM interval (Sluijs et al., 2011) |
| 349 | could be located, and the interval was reanalysed in a lower resolution (6 samples |
| 350 | over the PETM interval). For the remaining PETM, we used the published peak areas |
| 351 | (Sluijs et al., 2011) to calculate TEX $_{86}$, which is warranted given the new analytical |
| 352 | technique does not affect isoGDGT peak area ratios (Hopmans et al., 2016). For |
| 353 | palynology, we collated and revisited data presented in (Bijl et al., 2010; 2013b; |
| 354 | Brinkhuis et al., 2003; Houben et al., 2019; Sluijs et al., 2011) and generated higher- |
| 355 | resolution data for the Maastrichtian and Paleocene. |
| 356 | |
| 357 | 3. Methods |
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| 358 359 | 3.1 Organic geochemistry |
| | 3.1 Organic geochemistry 3.1.1 Extraction, column separation and analysis |
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| 359 360 | 3.1.1 Extraction, column separation and analysis |
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- 374 Since the discovery of isoGDGTs as proxy for SST (see Schouten et al. (2013)
- 375 for a review), several non-SST effects have been identified that may affect the
- 376 distribution of isoGDGTs in the sediment. Several indices have been developed to
- 377 identify most of the known sources of overprints (Table 1). Next to signalling SST-
- 378 unrelated influences on the isoGDGT pool, these indices also provide information on
- 379 the preverse g marine and paleoenvironmental and depositional conditions.
- 380
- 381 Table 1: Indices from iso- and brGDGTs and brGMGTs. For the chemical structure of
- these components see Fig. 3.

| Index | E are attace | D 6 | Cut-off | Gamma |
|-------------------------|---|---------------------|-----------------------|----------------|
| name | Equation | Proxy for | value | Source |
| TEX ₈₆ | GDGT2 + GDGT3 + Cren' | Sea surface | | (Schouten et |
| 1 LA86 | GDGT1 + GDGT2 + GDGT3 + Cren' | temperature | - | al., 2002) |
| BIT index | IIIa + IIIa' + IIa + IIa' + Ia | Terrestrial input | >0.4? Site- | (Hopmans |
| DITINUEX | Cren + IIIa + IIIa' + IIa + IIa' + Ia | rerrestriarinput | dependent | et al., 2004) |
| | 04 (50000) | Non-thermal | | (O'Brien et |
| fcren' | $\frac{\%Cren`}{\%Cren`+\%Cren}$ | contribution of | 0.25 | al., 2017) |
| | 90CTER + 90CTER | crenarchaeol isomer | | |
| | | Contribution by | | (Zhang et |
| Methane | GDGT1 + GDGT2 + GDGT3 | methane- | >0.3 | al., 2011) |
| Index | GDGT1 + GDGT2 + GDGT3 + Cren + Cren' | metabolising | 20.5 | |
| | | archaea | | |
| | | Contribution by | | (Weijers et |
| AOM ratio | GDGT2/Cren | anaeropbic methane | >0.2 | al., 2011) |
| | | oxydizers | | |
| GDGT2/3 | | Contribution by | | (Taylor et |
| ratio | GDGT2/GDGT3 | deep-dwelling | >5 | al., 2013) |
| Tatio | | archaea | | |
| Methano- | | Contribution by | | (Blaga et al., |
| genesis | GDGT0/Cren | methanogenic | >2.0 | 2009) |
| genesis | | archaea | | |
| Ring index | 0*%GDGT0 + 1*%GDGT1 + 2*%GDGT2 + | Non-pelagic GDGT | ∆RI >0.3* | (Zhang et |
| (RI) | 3*%GDGT3 + 4*%Cren + 4*%Cren' | composition | ∆KI >0.3 [×] | al., 2016) |
| MBT'5me | IIIa + IIIb + IIIc | Mean annual air | | (De Jonge et |
| MDI 5me | IIIa + IIIb + IIIc + IIa + IIb + IIc + Ia | temperature | - | al., 2014a) |
| CBT' | IIIb + IIb' + IIc' + Ia' | (soil-)pH | - | (De Jonge et |
| | IIIa + IIa + Ia | Com-Thu | | al., 2014a) |
| #rings _{tetra} | $\frac{lb+2*lc}{la+lb+lc}$ | | >0.7 | |
| | Ia + Ib + Ic | | | |



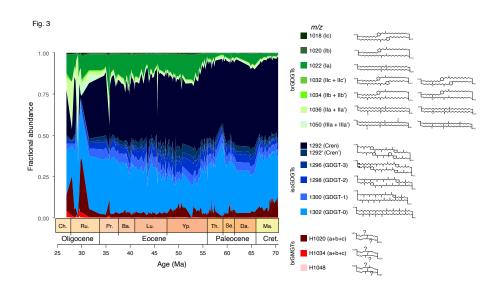


| | IIb + IIb' + 2 * IIc + 2 * IIc' | | | (Sinninghe |
|---------------------------|--|-------------------------------------|---------------------------------|--|
| #rings _{penta} | $\frac{1}{11a + 11a' + 11b + 11b' + 11c + 11c'}$ | | - | Damsté. |
| #rings _{penta5} | $\frac{11b + 2 * 11c}{11a + 11b + 11c}$ | Marine in situ brGDGT production | - | 2016) |
| #rings _{penta6} | $\frac{IIb' + 2 * IIc'}{IIa' + IIb' + IIc'}$ | · | - | |
| IR | $\frac{IIa' + IIb' + IIc' + Ia'}{IIa + IIa' + IIb + IIb' + IIc + IIc' + Ia + Ia'}$ | River in situ brGDGT | Depends on soil value | (De Jonge et al., 2014b) |
| IR _{hexa} | $\frac{Ia'}{Ia+Ia'}$ | production | | (Sinninghe Damsté, |
| IR _{penta} | $\frac{IIa' + IIb' + IIc'}{IIa + IIa' + IIb + IIb' + IIc + IIc'}$ | | - | 2016) |
| brGMGTI | H1020c + H1034a + H1034c H1020b + H1020c + H1034a + H1034b + H1 | | In lakes | (Baxter et al., 2019) |
| %brGMGT, %brGMGTa | $\frac{[brGMGT]}{[brGMGT] + [brGDGT]} \times 100$ | | In peats, | (Baxter et al., 2019) |
| cyclic | $\frac{[brGMGT]}{[Ia + IIa + IIIa + brGMGT]} \times 100$ | Temperature, | lakes | (Naafs et al., 2018a) |
| HMBT _{acyclic} , | H1020a + H1020b + H1 H1020a + H1020b + H1020c + H1034a + H | oxygenation? | In peats | (Naafs et al., 2018a) |
| HMBTacyclic | $\frac{H1020c}{H1020c + 1034b + H1048}$ | | In Arctic Ocean sediments | Sluijs et al., 2020, sensu Naafs et al., 2018a) |



* Cutoff depends on TEX₈₆ value. $\Delta RI = RI_{TEX} - RI$, where $RI_{TEX} = -0.77^*TEX_{86} + 3.32^*TEX_{86}^2 + 1.59$

384







386 Figure 3. Fractional abundances of the various branched (green) and isoprenoidal 387 (blue) GDGTs, and branched GMGTs (red) at ODP Site 1172. 388 389 390 All of the ratios that signal biases express the influence of this bias relative to 391 modern "normal" or idealized compositions. The abundance of overprinted 392 components is divided over either a total sum of overprinted and non-overprinted 393 components (closed sum; e.g., methane index, IR) or over only the non-overprinted 394 (ratio; GDGT2/3 ratio, AOM ratio). The complication is that a change in index value 395 can be caused by changes in the denominator or numerator. This is particularly 396 evident in the interpretation of the branched and isoprenoid tetraether (BIT) index. It 397 was proposed that the closed sum ratio between brGDGTs, which were thought to 398 derive exclusively from soils, and the isoGDGT crenarchaeol, which was thought to be 399 produced exclusively by marine archaea, could be an indicator for the relative 400 contribution of soil organic matter (OM) into marine sediments (Hopmans et al., <u>401</u> 2004). However, it was subsequently shown that crenarchaeol, but critically also 402 other isoGDGTs are produced on land (Weijers et al. (2006), and that a large 403 contribution of soil-OM to marine sediments can cause a bias in TEX_{86} . Weijers et al. 404 (2006) used an end-member model to calculate the potential bias in TEX₈₆-based SST 405 reconstructions in the Congo Fan and this shows significant bias (> 2° C) for BIT above 406 0.3. However, it is rather arbitrary to assign a cut-off for BIT based on the magnitude 407 of the SST bias alone, as this bias depends primarily on the difference between the 408 isoGDGT composition of the soil and marine endmembers. As a consequence, there is 409 no uniform cut-off value for BIT index above which TEX₈₆ should be discarded. 410 Secondly, because the BIT index is a closed sum, the index is equally affected by 411 increased production of crenarchaeol as it is by the input of brGDGTs. Although this 412 could be verified with absolute concentrations of GDGTs, these are not always 413 presented. Third, brGDGTs are produced in situ in the marine realm (Dearing 414 Crampton-Flood et al., 2019; Peterse et al., 2009; Sinninghe Damsté, 2016). The 415 corollary of this is that the cut-off value for BIT to infer an overprint in the isoGDGT-416 based SST signal should be assumed to differ per environmental setting, and perhaps 417 also through time if the setting changes majorly (e.g., Sluijs et al., 2020). As a result,





| 418 | cut-offs were set higher (e.g., 0.4; Bijl et al., 2013a) when no relationship between |
|-----|---|
| 419 | TEX_{86} and BIT index values was detected. This approach is also not without |
| 420 | complications, because the abundance and composition of isoGDGTs (including |
| 421 | crenarchaeol) in soil material varies (Weijers et al., 2006; De Jonge et al., 2015; De |
| 422 | Jonge et al., 2016; Naafs et al., 2019; Sluijs et al., 2020). This in turn implies that in |
| 423 | samples with high BIT (Weijers et al., 2006), the terrestrially-derived isoGDGT |
| 424 | contribution will not be uniform, and might become impossible to detect in a $\ensuremath{TEX_{86}}\xspace$ |
| 425 | BIT index cross plot. In some <mark>paleo-environmental</mark> settings that evidently lack high |
| 426 | input of soil-OM, BIT index values are high (Leutert et al., 2020). For reasons given |
| 427 | above, this could either mean very low crenarchaeol production in the marine realm, |
| 428 | or in situ production of brGDGTs. These two scenarios make for a completely |
| 429 | different paleoenvironmental interpretation, and fail to indicate whether the |
| 430 | brGDGTs can be used to reconstruct MAAT of the hinterland or whether isoGDGTs |
| 431 | can be used for reliable paleothermometry. For that, it is important to assess the |
| 432 | sources of brGDGTs, as brGDGTs produced in rivers (De Jonge et al., 2014b; Zell et al., |
| 433 | 2013) or the (coastal) marine environment (Dearing Crampton-Flood et al., 2019; |
| 434 | Peterse et al., 2009; Sinninghe Damsté, 2016) may contribute and thereby bias the |
| 435 | initial soil-derived brGDGT signal, affecting the reliability of MAAT reconstructions. |
| 436 | Fortunately, production of brGDGTs in the marine realm can be recognized based on |
| 437 | the weighed number of rings of the tetramethylated brGDGTs, quantified in the |
| 438 | #rings _{tetra} , where values >0.7 indicate a purely marine source of the brGDGTs |
| 439 | (Sinninghe Damsté, 2016). Similarly, brGDGTs that are produced in rivers are |
| 440 | characterized by a relatively high abundance of the 6-methyl brGDGTs relative to the |
| 441 | 5-methyl brGDGTs, expressed as the Isomerization Ratio (IR; De Jonge et al., 2014b). |
| 442 | For the other <mark>overprinting indices,</mark> cut-off values may not be as uniformly |
| 443 | applicable to all depositional settings either. Leutert et al. (2020) showed that |
| 444 | modern samples exceeding cut-off values for the GDGT2/3 ratio and methanogenesis |
| 445 | index do not have anomalous TEX $_{ m 86}$ index values in the modern core-top dataset |
| 446 | based on the Ring index. This suggests that the TEX_{86} index value is not equally |
| 447 | influenced by non-thermal contributions in all depositional settings. As a result, |
| 448 | evaluating non-thermal contributions on $\ensuremath{\text{TEX}_{86}}$ index values should consider the |
| 449 | depositional and environmental setting along with the cut-off values. |
| | |





| 450 | |
|----------------|--|
| 451 | 3.1.3 Calibrations for TEX $_{86}$ and MBT' $_{5me}$ |
| 452 | An extensive number of calibrations has been proposed to convert TEX ₈₆ index |
| 453 | values to sea surface temperatures. At the heart of the calibration discussion is the |
| 454 | uncertainty whether the TEX $_{86}$ to-SST relationship continues in a linear way beyond |
| 455 | the modern SST range, or in an exponential way (Hollis et al. 2019). The answer to |
| 456 | that is beyond the scope of this paper, and we refer to (Cramwinckel et al., 2018; |
| 457 | Hollis et al., 2019; O'Brien et al., 2017; Tierney et al., 2017) for detailed discussions, |
| 458 | specifically about using TEX$_{86}$ in regions with SSTs warmer than modern. Following |
| 459 | recent recommendations (Hollis et al. 2019), we here apply several calibrations to |
| 460 | convert our TEX $_{86}$ values to SSTs. We apply an exponential calibration (Kim et al., |
| 461 | 2010) for which we acknowledge that this calibration suffers from a regression |
| 462 | dilution bias (Tierney and Tingley, 2015), and two linear calibrations; one on the |
| 463 | warm part (>15°C) of the core-top dataset (O'Brien et al., 2017) and one using |
| 464 | Bayesian statistics (BAYSPAR; Tierney and Tingley, 2015) to convert our TEX $_{86}$ values |
| 465 | to SSTs. BAYSPAR produces a linear calibration based on a subset of the core top data |
| 466 | with similar GDGT assemblages as the measured \ensuremath{TEX}_{86} , with a user-defined tolerance. |
| 467 | The larger the subset of core tops (i.e., when the number of identified core-top |
| 468 | analogues to the sample TEX $_{86}$ values is large), the closer the Bayesian calibration will |
| 407 | approach a global linear regression. For MAAT reconstructions, we use the Deming |
| 470 | regression of the soil-specific calibration of Naafs et al. (2017b; MAAT $_{\text{soil}}$) and the |
| 471 | Bayesian BayMBT $_{0}$ of Dearing Crampton-Flood et al. (2020). The latter follows the |
| 472 | approach of BAYSPAR, but then for $\mathrm{MBT'}_{\mathrm{5me}}$. We will compare the calibrations for |
| 473 | both proxies to middle Eocene $U^{K'}_{37}$ -based SSTs (from Bijl et al., 2010, but using |
| 474 | BAYSPLINE calibration of Tierney and Tingley, 2018) and Paleocene–early Eocene |
| 475 | sporomorph-based MAAT estimates (Contreras et al., 2014) from the same site. |
| 476 | |
| 477 | Table 2. Calibration equations for TEX ₈₆ and MAAT |

477 Table 2. Calibration equations for TEX_{86} and MAAT

| Calibration | Equation | Туре | Proxy for | Source |
|-----------------------------------|---|----------------------|---------------------|--------------------|
| SST _{exp} | 68.4 * log10(TEX ₈₆) + 38.6 | Exponential | Mean annual SST (0– | (Kim et al., 2010) |
| (TEX ₈₆ ^H) | | (regression | 20m) | |
| | | dilution) | | |





| SST _{lin} | 58.8 * TEX ₈₆ - 11.18 | Linear | Mean annual SST (0– | (0'Brien et al., 2017) |
|---------------------|------------------------------------|------------|-----------------------|------------------------|
| | | | 20m) | |
| BAYSPAR | Prior mean = 20, prior std | Bayesian | Mean annual SST (0- | (Tierney and Tingley, |
| | = 10, search tolerance = | linear | 20m) | 2015) |
| | 0.15 | | | |
| MAATsoil | 40.01 * MBT' _{5me} -15.25 | Linear, | Mean annual air | (Naafs et al., 2017b) |
| | | Deming | temperature (for days | |
| | | regression | above freezing) | |
| BayMBT ₀ | Prior mean = 20, prior std | Bayesian | Mean annual air | (Dearing Crampton- |
| | = 15 | linear | temperature | Flood et al., 2020) |

478 479

3.1.4 R-script for data analysis and evaluation

480 To facilitate systematic calculation of GDGT ratios, data analysis, visualization,

481 and evaluation, we constructed a set of R markdowns

482 (<u>https://github.com/bijlpeter83/RGDGT.git</u>) that can be applied to any time- or

483 depth series of isoGDGT, brGDGT and/or brGMGT data. The R script loads peak areas

484 of GDGTs/GMGTs from Microsoft excel spreadsheets, calculates and plots fractional

485 abundances, overprinting indices and paleotemperature time or depth series.

486 487

488

495

3.2 Palynology

3.2.1 Sample processing

489 Palynological sample processing techniques were published in the original

490 papers describing these datasets (Bijl et al., 2010; 2013b; Brinkhuis et al., 2003;

491 Houben et al., 2019; Sluijs et al., 2011). We followed the same procedure for the new

492 Maastrichtian–Paleocene samples. The different publications do indicate differences

493 in the employed sieve mesh sizes (10 or 15 μm) but given all counted dinocysts are

494 larger than 15 μ m, this has not led to differences in the dinocyst results.

3.2.2 Taxonomy

496 We used the taxonomic framework cited in Williams et al., (2017) in our

497 counts down to the species level, with one exception. For the Wetzelielloidae

498 subfamily, we follow Bijl et al. (2017), for reasons stated therein. We also follow the

499 supra-generic classification based on Fensome et al. (1993); genera described post-

500 1993 were added into that classification accordingly.

5013.2.3 Ecological affinities of dinocyst ecogroups

19





- 502 In this paper, we present our results plotted in organized and complexes, 503 which groups dinocyst species with similar ecologic another ties based on modern (for 504 extant taxa) and empirical (for extant and extinct taxa) data (Frieling and Sluijs, 2018; 505 Sluijs et al., 2005). The species of which the ecologic affinity was not assessed 506 previously were included into larger groups based on shared morphological 507 characteristics; primarily tabulation, archaeopyle and cyst- and process shapes. For example, the "Approximation complex" is equated to "Wetzellioids" following Frieling 508 509 et al. (2014; see Table S2 for taxonomic grouping). We directly compare dinocyst- and 510 terrestrial palynomorph indices with GDGT-based indices (following the approach of 511 Frieling and Sluijs, 2018) to arrive at multi-proxy reconstructions of SST, river runoff 512 and distance to shore (Table 3). For this, we resampled and binned GDGT-based 513 indices to the sample resolution of the dinocyst data.
- 514
- 515 Table 3. Environmental parameters, and their corresponding GDGT indices and
- 516 dinocyst eco-groups, based on Sluijs et al. (2005), Frieling and Sluijs (2018) and Sluijs
- 517 and Brinkhuis (2009).

| Environmental | GDGT index | Dinocyst eco-group |
|----------------------|-------------------|---|
| parameter | | |
| Sea surface | TEX ₈₆ | %Thermophiles (Wetzellioideae, |
| temperature | | Hafniasphaera spp., Florentinia reichartii, |
| | | Polysphaeridium spp, Homotryblium spp., |
| | | Heteraulacacysta spp., Eocladopyxis spp., |
| | | Dinopterygium spp.) |
| Runoff, fresh water, | IR, BIT | %Senegalinium cpx, %Phthanoperidinium |
| salinity | | spp., %Terrestrial palynomorphs |
| Distance to shore | BIT, | %Open marine (<i>Apectodinium</i> spp., |
| | #ringstetra | Impagidinium spp., Operculodinium spp., |
| | | Spiniferites spp.), %Glaphyrocysta cpx., |
| | | %Epicystal Goniodomids |
| | | (Polysphaeridium spp, Homotryblium spp., |
| | | Heteraulacacysta spp., Eocladopyxis spp., |





Dinopterygium spp.), %Terrestrial palynomorphs 518 519 520 3.2.4 Diversity and variability indices 521 To gain insight into the diversity and variability of the dinocyst assemblage 522 through time, we employ several indices (Table 4), and compare their results at 523 species and ecogroup level. We note that diversity in dinocyst taxonomy is 524 complicated since dinocysts are dominantly produced during the hypnozygotic phase 525 of the dinoflagellate life cycle following sexual reproduction. Only ~15% of modern 526 dinoflagellates features this cyst stage (Fensome et al., 1993), which leaves a large 527 proportion of the biological group unrepresented in the fossil record, including for 528 example coral and foraminifer symbionts, but also more closely related free-**F 2 0** swimming taxa. Dinocyst diversity can therefore only be used as an indicator for dinocyst-producing dinoflagellates. As a further complication, taxonomic divisions of ວ່ວປ 531 cysts, although strongly related to dinoflagellate morphology and taxonomy, is fully 532 based on the morphology of dinocysts. Even modern dinoflagellate-dinocyst 533 relationships are often not unambiguous, Some dinoflagellate species produce 534 various cyst morphologies (Rochon et al., 2008) that represent multiple cyst genera 535 or species. Some of these cyst morphological variations are the result of ecology 536 (Mertens et al., 2011). This complicates comparing cyst datasets on the species level 537 and affects assessing biological diversity based on cysts. On the genus level, 538 taxonomic division of cysts is in most cases based on plate tabulation, which is the 539 morphologic feature that has the closest relationship to dinoflagellate biological 540 diversity (Fensome et al., 1993). However, ecologic or biogeographic affinities have 541 been established for some dinocysts on a species level (e.g., Frieling and Sluijs, 2018), 542 which does suggest subtle morphological features may have biologic and ecologic 543 significance. The eco-groups we use here are the result of extensive reviews of 544 empirical data. These groups combine dinocyst genera and species with 545 fundamentally similar plate tabulations, and thus probably group cysts of closely 546 affiliated biological dinoflagellate species, and, as such also ecologic affinities. For our





- 547 diversity calculations we use both the ungrouped data on species level and the
- 548 diversity in dinocyst ecogroups.
- **F** 40 As the simplest approximation of biological diversity, the richness R in terms of dinocyst taxa was summed. Furthermore, several diversity measures were ว่อป 551 calculated using the R package Vegan (Oksanen et al., 2015). Of these, Fisher's alpha 552 (α) is based on the count data, whereas the Shannon index (H') and Simpson index 553 (D) derive from the relative abundance data. Finally, we employed the Σcv metric 554 (Gibbs et al., 2012), which we here dub the "Gibbs index", and which provides a 555 measure for assemblage variability. Together, these metrics can give insight into 556 changing stability and diversity of these regional dinocyst assemblages over the 557 Maastrichtian to early Oligocene. 558 559 Table 4. Diversity and variability indices for (fossil) assemblages. In the Shannon and 560 Simpson indices, p_i represents the proportional abundance of the *i*th taxon of the total
- 561 amount of taxa R. The Gibbs index summates the coefficients of variation (SD/mean)
- of all taxa (*i* to *R*) over a certain rolling window $t_1 \rightarrow t_2$. In Fisher's alpha, the α
- 563 parameter is estimated from the dataset in which *S* is the expected number of species
- with an abundance of *n*. *x* represents a nuisance parameter estimated from the
- 565 dataset, generally between 0.9 and 1. While the Gibbs index is calculated over a
- 566 rolling window, the other indices are calculated per sample.

| Index | Equation | Source |
|--------------------|---|-----------------------|
| Shannon index (H') | $H' = \sum_{i=1}^{R} p_i * \ln p_i$ | (Shannon, 1948) |
| Simpson index (D) | $D = \sum_{i=1}^{R} p_i^2$ | (Simpson, 1949) |
| Gibbs index (Σcv) | $\Sigma cv = \sum_{i=1}^{R} \left(\frac{SD_{i,t_1 \to t_2}}{mean_{i,t_1 \to t_2}} \right)$ | (Gibbs et al., 2012) |
| Fisher's alpha (α) | $S_n = \frac{\alpha x^n}{n}$ | (Fisher et al., 1943) |

567

568

3.3 Comparison of GDGT and dinocyst assemblage data



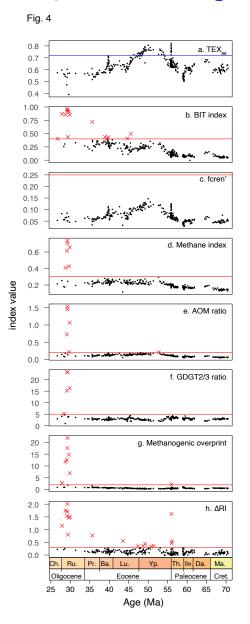


| 569 | Both dinocyst ecogroups and GDGT indices bear information on SST, runoff |
|-----|--|
| 570 | (\sim salinity), nutrients and marine primary productivity, and relative distance to shore |
| 571 | (Table 4). As dinocyst and GDGT analyses were performed on partly separate |
| 572 | datasets, the highest resolution dataset (dinocysts) was linearly resampled to the |
| 573 | depth intervals of the GDGT data to facilitate inter-comparison. |
| 574 | |
| 575 | 4. Results |
| 576 | |
| 577 | 4.1 GDGTs |
| 578 | The resulting dataset has an average temporal resolution of ${\sim}130$ kyrs for the |
| 579 | time interval between 70 and 30 Ma. IsoGDGTs dominate the pool of GDGTs |
| 580 | (particularly GDGT-0 and crenarchaeol), with a gradual increasing relative abundance |
| 581 | of brGDGTs (particularly Ia) throughout the record (Fig. 3). BrGMGTs are in low |
| 582 | (<10%) relative abundance, except in the Oligocene, where they account for up to |
| 583 | 30% of the total GDGT/GMGT pool. |
| 584 | 4.1.1 Isoprenoidal GDGTs |
| 585 | Before the TEX $_{86}$ index results (Fig. 4a) can be interpreted in terms of SST, we |
| 586 | assess whether the isoGDGTs are primarily derived from the sea surface, or have |
| 587 | received contributions from non-pelagic sources. Across the Ypresian, the BIT index |
| 588 | (Fig. 4b) increases from values around 0.1 in the underlying sediments to values at |
| 589 | times exceeding 0.4. Most EECO and middle Eocene values, however, have BIT index |
| 590 | values between 0.2 and 0.35. The remaining sediments have BIT index values of |
| 591 | around 0.3, with the exception of some upper Eocene and Oligocene sediments that |
| 592 | occasionally have values around 0.75. There is no correlation between \mbox{TEX}_{86} and BIT |
| 593 | in our entire dataset (2.15). None of the samples have fcren' (Fig. 4c) values above |
| 594 | the cut-off of 0.25, gesting no non-temperature related contribution of the |
| 595 | crenachaeol isomer to the isoGDGTs. In fact, the consistent trend in (fcren') and the |
| 596 | TEX ₈₆ index (Fig. 4) (and TEX ₈₆ ^L , not shown) demonstrates the temperature |
| 597 | sensitivity of cren' and confirms that discarding it from the index as was done for the |
| 598 | cold-temperature index $\mathrm{TEX}_{86}{}^{\mathrm{L}}$ (Kim et al., 2010) is not justified here. With the |
| 599 | exception of 6 samples in the Oligocene, all samples have methane index values below |
| 600 | the conservative cut-off value of 0.3 (Fig. 4d), although in the early and middle |





- 601 Eocene, methane index values do approach the cut-off. AOM ratio values suggest an
- 602 overprint from anaerobic methane oxidisers in one sample in the early Eocene that
- also has a high BIT index, and 5 samples from the Oligocene where isoGDGTs
- 604 supposedly received a contribution of anaerobic methane oxidisers (Fig. 4e). As a
- 605 result, these samples were discarded for TEX₈₆



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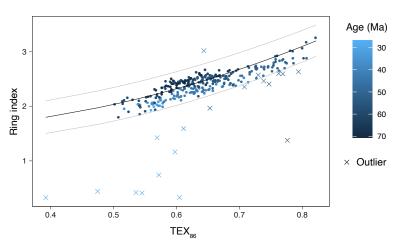


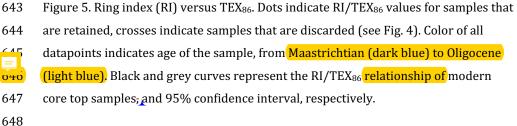
| 607 | Figure 4. TEX $_{\rm 86}$ index and overprinting indices. For the equations of these indices see |
|---------------|--|
| <u>(18</u> | Table 1. Red line indicates the cut-off value, red crosses indicate samples exceed the |
| ۲09 = • | cutoffa. TEX ₈₆ (Schouten et al., 2002), with blue line indicating the maximum modern |
| 610 | core-top value (~0.72). b. BIT index (Hopmans et al., 2004). c. fcren' (O'Brien et al., |
| 611 | 2017), no samples discarded; d. Methane index (Zhang et al., 2011), $n_{discarded}$ = 6. e. |
| 612 | AOM ratio (Weijers et al., 2011), n _{discarded} = 6. f. GDGT2/3 (Taylor et al., 2013), |
| 613 | conservative cut-off of 5, $n_{discarded}$ = 4, one sample retained for absence of anomalous |
| 614 | Ring index values (see text). g. Methanogenesis (Blaga et al., 2009), cut-off of 2.0, |
| 615 | $n_{discarded} = 8. h. \Delta RI n_{discarded} = 18.$ |
| 616 | |
| 617 | |
| 618 | |
| 619 | Values signalling a deeper water column overprint (GDGT2/3 ratio) fall below |
| 620 | 5 for most of the data (Fig. 4f), which was expected given the depositional setting on a |
| 621 | continental shelf. A total of 5 samples has water GDGT2/3 values exceeding 5, 4 of |
| 622 | which also had other indications of overprinted isoGDGT signals. We discard eight |
| 623 | TEX_{86} datapoints because GDGT-0/crenarchaeol ratios are over 2 (Fig. 4g). One of |
| 624 | these is from the PETM, with a value just above the cut-off. The others represent |
| 625 | Oligocene samples. |
| 626 | In total, 18 samples fall outside the range of RI values that characterize |
| 627 | modern pelagic-derived isoGDGT compositions (ΔRI >0.3; Fig. 4h, or 95% confidence |
| 628 | interval; Fig. 5). Nine of these 18 come from the interval >43 Ma and have no other |
| 629 | indications of overprints. These samples fall just outside the 95% confidence interval |
| 630 | limits (Fig. 5) and do follow the overall RI/TEX $_{86}$ trend, but we here discard them to |
| 631 | be conservative. The other 9 samples with too high ΔRI come from the late Eocene- |
| 632 | Oligocene and have BIT index values over 0.4. Two of these samples have no other |
| 633 | overprint indication, the other 7 have multiple other overprints. One sample has RI |
| 634 | values above the range of modern samples (with no other overprinting indications), |
| 635 | the others have too low RI values. Eight samples with normal RI values do have |
| 636 | overprints based on the other indices: BIT index values slightly over 0.4 (6 samples), |
| 637 | GDGT2/3 ratio values over 5 (1 sample), and AOM ratio over 0.2 (1 sample). |
| 638 | Following Leutert et al. (2020) we retained the samples with normal RI values and |





- high BIT or GDGT2/3 ratio values. After considering all these potential biases, and
- retaining those with normal isoGDGT distributions, we discard a total of 19 samples.
- 641 Most discarded samples are from the Oligocene and meet multiple overprint criteria. Fig.5





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4.1.2 Branched GDGTs

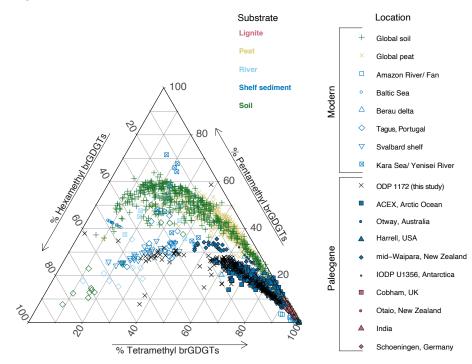
Before brGDGT distributions can be interpreted in terms of air temperature, 651 652 we assess whether they are primarily derived from soils, or have received 653 contributions from river-, or marine in situ production. The relative abundance of 654 tetra-, penta- and hexamethylated brGDGTs in the samples from 1172 deviates from 655 that in modern and Paleogene soils and peats (Fig. 6): it has either comparatively less 656 tetra-, less penta-, or more hexamethylated brGDGTs, and the data split into 2 657 clusters. The largest cluster follows the distribution observed in modern soils and 658 peats with an offset (Fig. 6). This generally indicates brGDGT contributions from 659 sources other than soils. The smaller second cluster, containing exclusively mid-to-660 late Paleocene samples, is indeed close to that of modern continental shelf sediments





661 (Svalbard and Berau delta; Dearing Crampton-Flood et al., 2019; Sinninghe Damsté, 662 2016) for which in situ brGDGT production substantially exceeds soil contributions. 663 This would generally suggest that the smaller cluster, and perhaps also the larger 664 cluster of samples, have contributions from marine in-situ brGDGT production. 665 However, unlike those modern marine sediments, our entire record does not show 666 elevated #ringstetra values that are associated to marine in situ-produced brGDGTs 667 (Fig. 7). The low #ringstetra values on our record (<0.3) suggest brGDGTs have a 668 terrestrial source. 669





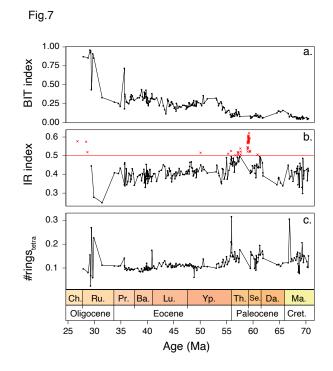


671 Figure 6, Ternary diagram showing fractional abundance of tetra-, penta- and
672 nexamethylated brGDGTs from modern soils (Dearing Crampton-Flood et al., 2020),
673 peats (Naafs et al., 2017b), rivers (Tagus River, Portugal (Warden et al., 2016) and
674 Yenisei River (De Jonge et al., 2015)) and marine sediments (Kara Sea (De Jonge et al.,
675 2015), Svalbard (Dearing Crampton-Flood et al., 2019), Berau delta (Sinninghe
676 Damsté, 2016), Portugal (Warden et al., 2016), Baltic Sea (Warden et al., 2018), and





- 677 Amazon Fan (Zell et al., 2014)), Paleogene lignites (Cobham, Schoeningen, India and
- Otaio (Naafs et al., 2018b)) and Paleogene marine sediments (Arctic Ocean, IODP
- 679 Arctic Coring Expedition ACEX (Willard et al., 2019), Otway Basin, Australia
- 680 (Huurdeman et al., 2020), Gulf of Mexico coastal plain, Harrel core (Sluijs et al., 2014),
- New Zealand (Pancost et al., 2013) and Antarctic Margin IODP Site U1356 (Bijl et al.,
- 682 2013a)). Data from ODP Site 1172 in black crosses.



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Figure 7. a. BIT index, b. IR (with cut-off value of the Australian soil IR of 0.5 and discarded MAAT samples in red crosses) and c. #rings_{tetra} from Site 1172.

In the modern soil- and peat brGDGT dataset #rings_{penta} and #rings_{tetra} are below 0.7 and follow a coherent 1:1 correlation (Fig. 8a). Values >0.7 suggest brGDGTs cannot come purely from soils (Sinninghe Damsté 2016). All samples from 1172 are below 0.7 for both these indices, which suggests a terrestrial origin is likely, and a strong contribution by marine in situ produced brGDGT is unlikely. We do note that while #rings_{tetra} is below 0.3, #rings_{penta} ranges up to 0.6. This diverts from the

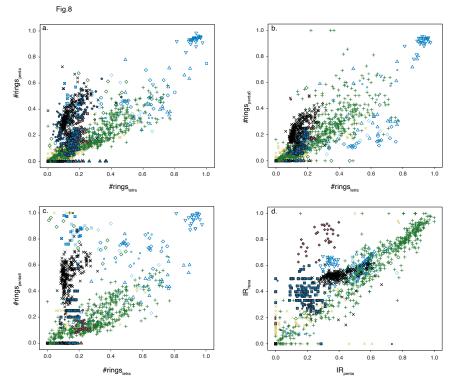




| 694 | 1:1 line but brings the brGDGT dataset from Site 1172 within the area of other |
|-----|--|
| 695 | Paleogene marine brGDGT datasets (Fig. 8a). This is consistent with the relative |
| 696 | abundances of tetra-, penta- and hexamethylated brGDGTs plotted in the ternary |
| 697 | diagram, where all Paleogene data plotted offset to that in modern soils. $\#rings_{tetra}$ |
| 698 | and $\#rings_{penta}$ for the 5- (Fig. 8b) and 6-methyl brGDGTs (Fig. 8c) separately shows |
| 699 | that the diversion of the 1172 data from modern soils and peats is primarily caused |
| 700 | by a larger degree of cyclisation of the 6-methyl brGDGTs, suggesting an influence by |
| 701 | river brGDGT production. |
| 702 | The samples within the Paleocene cluster do have relatively high IR. The IR in |
| 703 | our record averages \sim 0.4, but is elevated between 62 Ma and 54 Ma, with peak values |
| 704 | in the mid-Paleocene up to 0.6 (Fig. 7). Although based on only two datapoints, |
| 705 | modern southeast Australian soils have IR values of maximum 0.5 (De Jonge et al., |
| 706 | 2014a). Hence, any values exceeding that of the corresponding hinterland soils could |
| 707 | indicate a contribution by non-soil sources (be it marine or river in situ production; |
| 708 | De Jonge et al., 2014a). Given that #rings indices values argue against a large |
| 709 | contribution from marine in-situ production, we suggest that river-produced |
| 710 | brGDGTs contribute to the GDGT pool in the Paleocene samples with high IR. |
| 711 | Therefore, for MBT' $_{\rm 5me}$ calculation, we eliminate all brGDGT samples in our record |
| 712 | that have IR values above that of Australian soils. The IR_{penta} and IR_{hexa} are well within |
| 713 | the range of the modern soil and peat data (Fig. 8d) and settings affected by marine or |
| 714 | river in situ production (Svalbard, Kara / Yenisei), and plot far away from lignite |
| 715 | deposits. |
| 716 | |







717

Figure 8. Crossplots of various brGDGT indices. a. #rings_{tetra} vs. #rings_{penta}, #rings_{tetra}
vs. #rings_{penta5}, #rings_{tetra} vs. #rings_{penta6}, IR_{penta} vs. IR_{hexa}. For color and symbol legend
see Figure 6.

721

722 The consistent offset in brGDGT composition in Paleogene marine sediments 723 relative to modern soils can potentially be explained by non-soil contributions even 724 though many (most) indices suggest soil-dominated sourcing. Notably, not only 725 Paleogene marine sediments from Site 1172 are offset from present-day soils, but this 726 extends to other sites as well, including Paleogene lignites that are not influenced by 727 marine contributions. We thus argue that there may be an additional evolutionary- or 728 bacterial community factor that is at least in part responsible for the observed offset 729 brGDGT distributions during the Paleogene, and argue that, with some exceptions, 730 where indices do suggest non-soil contributions, Paleogene marine sediments contain 731 a dominant soil-derived brGDGT signal in spite of their offset composition. 732





| 733 | |
|-----|--|
| 734 | 4.2 SST and MAAT reconstructions |
| 735 | 4.2.1 SST |
| 736 | By removing all samples with potential overprint from the record we can now |
| 737 | assess the trends in TEX_86-based SST (Fig. 9a). Reconstructed SSTs differ t_{Θ} up to 4 °C |
| 738 | between the exponential SST_{exp} and the linear SST_{lin} calibration in the warmest |
| 739 | intervals, and progressively less with lower SSTs (Fig. 9a). The BAYSPAR method, |
| 740 | which uses a linear relation between SST and <mark>TEX86</mark> g enerates the lowest SSTs for |
| 741 | the low TEX $_{86}$ values in the Maastrichtian, Paleocene and middle–late Eocene, and |
| 742 | SSTs in between those based on the linear and exponential calibrations for the |
| 743 | highest TEX ₈₆ index values (Fig. 9a). $U^{K'}_{37}$ -based SSTs (Bijl et al., 2010, with |
| 744 | BAYSPLINE calibration) around the MECO are most consistent with the high SSTs |
| 745 | $\frac{1}{1000}$ based on the linear TEX _{OBL} calibration, although in this TEX ₈₆ range all calibrations |
| 746 | yield SSTs within error (Fig. 9a). |
| 747 | In general, Maastrichtian 🔁 5–66.7 Ma) SSTs show a gradual cooling trend |
| 748 | towards the late Maastrichtian–early Danian hiatus of ~800 kyrs. The early Danian |
| 749 | (~65 Ma) is roughly 3 °C warmer than the late Maastrichtian (~67 Ma). The mid- |
| 750 | Paleocene (62–59 Ma) shows two cooling steps: a \sim 2 °C cooling at 62 Ma, followed by |
| 751 | a return to higher SSTs roughly at 60 Ma, and a ${\sim}5$ °C cooling to minimum values of |
| 752 | around 18°C around 59.5 Ma, where the record is truncated by a hiatus. |
| 753 | A hiatus straddling the early Thanetian (59.0–57.7 Ma) likely obscures the |
| 754 | onset of late Paleocene warming out of the mid-Paleocene temperature minimum. |
| 755 | Background latest Paleocene SSTs fluctuate considerably, but the PETM warming |
| 756 | clearly stands out (Sluijs et al., 2011), with a magnitude (5-7 °C depending on the |
| 757 | calibration) comparable to that in other mid- to high latitude sections and similar to |
| 758 | the global average (Frieling et al., 2017). Post-PETM SSTs drop back to pre-PETM |
| 759 | values completely, followed by renewed warming towards the EECO. The magnitude |
| 760 | of warming is 6 to 11 °C depending on the choice of calibration. Smaller early Eocene |
| 761 | hyperthermal events , if represented in our record, do not clearly stand out at Site |
| 762 | 1172. |
| 763 | Highest SSTs of the EECO are slightly lower than peak PETM values in all |
| 764 | calibrations, consistent with other records (The lis et al., 2020). However, a hiatus |

31





- between 52.7 and 51.4 Ma may conceal peak EECO temperatures (Hollis et al., 2019;
- 766 Lauretano et al., 2018). A gradual SST decline commences around 49.2 Ma and
- 767 terminates the EECO, which is broadly coeval with cooling inferred from benthic
- for a miniferal isotope records (Lauretano et al., 2018) and proxy records from New
- 769 Zealand (Dallanave et al., 2016; Crouch et al., 2020).
- 770

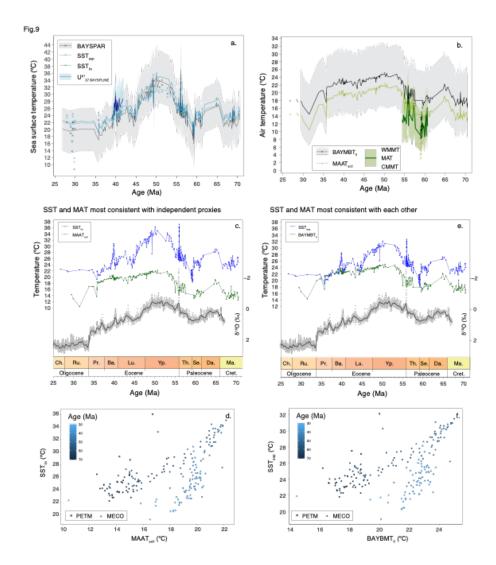




Figure 9. Paleothermometers. a. TEX₈₆ based SST reconstructions, using a Bayesian
(BAYSPAR; black, with 90% confidence interval in grey envelope; Tierney and





| 774 | Tingley, 2015) an exponential (SST $_{exp}$; light blue; Kim et al., 2010) and a linear (SST $_{lin}$; |
|-----|--|
| 775 | dark blue; O'Brien et al., 2017) calibration. Also plotted are $U^{K'}_{37}$ -based SSTs (thick |
| 776 | blue line; from Bijl et al., 2010, recalculated using BAYSPLINE of Tierney and Tingley, |
| 777 | 2018). Discarded samples are indicated by the crosses. b. brGDGT-based MAAT |
| 778 | reconstructions using the Bayesian (BayMBT ₀ ; black, with 90% confidence interval in |
| 779 | grey envelope; Dearing Crampton-Flood et al., 2020) and soil (MAAT _{soil} ; light green; |
| 780 | |
| | Naafs et al., 2017b) calibrations. Discarded samples are indicated in crosses. Also |
| 781 | plotted are pollen-based NLR-approach MAT reconstructions (thick dark green, with |
| 782 | light green envelope representing coldest and warmest month mean temperatures; |
| 783 | from Contreras et al., 2014). c. SST and MAAT reconstructions using the calibrations |
| 784 | that are most consistent with independent proxies from this site (Bijl et al., 2010; |
| 785 | Contreras et al., 2014), with the CENOGRID benthic foraminifer oxygen isotope splice |
| 786 | of Westerhold et al., 2020 (10pnt loess smooth in grey, 500 pnt loess smooth in |
| 787 | black). d. cross-correlation between SST_{lin} and $MAAT_{soil}$. e. SST and <code>MAAT</code> |
| 788 | reconstructions using the calibrations that are, in absolute temperature, most |
| 789 | consistent with each other, with the CENOGRID benthic foraminifer oxygen isotope |
| 790 | splice of Westerhold et al., 2020 (10pnt loess smooth in grey, 500 pnt loess smooth in |
| 791 | black). f. cross-correlation between SST_{exp} and $BAYMBT_0$. |
| 792 | |
| 793 | |
| 794 | Post-EECO cooling is gradual and of small magnitude (2 °C from peak EECO |
| 795 | values) until about 46.4 Ma, after which it accelerates (5 $^{\circ}$ C) between 46.2 Ma and |
| 796 | 44.4 Ma. The age model in this particular part of the sequence is <mark>complicated,</mark> due to a |
| 797 | problematic assignment of chron C21n (Bijl et al., 2010; 2013b). Following the |
| 798 | minimum <mark>SSTS</mark> at 44.4 Ma, SSTs rise again towards a plateau at 42.7 Ma. An SST |
| 799 | minimum of \sim 22 °C is observed just prior to the MECO, at 40.2 Ma, followed by MECO |
| 800 | warming of about 5–7 °C reached at 39.9 Ma. Post-MECO cooling seems more gradual |
| 801 | than MECO warming, although this might be the result of sedimentation rate changes |
| 802 | (Bijl et al., 2010), which are not accounted for in our age model. SSTs are almost |
| 803 | identical to those for the MECO at nearby Site 1170 on the South Tasman Rise |
| 804 | (Cramwinckel et al., 2020). Post-MECO cooling transitions into gradual late Eocene |
| 805 | cooling, down to a minimum of ${\sim}19$ °C at 35.7 Ma. The following latest Eocene to |
| | |

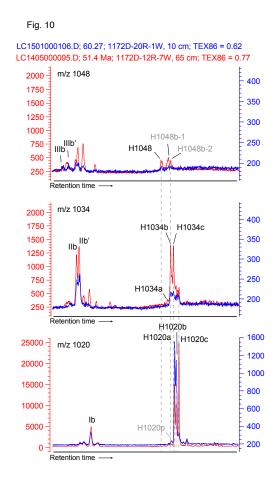




| 806 | Oligocene TEX $_{86}$ -based SSTs (35–27 Ma) are 3 °C warmer, with surprisingly no |
|-----|---|
| 807 | cooling associated to the Eocene–Oligocene transition (Houben et al., 2019). |
| 808 | |
| 809 | 4.2.2 MAAT |
| 810 | After excluding samples with IR values above those of local soils, we present |
| 811 | MBT' $_{5me}$ -based MAAT reconstructions. These differ substantially (up to 5°C) per |
| 812 | calibration chosen (Fig. 9b). The $MAAT_{soil}$ calibration yield consistently cooler MAATs |
| 813 | (2–3°C) compared to the BAYMBT $_{0}$ calibration (Fig. 9b), which may be because the |
| 814 | $MAAT_{soil}$ is calibrated against average temperature of the days above freezing, while |
| 815 | $BAYMBT_0$ is calibrated against mean annual temperature. Both MAAT calibrations fall |
| 816 | between the MAT and the warmest month mean temperature reconstruction of the |
| 817 | sporomorph nearest living relative (NLR)- based temperature reconstruction |
| 818 | (Contreras et al., 2014), and calibration errors largely overlap. |
| 819 | Independent of the calibration chosen, MAAT evolution reveals late |
| 820 | Maastrichtian-early Danian warming of 3°C, gradual 4°C cooling towards the mid- |
| 821 | Paleocene, gradual 8° C warming towards the EECO, with a conspicuous absence of |
| 822 | warming at the PETM. Gradual cooling of $4^\circ C$ starts at 49.5 Ma and continues into the |
| 823 | Late Eocene. MAAT reaches a minimum at 43 Ma, followed by a plateau-phase. There |
| 824 | is little warming that can be associated to the MECO. MAAT shows a conspicuous |
| 825 | transient 3 $^{\circ}\mathrm{C}$ cooling in the Late Eocene, at the onset of glauconite deposition. The |
| 826 | Eocene-Oligocene Transition is characterized by another 5°C MAAT cooling. |
| | |







827 828

Figure 10. Stack of 2 UHPLC-MS chromatograms (1172D-12R-7W, 65cm in red and
1172D-20R-1W, 10cm in blue) of *m/z* 1048, 1034 and 1020 traces, between ~40 and
~60 minutes retention time, showing the pattern of peaks corresponding to brGDGTs
and brGMGTs. Peak labels refer to the molecular structures in Fig. 3.

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834

835 4.3 brGMGTs

We detected brGMGTs throughout the record (see Fig. 10 for typical
chromatograms). The exact molecular structure and position of the C-C bridge is still
unknown but based on visual comparison with chromatograms of brGMGTs in

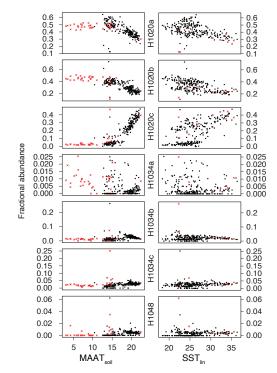
African lake sediments (Baxter et al., 2019), all 7 known brGMGTs can be identified.





- 840 Next to H1020a, b, and c as most abundant brGMGTs, also H1034a, b and c are
- 841 detected, although in some samples in the Maastrichtian, individual peaks could not
- be separated. Traces of H1048 can be seen at times, suggesting presence of this
- compound as well, albeit in low relative abundance. Next to these previously
- 844 described peaks, we here recognized several other peaks that consistently occur at
- 845 Site 1172: one compound that elutes just prior to H1020a, which we term H1020p
- 846 (Fig. 10). ~1 minute after H1048 elutes, two more peaks appear which we here term
- 847 H1048b-1 and H1048b-2 (Fig. 10). Because these have so far unknown affinities or
- 848 molecular structure, we do not include them here further in our calculations.





849

850 Figure 11. Fractional abundances of brGMGTs plotted against BAYMBT₀ and SST_{exp}.

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852

853The fractional abundances of the H1020 isomers show qualitatively similar854relations to temperature as in the East African lake dataset of Baxter et al. (2019):

855 H1020b abundance has a negative relationship with MAAT, while H1020c has a

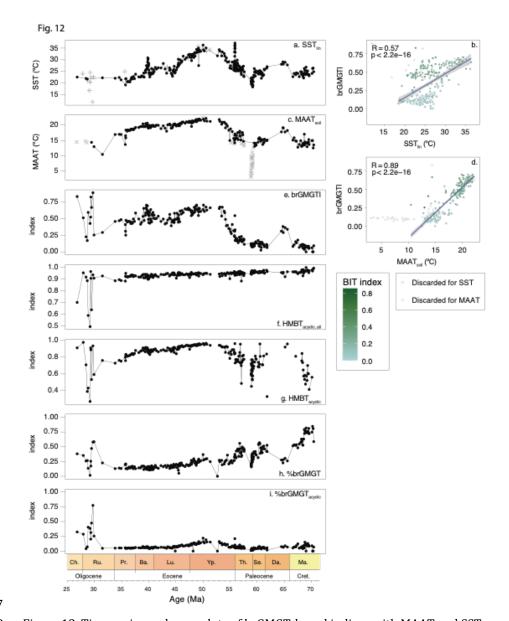




| 856 | positive relationship (Fig. 11). In addition, H1020a seems negatively correlated with |
|-----|--|
| 857 | MAAT in our data, although this compound does not show any relation to |
| 858 | temperature in lakes (Baxter et al., 2019). The relative abundances of the H1034 |
| 859 | isomers do not show a clear relationship with MAAT (Fig. 11), similar as in the |
| 860 | tropical lake dataset, where the scatter increases at lower (H1034b) or higher |
| 861 | (H1034a, c) temperatures (Baxter et al., 2019). |
| 862 | Downcore trends in brGMGTI are primarily controlled by the relative |
| 863 | abundance of H1020c and closely follow those in both SST and MAAT. Upon cross- |
| 864 | correlation, we note that brGMGTI has a higher correlation with MAAT (R = 0.89) |
| 865 | than with SST (R=0.57; Fig. 12). Interestingly, the correlations between $brGMGTI$ and |
| 866 | temperature indices only hold for part of the dataset. Samples with high IR values, for |
| 867 | which brGDGT-based MAAT could not be reliably interpreted, fall outside the |
| 868 | correlation (grey crosses in Fig. 12). This is probably due to the supposed |
| 869 | contribution of river-derived brGDGTs that may bias MAAT towards lower |
| 870 | temperatures. In addition, we also note that brGMGTI approaches 'saturation' |
| 871 | (brGMGTI = 0) at reconstructed MAAT of 15° C. The correlation with temperature |
| 872 | (MAAT and SST) is divided into 2 clusters. BrGMGTI values for samples from the |
| 873 | younger part of the record, with BIT index <~0.3, seem to correlate better to |
| 874 | temperature than those from the older part of tha record, with BIT index values |
| 875 | > \sim 0.3 (Fig. 12). In the Oligocene, where many isoGDGT signals were evidently |
| 876 | overprinted, the %brGMGTs increases (Fig. 3, 12d), and the brGMGT composition is |
| 877 | characterized by a relatively higher abundance of H1020c, H1034b and H1048 |
| 878 | compared to the rest of the record (Fig. 3; 12). |
| 879 | There is no clear relationship between %brGMGTs and MBT' $_{\mathrm{5me}}$ -based MAAT |
| 880 | (Fig. 12h, i). This slightly differs from the Paleocene-Eocene Arctic Ocean record |
| 881 | (Sluijs et al., 2020), where the brGMGTI has no correlation to temperature, while |
| 882 | %brGMGTs do. This may indicate differences in sources of brGMGTs between the |
| 883 | sites, differences in oceanographic settings, or a non-temperature control on their |
| 884 | distribution. HMBT _{acyclic} does seem to show similar trends to the MBT' _{5me} -based |
| 885 | MAAT (Fig. 12g) , like in the Arctic Ocean (Sluijs et al., 2020). |
| 886 | |







887

Figure 12. Time series and crossplots of brGMGT-based indices with MAAT and SST
reconstructions. (a) SST_{lin} (b) crossplot of SST_{lin} and brGMGTI, (c) MAAT_{soil}, (d)
crossplot of MAAT_{soil} and brGMGTI, (e) brGMGTI, (f) HMBT_{acyclic}, all, (g) HMBT_{acyclic}, (h)

891 %brGMGT (Baxter et al., 2019), (i) %brGMGT_{acyclic} (Naafs et al., 2018). For equations

see Table 1). Colour variable in the crossplots indicate the BIT index value.



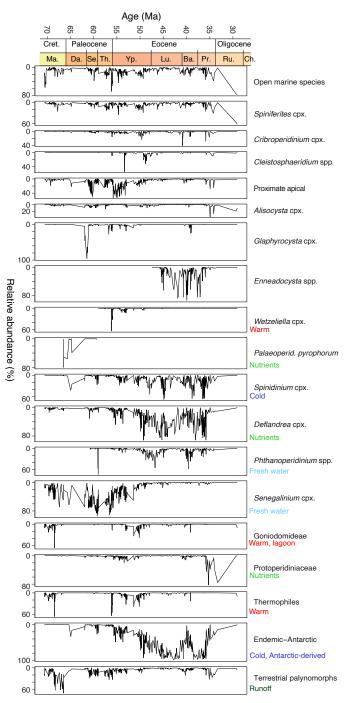


| 894 | 4.4 Palynology |
|------------------|--|
| 895 | 4.4.1 Assemblages |
| 896 | Dinocyst assemblages are dominated by Manumiella spp. throughout the |
| 897 | Maastrichtian, together with Goniodomideae (notably Dinopterygium spp.), and |
| 898 | Cerebrocysta cpx. (Fig. 13). From the late Maastrichtian onwards, Senegalinium cpx. |
| 899 | increases gradually in relative abundance, interrupted by acmes of Palaeoperidinium |
| 900 | <i>pyrophorum</i> in the Danian and <i>Glaphyrocysta</i> cpx. in the lower Selandian. Relative |
| 901 | abundances of <i>Senegalinium</i> cpx. reach maximum values during the mid-Paleocene |
| 902 | and during the Paleocene–Eocene transition, while the PETM itself is characterised by |
| 903 | abundant Apectodinium (~30%) and common Senegalinium cpx. and Goniodomideae |
| 904 | (Sluijs et al., 2011). After the PETM, <i>Senegalinium</i> cpx. abundance s temporarily |
| 905 | increase, and are then me laced by <i>Deflandrea</i> cpx, <i>Spinidinium</i> cpx and <i>Elytrocysta</i> |
| | spp. and low abundances of various other genera, e.g., <i>Wetzeliella, Hystrichokolpoma</i> , |
| <mark>90/</mark> | Goniodomideae (Fig. 13). From 45 Ma to ~37 Ma, <i>Enneadocysta</i> spp. alternates |
| 908 | dominance with <i>Deflandrea</i> cpx. and <i>Spinidinium/Vozzhennikovia</i> cpx, with |
| 909 | contributions of Phthanoperidinium spp. (Röhl et al., 2004a). Protoperidinioid |
| 910 | dinocysts appear commonly in the late Eocene, around 35.5 Ma (Houben et al., 2019; |
| 911 | Sluijs et al., 2003; Fig. 13). |
| 912 | |
| 913 | 4.4.2 Diversity and variability |
| 914 | Together with the changing assemblage composition are some strong and |
| 915 | coherent changes in all diversity and variability indices (Fig. 14). First, results are |
| 916 | highly similar for the full species-level dataset and the grouped assemblages, |
| 917 | indicating robustness of both the grouping and diversity analysis. The Maastrichtian- |
| 918 | Paleocene assemblages are characterized by relatively low diversity, which increases |
| 919 | towards the EECO, as variability indicated by the Gibbs index decreases. After this, |
| 920 | diversity drops and variability increases during middle–late Eocene cooling \sim 49–38 |
| 921 | Ma, with an interruption around 41–39 Ma. Towards the top of the record, both |
| 922 | diversity and variability increase. |
| 923 | |





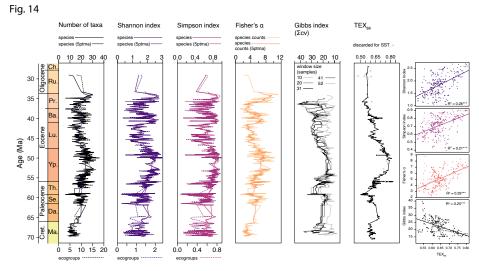
Fig. 13







- 925 Figure 13. Dinocyst ecogroups and %terrestrial palynomorphs from Site 1172. From
- 926 Fent to right, ecogroups are ordered in an in-shore-to-offshore transect. Relative
- 927 abundance ticks represent 20%.
- 928



929

930 Figure 14. Dinocyst taxonomic diversity and variability through time. From left to

931 right are plotted number of taxa (black), Shannon index (purple), Simpson index

932 (pink), Fisher's alpha (orange) (species-based dataset in solid lines; ecogrouped

933 dataset in dashed lines) and Gibbs index (different window sizes in gray-black). TEX₈₆

- 934 plotted for comparison. Crossplots show correlation of these indices with TEX₈₆.
- 935

937

936 **5. Discussion**

5.1 Air and sea surface temperature evolution compared

938The near-shore depositional setting of our record should have captured a939coastal terrestrial environment with similar MAATs similar to the local SSTs. Indeed,940the brGDGT-based MAAT record shows strong similarities to SST particularly in941multi-million year trends (Fig. 9). Specifically, the early Danian is warmer than the942Maastrichtian, and bothe records show a 2-step Paleocene cooling, late Paleocene943warming towards peak values in the EECO, and middle-late Eocene cooling. On944shorter time scales both records, occasionally even on sample-to-sample level, show

945 synchronous variability (Fig. 9c, d). The independent SST (based on alkenones; Bijl et





| 946 | al., 2010) and air temperature reconstructions (based on pollen assemblages; |
|------------------|---|
| 947 | Contreras et al., 2014) show the best fit with the absolute SST values derived from the |
| 948 | linear calibration for TEX $_{86}$ (BAYSPAR or SST $_{ m lin}$; generating the highest SST |
| 949 | estimates), and MAATs based on the calibration using Deeming regression (MAAT $_{ m soil}$), |
| 950 | respectively. These are the two calibrations with the largest absolute temperature |
| 951 | offset, particularly in the warm EECO (Fig. 9c). If the local terrestrial climate in the |
| 952 | nearby river catchment is largely controlled by ocean temperature, the absolute mean |
| 953 | annual SST and MAAT should be broadly similar i n the integrated signal of our |
| 954 | samples . This could be an argument to choose transfer functions for SST and MAAT |
| 955 | whereby absolute temperature estimates overlap most: SST_{exp} and the Bayesian |
| 956 | $BAYMBT_{0_k}$ These generate overlapping absolute temperatures for the colder middle- |
| 957 | late Eocene and Paleocene, but in the warmer Eocene time intervals SSTs remain |
| 958 | consistently warmer (by ~8 °C). The lower MAATs cannot be explained by a |
| 959 | saturation of the MBT' $_{\rm 5me}$ index that is underlying the paleothermometer, as |
| 960 | maximum recorded MBT' _{5me} relaes are around 0.85 in the EECO. Accounting for the |
| 961 | calibration errors of SST (± 2.5 C) and the large calibration error bar of BAYMBT $_0$ in |
| 962 | principle could resolve the offset but applying the extremes in calibration error to |
| 963 | TEX_{86} and MBT' $_{5\text{me}}$ to make them consistent would make both profoundly |
| 964 | inconsistent to the other independent SST and MAAT proxies from the same samples. |
| 965 | More importantly, the colder MAAT compared to SST seems to be a consistent feature |
| 966 | in many regions where TEX $_{86}$ and MBT' $_{\rm 5me}$ were applied together (see e.g., Hollis et |
| 967 | al., 2019 and compare Willard et al., 2019 and Sluijs et al., 2020). The offset between |
| 968 | MAAT and SST would have been more variable between sites if it were only the result |
| 969 | of calibration errors. TEX $_{86}$ -based SSTs in the southwest Pacific realm have been on |
| 970 | the high end of many multi-proxy-based temperature reconstructions for the Eocene |
| 971 | (Hollis et al., 2012), and therefore may be assumed to have an as yet poorly |
| 972 | understood warm bias. However, the consistency of $\ensuremath{TEX_{86}}\xspace$ -based SSTs with other SST |
| 973 | proxies for specific time intervals and locations (Bijl et al., 2010; Hines et al., 2017) |
| 074 | suggests the SST-MAAT offset <mark>cannot be easily reconciled by only invoking a warm</mark> |
| ز ر د | $\overline{\mathrm{bias}}$ in TEX ₈₆ , MAAT reconstructions on the other hand have been broadly consistent |
| 976 | with nearest living relative based temperature reconstructions on pollen assemblages |
| 977 | in many applications in the Eocene (Contreras et al., 2013; Contreras et al., 2014; |





| 978 | Pancost et al., 2013; Pross et al., 2012; Willard et al., 2019), but both brGDGT and |
|--|--|
| 979 | plant-based temperature estimates are arguably prone to cold biases at the high- |
| 980 | temperature end (e.g. Naafs et al. 2018, Van Dijk et al., 2020). In addition, it may be |
| 981 | that the same evolutionary or bacterial community factors that make Paleogene |
| 982 | brGDGT assemblages deviate from that in modern soils, may also cause a deviation in |
| 983 | the calibration to MAAT. This may resolve some of the offset between MAAT and SST |
| 984 | reconstructions. Although this implies that quantitative MAAT estimates based on |
| 985 | $\mathrm{MBT'}_{\mathrm{5me}}$ in non-analogue settings such as the present one should be taken with care, |
| 986 | the strong temperature dependence in Paleogene brGDGTs cannot be ignored. |
| 987 | Another surprising result is that the brGDGT-based MAAT record does not |
| 988 | capture PETM and MECO warming (Fig. 9c), ^{in d} ependent of the calibration chosen. |
| 989 | This is remarkable for several reasons: (1) brod GTs were measured on the same |
| 990 | samples as isoGDGTs, which precludes a sampling bias; (2) SST changes of longer |
| 991 | duration (Maastrichtian–Danian warming, mid-Paleocene cooling and early Eocene |
| 992 | warming towards the EECO) are represented in the MAAT record; (3) The duration of |
| 993 | PETM (\sim 150-250 kyrs; Röhl et al., 2007, Zeebe & Lourens, 2019) and MECO (\sim 400 |
| 994 | kyrs; Bohaty et al, 2009) is too long to explain the absence in MAAT warming with a |
| 995 | lag in soil-derived OM delivery to the ocean (up to several kyr, see, e.g., Feng et al., |
| 996 | 2013; Schefuß et al., 2016; Huurdeman et al., 2020); (4) seems incompatible with the |
| 007 | |
| 997 | fact that other low-amplitude, shorter-term SST changes are reflected in the MAAT |
| 997 998 | fact that other low-amplitude, shorter-term SST changes are reflected in the MAAT record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term |
| | |
| 998 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term |
| 998 999 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term climate variability on the time resolution of the samples; (5) Other PETM records in |
| 998 999 1000 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term climate variability on the time resolution of the samples; (5) Other PETM records in the region do show a temperature response in the PETM in various proxies including |
| 998 999 1000 1001 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term climate variability on the time resolution of the samples; (5) Other PETM records in the region do show a temperature response in the PETM in various proxies including brGDGTs (Hollis et al., 2012; Pancost et al., 2013, Huurdeman et al. 2020). A lack of |
| 998 999 1000 1001 1002 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term climate variability on the time resolution of the samples; (5) Other PETM records in the region do show a temperature response in the PETM in various proxies including brGDGTs (Hollis et al., 2012; Pancost et al., 2013, Huurdeman et al. 2020). A lack of MAAT rise during the PETM and the MECO could be explained by a switch from |
| 998 999 1000 1001 1002 1003 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term climate variability on the time resolution of the samples; (5) Other PETM records in the region do show a temperature response in the PETM in various proxies including brGDGTs (Hollis et al., 2012; Pancost et al., 2013, Huurdeman et al. 2020). A lack of MAAT rise during the PETM and the MECO could be explained by a switch from brGDGT sourcing from soils to (peaty) lakes,t. ich are notoriously cold-biased |
| 998 999 1000 1001 1002 1003 1004 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term climate variability on the time resolution of the samples; (5) Other PETM records in the region do show a temperature response in the PETM in various proxies including brGDGTs (Hollis et al., 2012; Pancost et al., 2013, Huurdeman et al. 2020). A lack of MAAT rise during the PETM and the MECO could be explained by a switch from brGDGT sourcing from soils to (peaty) lakes, which are notoriously cold-biased (Tierney et al., 2010). Paleocene–Eocene peats are abundant in southeast Australia |
| 998 999 1000 1001 1002 1003 1004 1005 | record (Fig. 9c, d), which suggests that soil-derived OM did capture short-term climate variability on the time resolution of the samples; (5) Other PETM records in the region do show a temperature response in the PETM in various proxies including brGDGTs (Hollis et al., 2012; Pancost et al., 2013, Huurdeman et al. 2020). A lack of MAAT rise during the PETM and the MECO could be explained by a switch from brGDGT sourcing from soils to (peaty) lakes, which are notoriously cold-biased (Tierney et al., 2010). Paleocene–Eocene peats are abundant in southeast Australia (Holdgate et al., 2009), and it is possible that a contribution from peat lakes at times |





| 1009 | excluded, is that a dominant fraction of the brGDGT was produced in situ and subdues |
|--|---|
| 1010 | the temperature response during these warming phases. |
| 1011 | The subdued middle-late Eocene cooling in the air temperature record |
| 1012 | compared to the strong decrease in SST is particularly clear when cross-plotting SST |
| 1013 | versus MAAT (Fig. 9c, d): the relationship between the two proxies is different in the |
| 1014 | Maastrichtian–early Eocene compared to the middle–late Eocene. This may be related |
| 1015 | to the start of a pregressive northward tectonic drift of the Tasmanian hinterland, |
| 1016 | which occurred around the same time (Fig. 1). This puts the hinterland of the soil- |
| 1017 | derived brGDGTs into warmer climate zones throughout the middle–late Eocene, |
| 1018 | while the ETP remained under influence of the Antarctic-derived Tasman Current |
| 1019 | (TC) through that time (The ber et al., 2004). The TC cools and likely strengthens in the |
| 1020 | middle and late Eocene. The terrestrial climate cooled as well, but this signal will be |
| 1021 | subdued if the soil material is sourced from a progressively lower-latitude |
| 1022 = 1023 | environment as Australia drifted northward. <mark>Yet, one would expect that the close</mark> |
| 1023 | coupling between land- and seawater temperature in near-shore environments |
| 1024 | would not capture such tectonic effects. |
| | |
| 1025 | The correlation between both temperature proxies changes once more during |
| 1025 1026 | The correlation between both temperature proxies changes once more during the late Eocene -Oligocene, when they show a sharp cooling of 2–3 °C at the onset of |
| | |
| 1026 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of |
| 1026 1027 | the late Eocene–Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at \sim 35.5 Ma. Whereas this is merely a continuation of |
| 1026 1027 1028 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at ~35.5 Ma. Whereas this is merely a continuation of progressive late Eocene cooling in the SST record, the decrease in MAAT is strongly |
| 1026 1027 1028 1029 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at ~35.5 Ma. Whereas this is merely a continuation of progressive late Eocene cooling in the SST record, the decrease in MAAT is strongly accelerated compared to middle–late Eocene trends. Another 3–4 °C MAAT drop |
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| 1026 1027 1028 1029 1030 1031 1032 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at ~35.5 Ma. Whereas this is merely a continuation of progressive late Eocene cooling in the SST record, the decrease in MAAT is strongly accelerated compared to middle–late Eocene trends. Another 3–4 °C MAAT drop occurs sometime between the late Eocene and the early Oligocene (likely associated with the Eocene–Oligocene transition), while SSTs returns to warmer, middle Eocene values in this interval. The continued sea surface warmth across the EOT has been |
| 1027 1028 1029 1030 1031 1032 1033 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at ~35.5 Ma. Whereas this is merely a continuation of progressive late Eocene cooling in the SST record, the decrease in MAAT is strongly accelerated compared to middle–late Eocene trends. Another 3–4 °C MAAT drop occurs sometime between the late Eocene and the early Oligocene (likely associated with the Eocene–Oligocene transition), while SSTs returns to warmer, middle Eocene values in this interval. The continued sea surface warmth across the EOT has been related to persistent influence of the proto-Leeuwin Current (Houben et al., 2019), |
| 102610271028102910301031103210331034 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at ~35.5 Ma. Whereas this is merely a continuation of progressive late Eocene cooling in the SST record, the decrease in MAAT is strongly accelerated compared to middle–late Eocene trends. Another 3–4 °C MAAT drop occurs sometime between the late Eocene and the early Oligocene (likely associated with the Eocene–Oligocene transition), while SSTs returns to warmer, middle Eocene values in this interval. The continued sea surface warmth across the EOT has been related to persistent influence of the proto-Leeuwin Current (Houben et al., 2019), which apparently retained a constant temperature across the EOT. Since MAAT |
| 1026102710281029103010311032103310341035 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at ~35.5 Ma. Whereas this is merely a continuation of progressive late Eocene cooling in the SST record, the decrease in MAAT is strongly accelerated compared to middle–late Eocene trends. Another 3–4 °C MAAT drop occurs sometime between the late Eocene and the early Oligocene (likely associated with the Eocene–Oligocene transition), while SSTs returns to warmer, middle Eocene values in this interval. The continued sea surface warmth across the EOT has been related to persistent influence of the proto-Leeuwin Current (Houben et al., 2019), which apparently retained a constant temperature across the EOT. Since MAAT decreased while SSTs remained high, the influence of regional oceanographic changes did not impact climate changes in the source region of brGDGTs during this time |
| 10261027102810291030103110321033103410351036 | the late Eocene Oligocene, when they show a sharp cooling of 2–3 °C at the onset of greensand deposition, at ~35.5 Ma. Whereas this is merely a continuation of progressive late Eocene cooling in the SST record, the decrease in MAAT is strongly accelerated compared to middle–late Eocene trends. Another 3–4 °C MAAT drop occurs sometime between the late Eocene and the early Oligocene (likely associated with the Eocene–Oligocene transition), while SSTs returns to warmer, middle Eocene values in this interval. The continued sea surface warmth across the EOT has been related to persistent influence of the proto-Leeuwin Current (Houben et al., 2019), which apparently retained a constant temperature across the EOT. Since MAAT decreased while SSTs remained high, the influence of regional oceanographic changes did not impact climate changes in the source region of brGDGTs during this time |
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| 1041 | throughflow of the proto-Leeuwin Current through the TG in the Late Eocene |
|------|--|
| 1042 | (Houben et al., 2019). More remarkably, the mid-Paleocene SSTs approach those of |
| 1043 | the warmest interglacial intervals of Oligocene Wilkes Land Margin, east Antarctica |
| 1044 | (Hartman et al., 2018), at a similar paleolatitude as the Paleocene ETP. This suggests |
| 1045 | that mid-Paleocene Antarctic-proximal temperatures were similar to those at times of |
| 1046 | major Antarctic glaciation during EOT. Previous work has indeed presented |
| 1047 | widespread evidence for <mark>low sea level during this time interval (Frieling et al., 2014;</mark> |
| 1048 | 2018; Guasti et al., 2006; Hollis et al., 2014), which combined with low SSTs suggests |
| 1049 | the presence of some continental ice on the Antarctic continent during this mid- |
| 1050 | Paleocene interval. |
| 1051 | |
| 1052 | |
| 1053 | 5.2 BrGMGTs |
| 1054 | The continuous presence of brGMGTs in our record appears unrelated to |
| 1055 | depositional conditions or varying relative contributions of terrestrial material, |
| 1056 | regesting that at least part of the brGMGTs have a marine origin. This confirms |
| 1057 | previous observations from the modern sediments and water column (Liu et al. 2012, |
| 1058 | Xie et al. 2014) and Paleogene Arctic Ocean sediments (Sluijs et al., 2020). Their |
| 1059 | sparse presence in modern soils (Peterse et al., pers comm based on metadata from |
| 1060 | De Jonge et al., 2019; Kirkels et al., 2020) : lso in line with this, although abundant |
| 1061 | occurrence in peats and lakes (Naafs et al., 2018, Baxter et al. 2019, Tang et al., 2021) |
| 1062 | shows clear potential for terrestrial input. Despite the uncertainties in sourcing of |
| 1063 | brGMGTs, we find a strong resemblance with the signals from tropical lakes. The |
| 1064 | brGMGTI, which was derived to quantify the temperature relation of brGMGTs in |
| 1065 | surface sediments of East African lakes (Baxter et al., 2019), seems to correlate to |
| 1066 | temperature in our record as well (Fig. $12a-d$), although the better correlation of |
| 1067 | brGMGTI with MAAT than with SST (Fig. 12a–d) seems to be at odds with the |
| 1068 | presumed marine source of brGMGTs in our record. Despite all potential |
| 1069 | complications, our study provides additional evidence that a temperature signal may |
| 1070 | be governing the distribution of the different brGMGT isomers, as proposed by |
| 1071 | (Baxter et al., 2019), but there are differences as well. For instance, H1020c is |
| 1072 | altogether absent in the cold mid-Paleocene part of our record. There are numerous |





| 1073 | ways in which this can be explained, e.g., the biosynthesis of H1020c occurs only |
|---|--|
| 1074 | above a certain temperature, or the microbes that biosynthesize H1020c do not live |
| 1075 | at low temperatures, or the site receives brGMGTs from a different catchment in |
| 1076 | colder climates. Indeed, H1020c was almost absent in the cold (<12°C), high-altitude |
| 1077 | tropical lake samples (Baxter et al. 2019). However, those cold lakes did have high |
| 1078 | relative abundance of H1048 (Baxter et al., 2019), which our record lacks in the cold |
| 1079 | mid-Paleocene <mark>, illustrating the differences between modern lakes and our marine</mark> |
| 1090 | depositional setting. |
| 1081 | The HMBT record does not reflect a temperature signal when using all |
| 1082 | brGMGT isomers in the calculation (Fig. 12f), as H1020 isomers are by far the most |
| 1083 | abundant brGMGTs at Site 1172, and any change in the abundance of H1020c is |
| 1084 | compensated by both H1020b and H1020a in our record. In contrast, only using the |
| 1085 | H1020c and H1034b isomers <mark>, which following the chromatographic peak</mark> |
| 1036 | identification of Sluijs et al. (2020) match the compounds used to derive the HMBT |
| 1087 | index based on brGMGTs in peats (Naafs et al., 2018), does show a correlation to |
| 1088 | temperature in our record (Fig. 12g). |
| 1089 | %brGMGT as calculated in Naafs et al., (2018) or as in Baxter et al., (2019) |
| 1090 | does not show a clear relation with temperature (Fig. 12h, i), in contrast to |
| 1091 | observations in modern peats (Naafs et al., 2018a) and lake sediments (Baxter et al., |
| 1092 | 2019). We do note the extremely high %brGMGT (~80%) in the Maastrichtian and |
| 1093 | |
| | Oligocene, much higher than seen anywhere so far, We also do not see a clear inverse |
| 1094 | Oligocene, much higher than seen anywhere so far, We also do not see a clear inverse relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be |
| 1094 1095 | |
| | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be |
| 1095 | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be because of the complexity in the relative abundance of b rGDGTs in the record, which |
| 1095 1096 | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be because of the complexity in the relative abundance of b rGDGTs in the record, which does not necessarily reflect the variation in terrestrial versus marine sourcing . |
| 1095 1096 1097 | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be because of the complexity in the relative abundance of b rGDGTs in the record, which does not necessarily reflect the variation in terrestrial versus marine sourcing . The higher relative abundance and change in composition of brGMGT |
| 1095 1096 1097 1098 | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be because of the complexity in the relative abundance of brGDGTs in the record, which does not necessarily reflect the variation in terrestrial versus marine sourcing. The higher relative abundance and change in composition of brGMGT composition in the Oligocene that coincides with abundant signs of overprints in all |
| 1095 1096 1097 1098 1099 | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be because of the complexity in the relative abundance of brGDGTs in the record, which does not necessarily reflect the variation in terrestrial versus marine sourcing. The higher relative abundance and change in composition of brGMGT composition in the Oligocene that coincides with abundant signs of overprints in all br- and isoGDGT indices may indicate a source change of brGMGTs in this interval, |
| 1095 1096 1097 1098 1099 1100 | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be because of the complexity in the relative abundance of brGDGTs in the record, which does not necessarily reflect the variation in terrestrial versus marine sourcing. The higher relative abundance and change in composition of brGMGT composition in the Oligocene that coincides with abundant signs of overprints in all br- and isoGDGT indices may indicate a source change of brGMGTs in this interval, e.g., related to the same changes in contribution that cause the overprints in |
| 1095 1096 1097 1098 1099 1100 1101 | relationship between %brGMGT and BIT index, as in Sluijs et al. (2020). This may be because of the complexity in the relative abundance of brGDGTs in the record, which does not necessarily reflect the variation in terrestrial versus marine sourcing. The higher relative abundance and change in composition of brGMGT composition in the Oligocene that coincides with abundant signs of overprints in all br- and isoGDGT indices may indicate a source change of brGMGTs in this interval, e.g., related to the same changes in contribution that cause the overprints in isoGDGTs. The differences in brGMGT signals between our record, the Eocene Arctic |



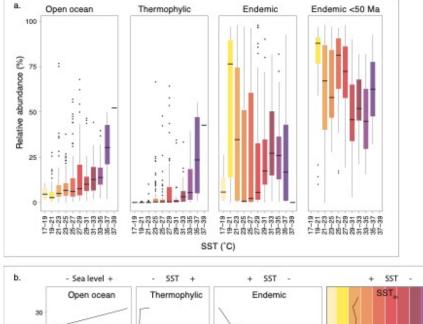


| 1105 | |
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| 1106 | |
| 1107 | 5.3 Integrated paleo environmental and -climatological |
| 1108 | 5.3.1 Sea level and ecosystem response to SST changes |
| 1109 | The abundance of temperature-responsive dinocyst eco-groups qualitatively |
| 1110 | confirm our SST trends. The thermophile dinocyst eco-group shows higher relative |
| 1111 | abundance at higher SSTs (Fig. 15). This eco-group has been empirically associated to |
| 1112 | SST based on its strong affinity to high SST in the global compilation of late |
| 1113 | Paleocene-early Eocene records (Frieling and Sluijs, 2018), and we here confirm this |
| 1114 | relationship for the longer early Paleogene. The relationship between isoGDGT-based |
| 1115 | SSTs and relative abundance of the supposedly cold-affiliated endemic dinocysts (Bijl |
| 1116 | et al., 2011) is not as clear. This might be because the majority of dinocyst species |
| 1117 | within the Feature lemic-Antarctic dinocyst ecogroup are restricted to the Eocene (Bijl et |
| 1118 | al., 2011). |
| 1119 | Indeed, from 50 Ma onwards, SST does have a relation with the %endemic |
| 1120 | Antarctic dinocyst abundance in the record (Fig. 15; Byret al., 2011), which may |
| 1121 | indicate a more distinct biogeographical separation through the development of |
| 1122 | stronger meridional gradients or evolutive adaptation of the endemic dinocyst |
| 1123 | community to more polar environments following the extreme warmth of the EECO. |
| 1124 | We particularly the strong correlation between TEX ₈₆ -based SST and relative |
| 1125 | abundance of more open ocean-affiliated dinocysts (Fig. 13), which suggests that SST |
| 1126 | correlates with regional sea level. SST-induced sea level changes hint at steric effects |
| 1127 | as driver. Indeed, because of the flatness of continental shelf areas during long-term |
| 1128 | greenhouse climates (Somme et al., 2009) small changes in regional sea level will |
| 1129 | cause large changes in distance to shore and associated characteristics, including e.g. |
| 1130 | salinity, nutrients and suspended sediment loads, which subsequently shapes the |
| 1131 | dinocyst assemblages. |
| 1132 | |





Fig.15 GDGT- and palynology-based proxies for SST



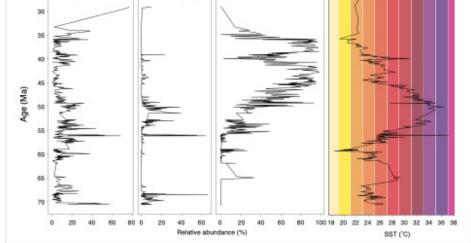




Figure 15. isoGDGT- and palynology-based proxies for SST. Prelative abundance of
open ocean, thermophylic and endemic-Antarctic dinocysts (total dataset and
samples <50Ma only) plotted against linearly interpolated (resampled) TEX₈₆-based
SST, in 2 degree bins. b. Time series of the same dinocyst ecogroups and SST.





| 1139 | The SST control on dinocyst assemblages is further demonstrated in diversity |
|--------------------|--|
| 1140 | and assemblage variability indices. Throughout the long time interval covered in our |
| 1141 | record, dinocyst richness and diversity show a remarkably similar pattern to $\mathrm{TEX}_{86}	ext{-}$ |
| 1142 | based SST. Species richness, as well as diversity expressed in the Shannon and |
| 1143 | Simpson indices and Fisher's alpha reach a maximum during the prolonged warmth |
| 1144 | of the EECO and drop during middle–late Eocene cooling. These relationships are |
| 1145 | demonstrated by modest correlation coefficient between diversity and ۲۳۲ ₈₆ (Fig. |
| 1146 | 13). This pattern holds for the long-term trends, as well as for the short-term PETM, |
| 1147 | but not for the MECO. Dinocyst diversity is known to vary in an inshore-to-offshore |
| 1148 | transect in the modern and Eocene (Brinkhuis, 1994; Pross and Brinkhuis, 2005). As |
| 1149 | our assemblages indeed consist of many mid-shelf species, the <mark>strong correlation</mark> |
| 1149 = 1130 | between diversity and SST might thus be indirectly related through habitat size, with |
| 1151 | expansion of the flat shelf area and thereby increase in shelf niches during the EECO |
| 1152 | (Somme et al., 2009). Variability as indicated by the Gibbs index records the opposite |
| 1153 | pattern to diversity for the long-term trends into and out of the EECO (1997). That |
| 1154 | is, the lower-diversity assemblages in general have a higher variability, or sample-to- |
| 1155 | sample fluctuations within the relative abundances. This indicates the higher |
| 1156 | diversity dinocyst assemblages might be more ecologically resilient. |
| 1157 | 5.3.2 Marine environmental response to runoff changes |
| 1158 | Senegalinium cpx. abundance broadly correlates with the IR (Fig. 16), which |
| 1159 | signals input of river-produced brGDGTs. Senegalinium cpx. is generally thought to |
| 1160 | have tolerated low salinity environments (Barke et al., 2011; Sluijs et al., 2005; Sluijs |
| 1161 | and Brinkhuis, 2009). These two proxies together confirm a large input of fresh water |
| 1162 | at this site during the mid-to-late Paleocene in line with high relative abundance of |
| 1163 | terrestrial palynomorphs, low abundances of open ocean dinocysts and high IR values |
| 1164 | (Fig. 16). Interestingly, Phthanoperidinium spp., which is also generally associated |
| 1165 | with low-salinity to near fresh-water conditions (Barke et al., 2011; Frieling and |
| 1166 | Sluijs, 2018; Sluijs and Brinkhuis, 2009) <mark>dees not correlate to IR (Fig. 16). Because we</mark> |
| 1167 | find different species of <i>Phthanoperidinium</i> spp. than those previously associated to |
| 1168 | fresh-water conditions (e.g., Barke et al., 2011), it could be that the fresh-water |
| 1169 | tolerance of Phthanoperidinium spp. was not shared among all species. It could also |
| 1170 | be that <i>Phthanoperidinium</i> spp. is slightly less euryhaline than <i>Senegalinium</i> . |
| | |





| 1171 | Tasmania was located at latitudes between 55° and 60° S, in the middle of the |
|------|---|
| 1172 | region of strong westerly winds, within the range of low atmospheric pressure and |
| 1173 | received a large amount of precipitation during the Paleocene (Huber and Caballero, |
| 1174 | 2011). River input from Tasmania and perhaps also Australia could have reached the |
| 1175 | site. Interestingly, while Tasmania migrated northwards only a few degrees latitude |
| 1176 | between 60 and 40 Ma (Fig. 1), the freshwater input decreased, based on both |
| 1177 | Senegalinium cpx., terrestrial palynomorph abundance (decimated abundance from |
| 1178 | 50 Ma onwards) and the IR (decrease to normal values around 54 Ma; Fig. 16). This |
| 1179 | suggests either a rerouting of river input or a <mark>drying of the hinterland.</mark> Support for the |
| 1180 | latter comes from clay mineralogical data, showing an increase in kaolinite starting at |
| 1181 | 50 Ma (Fig. 2; Robert, 2004). Kaolinite forms abundantly at the base of acidic peats |
| 1182 | (Staub and Cohen, 1978; Korasidis et al., 2019) and in old, leached soils, which were |
| 1183 | ubiquitous in the Eocene Australian hinterland (Holdgate et al., 2009). While the |
| 1184 | hinterland could well retain the kaolinite-rich clays during the Maastrichtian and |
| 1185 | Paleocene, because the site was under a year-round high precipitation regime, it was |
| 1186 | less efficient in doing so when the hinterland experienced drier, more variable |
| 1187 | climatic conditions as Australia migrated northward. The increase in kaolinite |
| 1188 | delivery to Site 1172 is hence interpreted as a signal of enhanced soil (or peat) |
| 1189 | erosion from the catchment areas in Tasmania and SW, as a result of a more variable |
| 1190 | -limate regime. |
| 1191 | |
| 1192 | 5.4 Environmental and climatological changes in the catchment |
| 1193 | The long-term trends in the BIT index are at odds with all the river runoff |
| 1194 | indicators in our data, BIT remains low during the presumed interval with increased |
| 1195 | river water discharge, as indicated by the high IR and abundance of <i>Senegalinium</i> and |
| 1196 | increases when this river-input signal ceases, This is unexpected , because the BIT |
| 1197 | index should reflect high input of soil-derived brGDGTs into the marine sediments, |
| | |

- 1198 associated with increased discharge (Hopmans et al., 2004). There are two ways to
- 1199 explain this signal. The BIT index could responding to marine crenarchaeol
- 1200 production, rather than to the terrestriar or GDGT flux. Indeed, marine productivity
- 1201 could have been spurred by runoff and associated nutrient delivery to the coastal
- 1202 zone during the Paleocene, and decreased in the Eocene, affecting BIT index relates.





| 1203 | Another explanation is that the BIT index has been influenced by crenarchaeol |
|------|--|
| 1204 | production in the river (Zell et al., 2013), although the river-produced crenarchaeol |
| 1205 | concentrations are normally negligible compared to that produced in the marine |
| 1206 | realm. Decreasing BIT index values with increasing river-and soil input has been |
| 1207 | related to catchment dynamics in the late Quaternary Zambezi river (Lattaud et al., |
| 1208 | 2017), involving vegetation dynamics controlling soil-OM input. Studies on modern |
| 1209 | soil-river-ocean pathways in the Amazon River system show that whereas river- |
| 1210 | suspended brGDGTs in its upper tributaries reflect catchment soils (Kirkels et al., |
| 1211 | 2020), brGDGTs in the lower Amazon receive increasing contributions of in situ |
| 1212 | produced brGDGTs (Zell et al., 2013), and marine surface sediments of the Amazon |
| 1213 | fan contain a mix of soil, riverine, and marine brGDGTs (Zell et al., 2014). In a high- |
| 1214 | precipitation, soil-rich environment like the Amazon basin, this can only occur if the |
| 1215 | river-produced brGDGT production progressively exceeds the soil-derived brGDGTs |
| 1216 | input in river water along the trajectory of the river, or when the soil-derived |
| 1217 | brGDGTs are preferentially remineralized during river transport. We here propose |
| 1218 | the following scenario to explain the BIT index trends: In the Paleocene– carly Eocene, |
| 1219 | excessive, year-round precipitation-fed river runoff caused strong fresh-water |
| 1220 | delivery into the sw Pacific. Soil-derived brGDGTs were partly remineralized in the |
| 1221 | river catchment, and the strongly diluted soil-derived OM was dominated by river- |
| 1222 | produced brGDGTs and crenarchaeol, both in river production and on the continental |
| 1223 | shelf, which suppressed the BIT index. This explains the provide the maxpectedly cold MAATs |
| 1224 | derived from the mid-Paleocene samples with high IR. The nigh riverine input did |
| 1225 | promote pelagic isoGDGT production: crenarchaeol which decreases the BIT index |
| 1226 | and isoGDGTs in general. In the early Eocene, precipitation in the hinterland |
| 1227 | decreased as the hinterland gradually drifted out of the zone of intense precipitation. |
| 1228 | This reduced river discharge, but at the same time the more dynamic climate regime |
| 1229 | caused more seasonal precipitation which increased destabingation of Australian and |
| 1230 | Tasmanian soils, leading to a higher abundance of soil-derived brGDGTs and kaolinite |
| 1231 | from destabilized soils in the river discharge, and relatively little river-produced |
| 1232 | brGDGTs (because enhanced soil-OM content increased turbidity) and river-produced |
| 1233 | crenarchaeol, which increased BIT index at the ETP . |





Fig. 16 GDGT- and palynology-based proxies for runoff

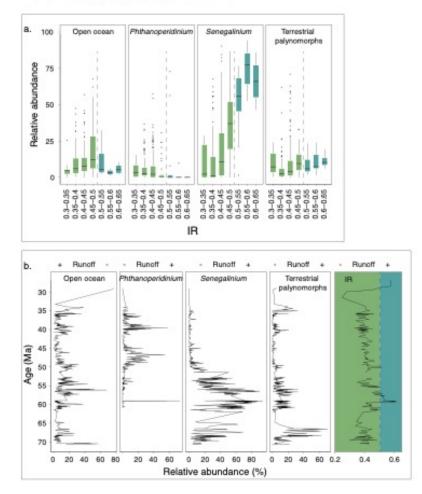


Figure 16. BrGDGT- and palynology-based proxies for river runoff. a. relative abundance of Open Ocean, *Phthanoperidinium* spp., *Senegalinium* cpx. and terrestrial palynomorphs in 0.05 index unit bins of the resampled IR. Dashed line separates elevated IR bins from non-elevated IR bins. b. Time series of the same palynology ecogroups and the IR.

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1244 **6. Conclusions**

| o. conclusions |
|--|
| We have critically reviewed the GDGT- and dinoflagellate cyst data in samples from |
| the Maastrichtian–lower Oligocene sediments at ODP Site 1172. IsoGDGTs represent |
| a pelagic signal throughout the Maastrichtian–Eocene and are influenced by |
| sedimentary-produced isoGDGTs in the Oligocene. BrGDGTs at our site are likely |
| primarily soil- or peat-derived, albeit evolutionary changes in brGDGT prodcution |
| may have altered the Paleogene soil brGDGT composition. Exceptions are the mid- |
| Paleocene, where river-produced brGDGTs influence the record, and the Oligocene, |
| where marine in situ produced brGDGTs dominate. TEX ₈₆ and MBT' _{5me} records reveal |
| the SST and MAAT evolution of the region, respectively, consistent with independent |
| existing paleotemperature reconstructions from the same samples. A temperature |
| offset between the SST and MAAT proxies remains poorly reconciled with the coastal |
| proximity of the site but is likely a combined effect of evolutionary differences in |
| MBT' _{5me} -MAAT relationships and a <mark>disparate integration of climate signals by both</mark> |
| proxies. The subdued air temperature cooling in the middle Eocene compared to the |
| SST cooling could in part reflect progressive northward tectonic drift of the |
| hinterland, while SSTs remained continuously influenced by the Antarctic-derived |
| Tasman Current, Ctan MAAT cooling occurred step-wise around the Eocene- |
| Oligocene transition. The absence of SST cooling around the Eocene–Oligocene |
| transition suggests some disconnection between oceanographic reorganisations and |
| regional MAAT changes. The absence of a MAAT response at the PETM and MECO |
| remains unresolved but might be related to shifting sources of brGDGTs during these |
| warm phases. |
| The relatively high IR, and low #rings _{tetra} firm dinocyst assemblages characteristic |
| of an inner mid-shelf depositional setting. we reconstruct strong river-runoff in the |
| Paleocene–early Eocene at this site, and normal open-marine conditions thereafter. |
| The latter reflects a drying and increased seasonality in precipitation in the |
| catchment as the continent tectonically migrates northwards, out of the region of |
| strong precipitation during the middle and late Eocene. The correlation between SST |
| and changes dinoflagellate cyst assemblages and biodiversity metrics suggest |
| temperature-controlled sea level changes influenced the site, probably through steric |
| effects. We find corroborating evidence for a temperature signal in brGMGT |
| |





1276 assemblages, further demonstrating the potential to develop novel proxies with more 1277 extensive studies on modern affinities. 1278 1279 **7.Supplements** 1280 Table S1: Age tie points 1281 • Table S2: grouping of dinocysts in ecogroups 1282 1283 8. Data availability 1284 Raw GDGT/GMGT and palynological data presented in this paper, as well as R 1285 markdown code for data analysis and visualization can be found on Github 1286 (https://github.com/bijlpeter83/RGDGT.git). Data was deposited at zenodo under 1287 DOI:10.5281/zenodo.4471204. 1288 1289 9. Author contributions 1290 AS (PETM) and PKB (part) prepared samples for GDGT analyses. PKB (part), AS 1291 (PETM) and CB (part) integrated UHPLC-MS results. AS (PETM) and PKB (part) 1292 analysed palynological samples. All authors contributed to the interpretation of the 1293 OG results, and PKB, AS, JF and MC interpreted dinocyst results. PKB wrote the R 1294 script for data analyses and visualization, drafted the figures and wrote the paper 1295 with input from all authors. 1296 1297 **10. Acknowledgements** 1298 This research used samples from the Ocean Drilling Program, which was funded 1299 through U.S. National Science foundation under the management of joined 1300 oceanographic institutions, inc. We thank Klaas Nierop, Desmond Eefting and Natasja 1301 Welters for technical/analytical support. PKB acknowledges funding through NWO 1302 Vernieuwingsimpuls Veni grant no. 863.13.002. This work was carried out under the 1303 program of the Netherlands Earth System Science Centre (NESSC), financially 1304 supported by the Dutch Ministry of Education, Culture and Science. MC and AS thank 1305 the Ammodo Foundation for funding unfettered research of laureate AS. PB and AS 1306 thank the European Research Council for Starting Grant 802835 OceaNice and 1307 Consolidator Grant 771497 SPANC, respectively. 1308





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