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Towards a Cenozoic History of Atmospheric CO₂
The Cenozoic CO₂ Proxy Integration Project (CenCO₂PIP) Consortium

Abstract: The geological record encodes the relationship between climate and atmospheric carbon dioxide (CO₂) over long and short timescales, as well as potential drivers of evolutionary transitions. However, reconstructing CO₂ beyond direct measurements requires the use of paleo-proxies and herein lies the challenge, as proxies differ in their assumptions, degree of understanding, and even reconstructed values. Here we critically evaluate, categorize, and integrate available proxies to create a high-fidelity and transparently constructed atmospheric CO₂ record spanning the past 66 million years. This provides clearer evidence for higher Earth System Sensitivity in the past and for the role of CO₂ thresholds in biological and cryosphere evolution.

14 The contribution of atmospheric CO₂ to Earth's greenhouse effect and the potential for variations
15 in the global carbon cycle to cause climate change has been known for more than a century (1),
16 but it was only in 1958 that direct measurements of the concentration of CO₂ in the atmosphere
17 (or molar mixing ratio - the mole fraction of a gas in one mole of air) were systematically
18 collected. Alongside reconstructions of the historical rise in Earth's surface temperature (2), this
19 record has become one of the most influential and scientifically valuable environmental time-
20 series, documenting the continuous rise in annual mean CO₂ from 315 parts per million (ppm) in
21 1958 to 419 ppm in 2022 (3). Projecting beyond these records to estimate how Earth's climate
22 will respond to further increases in CO₂ requires global climate models (4). However, while
23 successful in explaining observed historical climate change (2), models leave doubt as to whether
24 global mean temperature will rise linearly as a function of future doubling of CO₂ (i.e., an invariant
25 'climate sensitivity') or whether climate feedbacks will lead to an increasing (or 'state-
26 dependent') sensitivity of climate to CO₂ in the future (5, 6).

27 We can turn to the geological record to help constrain models and improve our
28 understanding of non-linearities in the climate system (e.g., 7), as it documents a variety of global
29 climate changes and critically, climate states warmer than today. Leveraging this record,
30 however, requires the paired quantification of both past atmospheric CO₂ and temperature. In
31 parallel with recent efforts to compile and vet paleo-temperature estimates (8), here we focus
32 on paleo-CO₂ estimates. Samples of ancient air can be extracted and analyzed from bubbles
33 preserved in ancient polar ice (9, 10), but continuous ice core records currently only extend our
34 knowledge of CO₂ back about 800 thousand years (kyr) (for a compilation, see 11), with isolated
35 time slices extending to ~2 Ma (million years ago) (12, 13). Importantly, at no point during the
36 Pleistocene (2.58 Ma to 11,700 years ago) did CO₂ come close to present-day values (419 ppm,
37 year 2022), with 300 ppm being the highest value measured to date (14). In contrast, depending
38 on the extent of future human emissions, atmospheric CO₂ could reach 600–1000 ppm by the
39 year 2100 (2). Feedbacks between changing climate and the carbon cycle may also amplify or
40 diminish emissions from surficial carbon reservoirs (e.g., thawing permafrost, adjustments in size
41 and composition of the terrestrial biosphere and marine carbon pool), creating additional
42 uncertainty in future CO₂ projections (15, 16). Past changes in CO₂ inherently include the role of
43 these feedbacks, and their study could help reduce uncertainty in Earth system models (17).

44 A solid understanding of atmospheric CO₂ variation through geological time is also
45 essential to deciphering and learning from other features of Earth's history. Changes in
46 atmospheric CO₂ and climate are suspected to have caused mass extinctions (e.g., 18, 19) as well
47 as evolutionary innovations (20, 21). During the Cenozoic, long-term declines in CO₂ and
48 associated climate cooling have been proposed as the drivers of changing plant physiology (e.g.,
49 carbon-concentrating mechanisms), species competition and dominance, and associated with
50 this, mammalian evolution. A more refined understanding of past trends in CO₂ is therefore
51 central to understanding how modern species and ecosystems arose and may fare in the future.

52 Extending the CO₂ record beyond the temporally restricted availability of polar ice
53 requires the use of 'proxies'. In essence, a CO₂ proxy could be any biological and/or geochemical
54 property of a fossil or mineral that responds to the concentration of ambient CO₂ when it is
55 formed. Unfortunately, unlike in the case of bubbles of ancient air trapped in polar ice, this
56 response is invariably indirect. The connection between a proxy signal and atmospheric CO₂ is

57 often strongly mediated via biological 'vital effects' (e.g., concentration of or discrimination
58 against certain molecules, elements or isotopes due to physiological processes such as
59 biomineralization, photosynthesis, respiration), may be indirectly connected to the atmosphere
60 via dissolution of carbon in seawater or lakes, may involve isotopic or other chemical
61 fractionation steps, or a combination of these. When preserved in terrestrial or marine
62 sediments, proxy substrates can also be impacted by post-depositional ('diagenetic') processes
63 that must be accounted for. Relationships between proxies and CO₂ are typically calibrated using
64 observations or laboratory experiments; in biological systems, these calibrations are often
65 limited to modern systems (e.g., modern organisms or soils), and applications to the distant past
66 focus on physiologically or physically similar systems preserved in the sediment and rock record
67 (e.g., similar fossil organisms or fossil soils). Most CO₂ proxies also require estimation of one or
68 more additional environmental parameters and hence depend on additional proxy records. The
69 complexity of proxy-enabled paleoclimate reconstructions thus presents a major challenge for
70 creating a self-consistent estimate of atmospheric CO₂ through geological time and requires
71 careful validation.

72 One of the first paleo-CO₂ proxies to be devised was based on the observation that
73 vascular plants typically optimize the density, size, and opening/closing behavior of stomatal
74 pores on their leaf surfaces to ensure sufficient CO₂ uptake while minimizing water loss (e.g., 22).
75 A count of stomatal frequencies then provides a simple proxy for the CO₂ concentration
76 experienced by the plant (23). Changes in ambient CO₂ can also drive a cascade of interrelated
77 effects on photosynthesis, the flux of CO₂ into the leaf (largely determined by stomatal size and
78 density), and the carbon isotopic fractionation during photosynthesis ($\Delta^{13}\text{C}$, 22, 23, 24). While
79 lacking functional stomata, non-vascular plants like liverworts also exhibit isotopic fractionation
80 during photosynthesis, and their $\delta^{13}\text{C}$ values are thus similarly controlled by ambient CO₂. The list
81 of terrestrial paleo-CO₂ proxies also includes inorganic carbonate nodules precipitated in ancient
82 soils (i.e., paleosols) as well as sodium carbonate minerals precipitated in continental lacustrine
83 evaporites. While the paleosol proxy uses the carbon isotope composition of carbonate nodules
84 and deconvolves the mixture of atmospheric and soil-respired CO₂ in soil porewaters using
85 models of soil CO₂ (25, 26), the nahcolite proxy is based on the CO₂ dependence of sodium
86 carbonate mineral equilibria (27, 28). Analogous to non-vascular plants on land, phytoplankton
87 fractionate carbon isotopes during photosynthesis in response to the concentration of dissolved
88 CO₂ in seawater, creating an isotopic signal stored in organic biomolecules that can be retrieved
89 from ocean sediments (29). Boron proxies recorded in fossil shells of marine calcifying organisms
90 are related to seawater pH, which in turn can be related back to atmospheric CO₂ (30, 31). A
91 detailed discussion of the analytical details, entrained assumptions, and inherent uncertainties
92 of currently available CO₂ proxies, plus summaries of recent advances and opportunities for
93 further validation, is presented in the Supplemental Material and in Table S1.

94 Although each of these proxies has been validated extensively, comparing
95 reconstructions from different proxies often reveals discrepancies. Prior compilations of paleo-
96 CO₂ and explorations of the CO₂-climate linkage already exist (32-34), however, those studies
97 applied limited proxy vetting, include CO₂ estimates that predate major innovations in some
98 methods, and use rather basic data interpolation to assess broad CO₂ trends. Earlier CO₂
99 reconstructions are also often insufficiently constrained by ancillary data (e.g., concomitant

100 temperature, isotopic composition of seawater or atmosphere) to be consistent with modern
101 proxy theory, have incomplete or missing uncertainty estimates for CO₂ and/or sample age, and
102 may exhibit fundamental disagreement with other proxies, leaving our current understanding of
103 past CO₂ incomplete.

104 In this study we present the results of a 7-year endeavor by an international consortium
105 of researchers whose collective expertise spans the reconstruction of paleo-CO₂ from all available
106 terrestrial and marine archives. We have jointly created a detailed, open-source database of
107 published paleo-CO₂ estimates including all raw and ancillary data together with associated
108 analytical and computational methods. Each record was vetted and categorized in view of the
109 most recent proxy understanding, with calculations adopting a common methodology including
110 full propagation of uncertainties. We focus our efforts here on the Cenozoic, when the spatial
111 distribution of continents and ocean basins, as well as the structure of marine and terrestrial
112 ecosystems, was similar to the modern, yet profound changes in CO₂ and climate occurred.
113 Identifying the most reliable Cenozoic CO₂ estimates published to date allows us to quantify
114 important physical (e.g., temperature, ice volume) and biological (i.e., physiological, ecosystem)
115 thresholds and tipping points.

116 We structure this investigation as follows: First we summarize the methodology by which
117 we assessed the CO₂ proxies and associated estimates. We then apply these methods to derive
118 a series of paleo CO₂ compilations comprised of data with different levels of quality or
119 confidence, and statistically integrate the 'top-tier' data to create a realization of the Cenozoic
120 variability in atmospheric CO₂. This is followed by a discussion of the climatic implications
121 (including climate sensitivity) of the paleo-CO₂ curve, and a presentation of an evolutionary
122 perspective. We finish with a roadmap for further advances in understanding past changes in
123 atmospheric CO₂.

124

125 **Critical assessment of atmospheric CO₂ proxies**

126 The basis of our synthesis is a set of comprehensive data templates documenting all types
127 of proxy data and their corresponding CO₂ estimates (a total of 6,247 data points). The completed
128 data sheets for each study can be accessed as the [paleo-CO₂ 'Archive'](#) in NOAA's National Climatic
129 Data Center (NCDC). These 'Archive' sheets report all underlying data at face value from the
130 original publications, but their unprecedented level of detail is designed to facilitate critical
131 evaluation and recalculation of each CO₂ estimate.

132 From the 'Archive', published CO₂ estimates were evaluated by teams of experts who are
133 active in validating and applying these proxies, and often included the original authors of the
134 respective data. No new proxy data were collected as part of this effort, but estimates were
135 recalculated where needed and possible, and age models were revised where new evidence was
136 readily accessible. Additionally, CO₂ and age uncertainties were updated, as necessary, to
137 consistently reflect propagated 95% confidence intervals. The vetting criteria are summarized in
138 Supplementary Table S1 and detailed [in paleo-CO₂ 'Product'](#) sheets. These CO₂ estimates are
139 categorized as follows: 'Category 1' estimates (Fig. 1a, 1,673 data points or ~27% of the original
140 total) are based on data whose uncertainty is fully documented and quantifiable in view of

141 current proxy understanding. 'Category 2' estimates (Fig. 1b, 1,813 data points) contain sources
142 of uncertainty that are not yet fully constrained. These uncertainties vary between proxies and
143 datasets, and include, e.g., insufficient replication, poorly constrained proxy sensitivity to
144 parameters other than CO₂, or extrapolation of calibration curves. 'Category 3' estimates (the
145 residual 2,761 data points or ~44% of the Cenozoic paleo-CO₂ estimates published to date) are
146 either superseded by newer, independently published evaluations from the same raw data, are
147 considered unreliable due to factors such as incomplete supporting datasets that prevent full
148 quantification of uncertainties, or outdated sample preparation methods.

149 Although objective criteria are applied throughout, the vetting process was particularly
150 challenging for the paleosol- and phytoplankton-based proxies because multiple approaches are
151 currently in use for interpreting these proxy data (35-41). Given the lack of a universally agreed-
152 upon method, we compare multiple approaches for treating the data of these two proxies
153 whenever possible. For the paleosol proxy, the greatest source of uncertainty is in the estimation
154 of paleo-soil CO₂ concentration derived from respiration. Two different approaches are
155 commonly used to do this. The first method is based on proxy-estimated mean annual rainfall,
156 while the second is based on soil order (i.e., the most general hierarchical level in soil taxonomy,
157 comparable to kingdom in the classification of biological organisms). However, few records in the
158 database allow for a direct comparison between the two approaches. An opportunity for
159 comparison exists with two Eocene records (37, 42), where re-calculation using each of the two
160 different methods leads to CO₂ estimates that do not overlap within 95% confidence intervals for
161 most stratigraphic levels (Fig. S6). This implies that the uncertainty in estimating paleo-soil CO₂
162 concentration derived from respiration cannot be fully quantified with either of these
163 approaches. Thus, most paleosol-based CO₂ estimates were designated as Category 2. For the
164 phytoplankton proxy, routinely applied methods differ in how algal cell size and growth rate are
165 accounted for, as well as the assumed sensitivity of algal $\delta^{13}\text{C}$ values to aqueous CO₂
166 concentration (see Supplementary Materials for details). Where data are available, we compare
167 both newer and traditional methods, finding that although there are deviations between the
168 resulting CO₂ estimates, they do agree within 95% confidence intervals. We hence assign many
169 phytoplankton CO₂ estimates to Category 1 and present mean CO₂ and uncertainty values that
170 reflect the range of results from the different methods.

171

172 **Towards a Cenozoic history of atmospheric CO₂**

173 Our composite Category 1 and 2 realizations of Cenozoic CO₂ (Figs. 1a and b) display much
174 better agreement among proxies than does the 'raw', un-curated collection ('Archive', Fig. 1c).
175 Encouragingly, objective criteria applied to the original data products automatically placed the
176 earlier-reported estimates of 'negative' CO₂, as well as some unusually high values, into Category
177 3, and without subjective intervention to otherwise filter them. We note that the Category 1
178 composite is now largely dominated by marine proxy estimates, with some intervals (e.g., the
179 middle Paleocene, ~63-57 Ma) very sparsely sampled. Furthermore, some intervals (e.g.,
180 Oligocene, Miocene) still exhibit significant differences between proxies; for instance, marine-
181 based CO₂ estimates start high and decline during the Oligocene (~34-23 Ma), whereas plant-
182 based estimates suggest overall lower and constant CO₂ (Fig. 1a). Estimates of global

183 temperature (Fig. 2b) during this time interval are largely invariant, which leaves us with the
184 questions of whether CO₂ and climate were decoupled during this interval, or whether there is a
185 systematic bias in the marine or plant-based CO₂ proxies and/or in the temperature proxies. All
186 proxies become more uncertain further back in time as our knowledge of vital effects in biological
187 proxy carriers, secular changes in the elemental and isotopic composition of ocean and
188 atmosphere, as well as proxy sensitivity to environmental parameters that change along with CO₂
189 (e.g., temperature, rainfall, see Supplementary Materials for details) becomes less certain. In
190 some cases, ancillary constraints and uncertainties are shared across multiple proxies (e.g.,
191 assumed atmospheric δ¹³C is common to proxies based on land plant δ¹³C, leaf gas exchange,
192 and paleosols), creating interdependence of estimates from seemingly independent proxies.
193 More robust paleo-CO₂ reconstruction thus requires not only continued application of all proxies
194 but also replication from different locations.

195 Although some uncertainties and proxy disagreements remain, the much-improved
196 agreement within the vetted paleo-CO₂ compilation gives us confidence that a quantitative
197 reconstruction of Cenozoic CO₂ based on the combined Category 1 data is possible. To do so, we
198 statistically model mean CO₂ values at half-million-year intervals, together with uncertainties in
199 age and proxy CO₂ estimates (Fig. 2a, see Supplementary Materials for details). Our choice of a
200 500-kyr resolution interval reflects a compromise driven by the proxy data compilation. Although
201 parts of the Cenozoic, particularly the Plio-Pleistocene, are sampled at higher temporal
202 resolution, the density of records remains relatively sparse throughout much of the Paleogene (1
203 datum per 190 kyr on average). As a result, the data (and in some cases the underlying age
204 models) are not suited to interpreting higher-frequency (e.g., Milankovitch-scale) variations in
205 atmospheric composition, and we focus here on low-frequency (e.g., multi-million year) trends
206 and transitions. Proxy sampling within some intervals may be biased toward conditions that
207 deviate from the 500-kyr mean (most notably here, the Paleocene-Eocene Thermal Maximum,
208 PETM). We do not attempt to remove this bias but recommend caution in interpreting any
209 features expressed at sub-million-year timescales.

210 This curve (Fig. 2a) allows us to constrain Cenozoic paleo-CO₂ and its uncertainty with
211 greater confidence than earlier efforts. The highest CO₂ values of the past 66 Myr appear during
212 the Early Eocene Climatic Optimum (EECO, ~53-51 Ma), while the lowest values occur during the
213 Pleistocene. In contrast to earlier compilations, which suggested early Cenozoic CO₂
214 concentrations <400 ppm (e.g., 33), rigorous data vetting and newly published records place early
215 Paleocene mean CO₂ in our reconstruction between 650 and 850 ppm. However, the Paleocene
216 remains data poor, and uncertainty in the curve remains large. Although the Paleocene record is
217 predominantly based on the boron isotope proxy (Fig. 1a), inclusion of other (non-marine) proxy
218 data does influence and refine the reconstruction through this epoch, supporting the value of
219 the multi-proxy approach (Fig. S10). Following the rapid CO₂ rise and fall associated with the
220 PETM at 56 Ma, mean CO₂ steadily rose to peak values of ~1600 ppm around 51 Ma during the
221 EECO. The middle and late Eocene recorded slightly lower values (800-1100 ppm). Mean CO₂
222 dropped to <600 ppm across the Eocene-Oligocene transition (EOT, 33.9 Ma) and reached values
223 that generally fall between ~400 and 200 ppm during the Miocene through Pleistocene, except
224 for a notable increase during the Middle Miocene (~17-15 Ma) to a mean of ~500 ppm.
225 Uncertainty in the mean CO₂ values drops substantially in the Plio-Pleistocene (see also Fig. S11),

226 as expected given a dramatic increase in data density. Our analysis suggests that ~14.5-14 Ma
227 was the last time 500-kyr-mean CO₂ value was as high as the present (Fig. S11), and that all Plio-
228 Pleistocene peak interglacial CO₂ concentrations were exceptionally likely less than those of the
229 modern atmosphere (Fig. S12). In contrast, prior to the Miocene, there is very little support
230 (<2.5% probability) for Cenozoic 500-kyr-mean CO₂ values reaching or falling below pre-industrial
231 levels.

232

233 **Climatic implications of the revised CO₂ curve**

234 **Relationship with global temperature change and climate sensitivity**

235 Our reconstructed Cenozoic CO₂ trends are broadly coherent with those for global
236 temperature as inferred, e.g., from the oxygen isotopic composition ($\delta^{18}\text{O}$) of fossil benthic
237 foraminifera shells (43, 44) and compilations of global surface temperature (45) (Fig. 2b). The
238 Paleocene and Eocene epochs display overall higher temperatures and atmospheric CO₂
239 concentrations as compared to the later Oligocene, Miocene, and Pliocene - consistent with a
240 predominantly greenhouse-gas regulated global energy budget. More specifically, the slow rise
241 and subsequent fall of CO₂ over the course of the Paleocene and Eocene are mirrored by global
242 temperatures, just as a transient Miocene CO₂ rise coincides with a period of warming at the
243 Miocene Climatic Optimum (MCO). The EOT is identifiable in both the CO₂ and temperature
244 records, despite the smoothing introduced by the curve fitting and 500-kyr binning interval.

245 Despite this overall agreement, rates and timing of CO₂ vs. temperature changes in the
246 two records are not always synchronized (Fig. 2a,b). For example, CO₂ appears broadly static or
247 even rising during the late Eocene (37-34 Ma) and late Miocene (11-5 Ma) despite global cooling
248 (see also 46) at these times. Conversely, decreasing CO₂ during the early Oligocene corresponds
249 with relatively stable global temperatures (Fig. 2b, but see also 47, 48) and ice volume (Fig. 2c) at
250 that time. We note that the reconstructed Oligocene CO₂ decrease is driven by the contribution
251 of marine proxies to the composite curve, whereas estimates from leaf gas exchange proxies are
252 low and broadly static (Fig. 1c), a discrepancy that cannot be resolved without further
253 experimentation and data collection. We caution that, even at the 500-kyr resolution of our
254 study, the relative timing of CO₂ and temperature change might be unresolved in poorly sampled
255 intervals (i.e., middle Paleocene), but should be well resolved during more recent, well sampled
256 intervals (i.e., late Miocene through present, Fig. S8). Is the occasional divergence of temperature
257 and CO₂ change evidence for occasional disconnects between CO₂ forcing and climate response?
258 Although one might posit bias in the CO₂ reconstruction, the strength of our multiproxy approach
259 is the reduced likelihood that multiple proxies exhibit common bias during particular periods of
260 the Cenozoic. We suggest that some cases of divergence between temperature and CO₂ could
261 reflect non-CO₂ effects on climate (e.g., changes in paleogeography affecting ocean circulation,
262 albedo and heat transport, 49), or the temperature reconstructions used herein could be biased
263 by non-thermal influences (e.g., uncertain elemental and isotopic composition of paleo-
264 seawater, physiological or pH effects on proxies, 48, 50).

265 Our updated CO₂ curve, in conjunction with existing global temperature reconstructions,
266 gives us the opportunity to reassess how climate sensitivity might have evolved through the

267 Cenozoic. The most commonly reported form of climate sensitivity is equilibrium climate
268 sensitivity (ECS), which focuses on fast feedback processes (e.g., clouds, lapse rate, snow, sea ice)
269 and is therefore best suited for predicting present-day warming ($\sim 3^\circ\text{C}$ for a doubling of CO_2 above
270 the pre-industrial condition, 2). Because the average temporal resolution of our CO_2 database is
271 coarser than 1000 years, we cannot estimate ECS directly. Instead, our data are most appropriate
272 for interpreting an Earth System Sensitivity ($\text{ESS}_{[\text{CO}_2]}$, following the taxonomy of 51) – the
273 combination of short-term climate responses to doubling CO_2 plus the effects of slower,
274 geological feedback loops such as the growth and decay of continental ice sheets. We compare
275 our reconstructed 500-kyr-mean CO_2 values with two different estimates of global surface
276 temperature. We apply the same Bayesian inversion model used in the CO_2 reconstruction to
277 derive 500-kyr-mean surface temperatures from the benthic foraminiferal $\delta^{18}\text{O}$ compilation of
278 Ref. (43), which we convert to temperatures using the methodology of Ref. (44) (Fig. 2b). In
279 addition, we pair a set of multiproxy global surface temperature estimates for eight Cenozoic
280 time intervals (Fig. 2b, 45) with posterior CO_2 estimates from time bins corresponding to each
281 interval. The two temperature reconstructions are broadly similar, although the benthic record
282 suggests relatively higher temperatures during the hothouse climate of the Paleocene and
283 Eocene, whereas the multiproxy reconstruction is elevated relative to the benthic record during
284 the Oligocene and Neogene.

285 The co-evolution of atmospheric CO_2 and global mean surface temperature (GMST) over
286 the Cenozoic is shown in Fig. 3. Because CO_2 is on a log scale, the slopes of lines connecting two
287 adjacent points in time reflect the average intervening $\text{ESS}_{[\text{CO}_2]}$. Benthic $\delta^{18}\text{O}$ -derived
288 temperatures suggest early Paleocene warming occurs with a very high $\text{ESS}_{[\text{CO}_2]}$ ($>8^\circ\text{C}$ per CO_2
289 doubling), although CO_2 uncertainties are large during this time interval. $\text{ESS}_{[\text{CO}_2]}$ steadily declines
290 towards the peak of Cenozoic warmth ~ 50 Ma, then steepens again to $\sim 8^\circ\text{C}$ per CO_2 doubling for
291 much of the cooling through to the EOT at ~ 34 Ma. In contrast, the multiproxy global temperature
292 record suggests a lower $\text{ESS}_{[\text{CO}_2]}$ of $\sim 5^\circ\text{C}$ between the early Eocene and earliest Oligocene. During
293 the Oligocene and early part of the Miocene, both temperature records imply a near-zero
294 $\text{ESS}_{[\text{CO}_2]}$, i.e., CO_2 values appear to decline with no appreciable global cooling. $\text{ESS}_{[\text{CO}_2]}$ implied by
295 both temperature reconstructions steepens again from the middle Miocene (~ 16 Ma) to present,
296 averaging 8°C per CO_2 doubling over the past 10 Myr.

297 An alternative perspective on early Cenozoic climate forcing was introduced by Ref. (44),
298 who hypothesized that all pre-Oligocene climate change was the response of direct and indirect
299 CO_2 radiative forcing plus long-term change in solar output (i.e., constant albedo). Given this,
300 they converted Paleocene and Eocene benthic $\delta^{18}\text{O}$ -derived GMST to estimates of CO_2 change
301 required to explain the temperature record. Our reconstruction offers a direct test of this
302 hypothesis, and although it compares well with the $\delta^{18}\text{O}$ approach of Ref. (44) throughout much
303 of the early Cenozoic, our curve suggests that the late Eocene decline in CO_2 was less severe than
304 expected under the constant albedo assumption (Fig. S13). This result is consistent with a
305 growing contribution of glacier and sea ice albedo effects (e.g., 52, 53) and the opening of
306 Southern Ocean gateways (e.g., 54) to climate cooling preceding the Eocene-Oligocene
307 boundary.

308 In summary, the Cenozoic compilation confirms a strong link between CO₂ and GMST
309 across timescales from 500 kyr to tens of Myr, with ESS_[CO₂] generally within the range of 5-8°C –
310 patterns consistent with most prior work (32-34, 45, 51, 55-60), and considerably higher than the
311 present-day ECS of ~3°C. Both temperature reconstructions imply relatively high ESS_[CO₂] values
312 during the last 10 Myr of the Cenozoic, when global ice volumes were highest. This agrees with
313 expectations of an amplified ESS_[CO₂] due to the ice-albedo feedback (61). However, even during
314 times with little-to-no ice (Paleocene to early Eocene), we find elevated values of ESS_[CO₂]
315 (approaching or exceeding 5°C per CO₂ doubling). This implies that fast, non-ice feedbacks, such
316 as clouds or non-CO₂ greenhouse gases (60, 62-65) were probably stronger in the early Paleogene
317 than they are in the present-day climate system (see also 5). The Oligocene to early Miocene is
318 the most enigmatic interval, with an apparent decrease in CO₂ despite relatively stable
319 temperature, implying near zero ESS_[CO₂]. It should be noted that this is one interval where
320 different CO₂ proxies disagree on CO₂ change (Fig. 1a), with relatively stable values from plants
321 but a decline in values from alkenones. More work is needed to confirm these CO₂ and
322 temperature findings, but if these estimates are correct, this could partly reflect transition from
323 a climate state too cold to support the strong fast feedbacks (e.g., clouds) of the early Eocene (5),
324 but not cold enough to generate strong ice-albedo feedback. Tectonic changes in the
325 arrangement of continents and the opening of critical ocean gateways may also be confounding
326 derivation of ESS_[CO₂] at that time (e.g., 49, 54).

327 **Relationship with the evolution of the cryosphere**

328 Our composite CO₂ record also enables reexamination of the evolution of Earth's
329 cryosphere (Fig. 2c) in relation to CO₂ radiative forcing. We use the sea level estimation of Ref.
330 (66) for this comparison because it covers the entire Cenozoic and is somewhat independent of
331 the benthic δ¹⁸O stack (43) used for the GMST derivation in Fig. 2b and also of the more recent
332 sea level reconstruction of Ref. (67). Although there are significant differences between the two
333 sea level estimates, the main features discussed herein are broadly consistent between them.
334 The establishment of a permanent, continent-wide Antarctic ice shield at the EOT (~34 Ma)
335 comes at the end of a ~10-Myr period of generally slowly decreasing CO₂. There is evidence for
336 isolated, unstable Antarctic glaciers at various points over the 10-Myr interval prior to the EOT
337 (50, 53, 66, 68), which is consistent with the increasing paleogeographic isolation of Antarctica
338 and Southern Ocean cooling (54), and CO₂ may have been sufficiently low to enable the repeated
339 crossing of a glaciation threshold by periodic orbital forcing. Tectonic cooling of Antarctica would
340 have progressively raised the CO₂ glaciation threshold, which has been modeled to be within 560-
341 920 ppm (69, 70). Our composite CO₂ record allows us to further assess this glaciation threshold
342 but requires determining the point during glacial inception when strong positive feedbacks (e.g.,
343 ice-albedo and ice sheet elevation) commenced and ice sheet growth accelerated (71). Using the
344 sea level curve of Ref. (66), we determine this point as 33.75 ± 0.25 Ma, where our composite CO₂
345 record suggests 719 $\frac{+180}{-152}$ ppm (95% CIs). Once established, the land-based Antarctic ice sheet
346 likely persisted for the remainder of the Cenozoic, although significant retreat of land-based ice
347 has been modeled (30-36 m sea level equivalent, 72) and estimated from proxies (Fig. 2c) for the
348 Miocene Climatic Optimum (MCO). 500-kyr-mean CO₂ values increased to ~500 ppm during the
349 MCO (Figs. 2a, S10), and benthic foraminiferal δ¹⁸O (Fig. 2b, 43) and clumped isotopes (50)

350 indicate warming. While the stability of the land-based Antarctic ice sheet depends on many
351 factors in addition to CO₂-induced global warming (e.g., hysteresis (73), bed topography (74)),
352 our composite record indicates that significant retreat of land-based ice did not occur below 441-
353 480 ppm (2.5-50 percentiles), and some land-based ice may have persisted up to 563 ppm (97.5
354 percentile) during the MCO. Excepting the MCO, atmospheric CO₂ has remained below our
355 current value of 419 ppm since the late Oligocene (Figs. 2a, S10), with relatively small sea-level
356 variations (up to ~20m, Fig. 2c and 67) being driven by orbitally-forced melting of the marine-
357 based ice sheet (e.g., 72, 75). Finally, at ~2.7 Ma, the transition to intensified northern
358 hemisphere glaciation and orbitally-driven glacial cycles coincided with CO₂ values that began
359 decreasing after a relative high during the Pliocene (Fig. 2a).

360

361 **Evolutionary implications of the revised CO₂ curve**

362 While geologic trends in terrestrial floral and faunal habitat ranges (e.g., 76, 77) and diversity
363 (e.g., 78, 79, 80) are largely thought to be controlled by temperature and associated climate
364 patterns, atmospheric CO₂ has been hypothesized to drive the evolution of biological carbon
365 concentrating mechanisms and their subsequent diversification in terrestrial plants (CCMs, Fig.
366 2d, 81, 82). Our realization of how atmospheric CO₂ has varied through the Cenozoic allows us to
367 re-examine this hypothesis. The two primary CCMs in terrestrial plants are the crassulacean acid
368 metabolism (CAM) and C₄ photosynthetic syndromes. CCMs in terrestrial C₄ and CAM plants
369 confer competitive advantages over the ancestral C₃ pathway under higher growing season
370 temperatures, low rainfall, and lower atmospheric CO₂. As a result, C₄ photosynthesis contributes
371 about 23% of today's global terrestrial gross primary production (GPP, 83).

372 Plant clades with the C₄ pathway first emerged in the early Oligocene (84, 85), yet did not
373 expand to ecological significance until the late Miocene (i.e., <5% GPP before ~10 Ma, Fig. 2d, 86,
374 87, 88). CAM plants (e.g., cacti, ice plants, agaves, and some orchids) underwent significant
375 diversification events around the late Oligocene and late Miocene (89-91). Taken together, two
376 general biological thresholds emerge based on our CO₂ record: (1) All known origins of C₄ plants
377 occurred when atmospheric CO₂ was lower than ~550 ppm (i.e., after 32 Ma, Fig. 2a,d, 84), which
378 is in agreement with theoretical predictions (92, 93). (2) All major Cenozoic CAM diversification
379 events coincided with intervals when CO₂ was lower than ~430 ppm (i.e., after 27 Ma, 89, 90).
380 Our record is thus consistent with decreasing atmospheric CO₂ (< 550 ppm) being a critical
381 threshold for the Cenozoic origin, diversification, and expansion of C₄ and CAM plants within
382 grasslands, arid habitats (such as deserts), and habits (such as epiphytes), and provides strong
383 data support for previous hypotheses (20, 84, 86, 88, 89, 92, 94, 95). Importantly, following their
384 origin in the early Oligocene, C₄ plants did not immediately proliferate. By ~24 to ~18 Ma, open
385 habitat grasslands are evident on most continents (96), yet widespread dispersal of C₄ plants was
386 delayed until the late Miocene, and without any apparent decline in CO₂ (Fig. 2d). Therefore, the
387 rise of C₄ plants to their dominance in many tropical and subtropical ecosystems was likely driven
388 (and maintained today) by other factors such as fire, seasonality of rainfall, and herbivory (i.e.,
389 grazing that keeps landscapes open) (97, 98). The temporal evolution of these factors warrants
390 further study as we move towards a future where CO₂ may rise above the 550-ppm threshold
391 that was key to the origin, taxonomic diversification, and spread of C₄ plants.

392

393 Terrestrial mammals evolved and adapted to the changing and more open floral
394 ecosystems of the late Cenozoic (99-101), and are thus indirectly linked to the 550-ppm
395 atmospheric CO₂ threshold discovered herein. In particular, dental wear patterns (such as the
396 shape of the chewing surface of a tooth, i.e., mesowear) and tooth morphology, such as crown
397 height, reflect an increasingly abrasive and tough diet (102, 103), and can be traced across many
398 herbivore lineages during this period. For instance, mesowear in North American Equidae (horses
399 and their ancestors, Fig. 2d) began to increase in the late Eocene, and steadily continued into the
400 Quaternary. Similarly, equids evolved high-crowned (hypsodont) teeth in the Miocene (103-105),
401 and their body size increased to accommodate higher intake of more abrasive, grassy vegetation
402 (Fig. 2d).

403 Evolutionary trends are a little less clear in the ocean, because marine algal CCMs are
404 ubiquitous and diverse in form (106) and are believed to have an ancient origin. Moreover, the
405 large spatial and seasonal variance of dissolved CO₂ in the surface ocean (as compared to the
406 relatively uniform seasonal and spatial concentration of CO₂ in the air) may somewhat decouple
407 their evolution from geologic trends in atmospheric CO₂. Evidence exists that marine algae, and
408 in particular the coccolithophores (i.e., the source of the alkenone biomarkers), express CCMs to
409 greater extent when CO₂ is lower (e.g., 107, 108, 109), with estimates of cellular carbon fluxes
410 suggesting enhanced CCM activity in coccolithophores began ~7-5 Ma (110). However, our
411 revised CO₂ curve displays mean atmospheric CO₂ broadly constant at 300-350 ppm since at least
412 ~14 Ma (Figs. 2a, S10), suggesting that increased CCM activity may reflect other proximal triggers,
413 perhaps involving changes in ocean circulation and nutrient supply.

414

415 **Perspectives and opportunities for further advances**

416 Our community-assessed composite CO₂ record and statistically modelled time-averaged
417 CO₂ curve exhibit greater clarity in the Cenozoic evolution of CO₂ and its relationship with climate
418 than was possible in previous compilations, and furthermore highlight the value of cross-
419 disciplinary collaboration and community building. Generating a paleo-CO₂ record with even
420 greater confidence requires targeted efforts using multiple proxies to fill in data gaps, higher
421 resolution and replication from multiple locations, and novel approaches to resolve remaining
422 differences between CO₂ proxy estimates. Specifically: although the number and diversity of
423 paleo-CO₂ proxy records continues to grow, data remain relatively sparse during several key parts
424 of the Cenozoic record (e.g., middle Paleocene, Oligocene). Moreover, records from the
425 Paleocene and Eocene are dominated by estimates from the boron isotope proxy, increasing
426 potential for bias. Targeted efforts are hence needed to expand the number and diversity of data
427 through these intervals and to refine multi-proxy reconstructions. Secondly, despite substantial
428 progress, there remains a lack of consensus regarding the identity and/or quantification of some
429 of the factors underlying each of the proxy systems analyzed here. New experimental and
430 calibration studies, particularly those that isolate and quantify specific mechanistic responses
431 and/or their interactions, need to be undertaken in order to reduce potential biases and
432 uncertainty for each method. For instance, the emerging fields of genomics, evolutionary and
433 developmental biology, and proteomics provide exciting new opportunities for improving and

434 understanding paleo-proxy systematics. Thirdly, and associated with improved experimental
435 quantification, refining our theoretical and mechanistic understanding of how proxies are
436 encoded will allow us to create explicit and self-consistent representations of the processes
437 involved. The development of proxy system forward models provides a promising leap in this
438 direction (e.g., 111). Bayesian statistical methods can then enable the full suite of models and
439 data to be integrated and constrain the range of environmental conditions, including
440 atmospheric CO₂ and other variables that are consistent with the multiproxy data (112, 113).
441 Finally, development of new proxies is also a realistic and desirable aim. For instance, while this
442 study focuses on more established proxies, new proxies such as coccolith calcite stable isotopes
443 (114) and mammalian bone and teeth oxygen-17 anomalies (115) show promising results for
444 reconstructing paleo-CO₂, but perhaps require further validation before they can be assessed
445 with confidence.

446 Proxies and proxy-based reconstructions of how atmospheric CO₂ has varied through
447 deep time have improved immeasurably over the past few decades. While they will never allow
448 us to reconstruct past CO₂ with the same fidelity as direct air measurement, our study shows how
449 community-based consensus assessment, together with a critical reanalysis of proxy models and
450 assumptions, can progressively move us towards a quantitative history of atmospheric CO₂ for
451 geological time.

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References and Notes

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721

722 **Data and materials availability:** The completed data sheets for each study can be accessed as the
723 [paleo-CO₂ 'Archive'](#) at NOAA's National Center for Environmental Information (NCEI). The
724 specific choice of category, as well as the updated CO₂ and age estimates, are documented in
725 'Product' sheets for each data set and proxy. In contrast to the 'Archive', which will grow with new
726 publications but will otherwise remain passive, the paleo-CO₂ 'Product' is a living database that
727 will be updated when newly published data or ancillary data constraints become available, and/or
728 methodological improvements are developed that enable modernization of previously
729 underconstrained datasets. The **Product** sheets created for this study can be accessed in NCEI, and
730 this is also the place where future data updates will be made available in consecutive versions of
731 the data 'Product'.

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822 **List of Supplementary Materials**

823 Supplementary Text, Sections 1-10, Figs. S1 to S13

824 Tables S1 to S3

825 References (118-439)

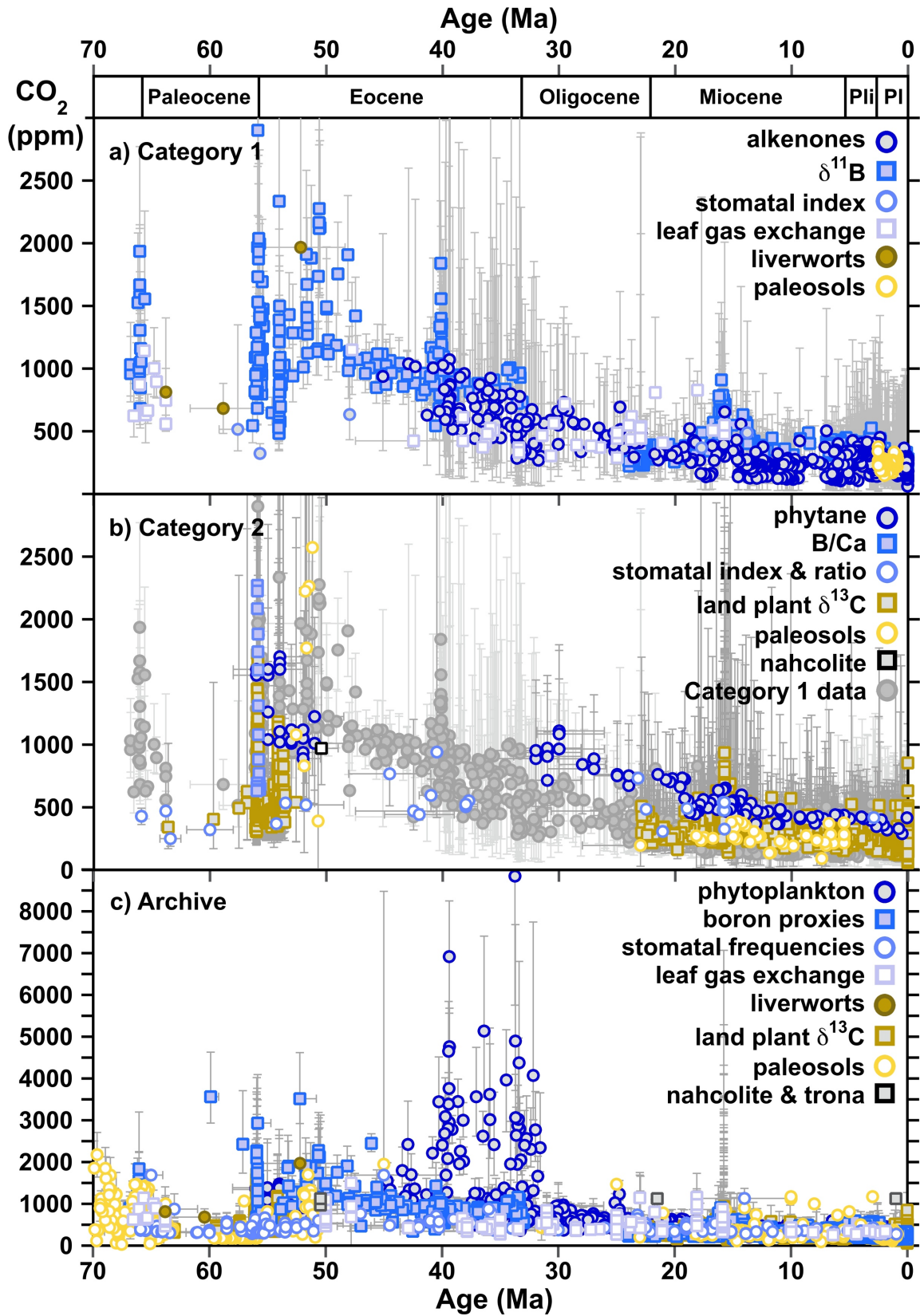
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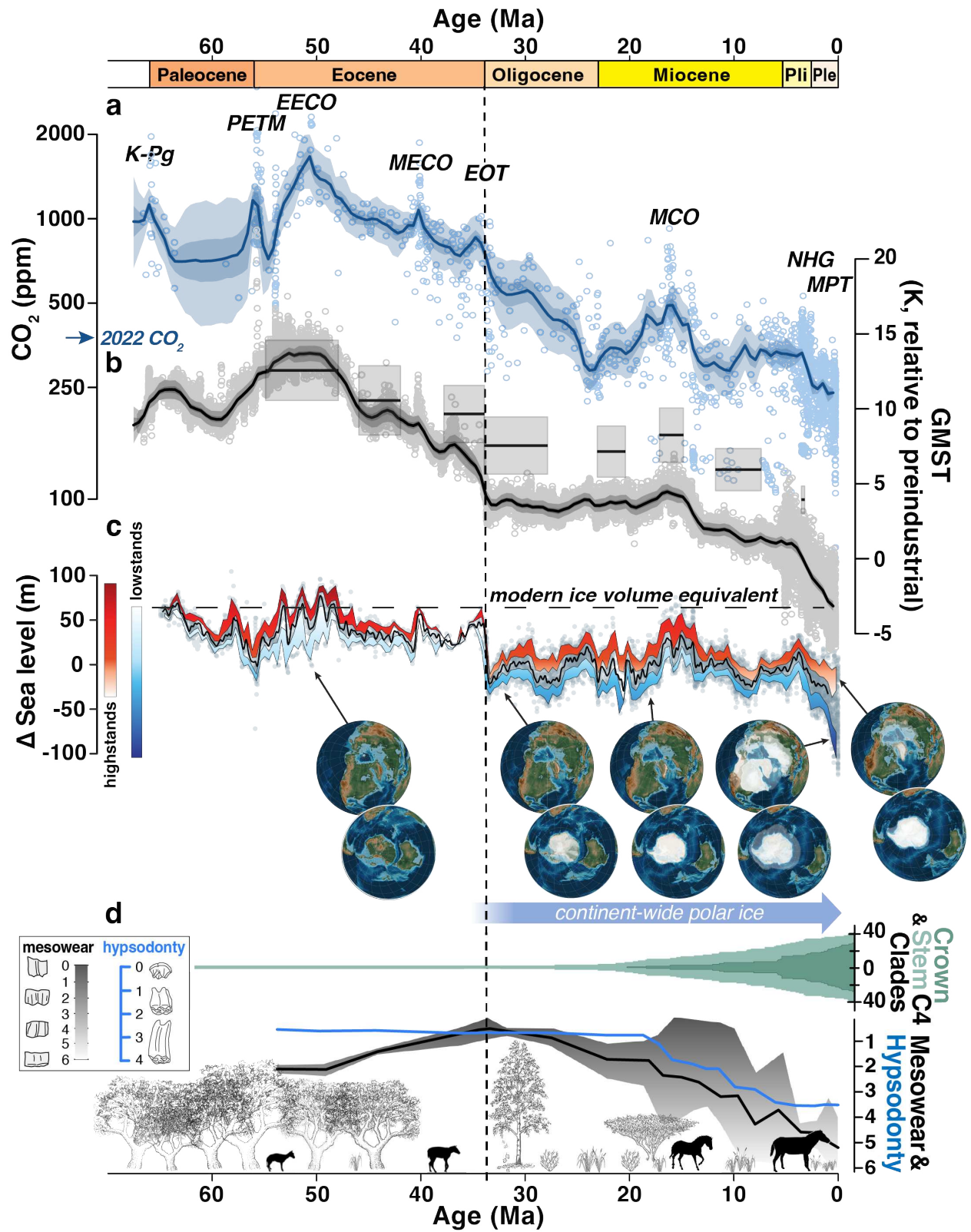
828 **Figures :**

829 **Fig. 1. Documentation and assessment of all Cenozoic paleo-CO₂ estimates published to**
830 **date.** Individual proxy estimates are defined by colored symbols in legends. **(a)** Vetted Category
831 1 estimates with their fully developed uncertainty estimates (95% CIs); age uncertainties have
832 been updated or established to the best of current understanding. **(b)** Vetted Category 2 estimates
833 whose uncertainty is not yet fully constrained. Category 1 data are shown in grey for reference.
834 **(c)** Archive compilation of all CO₂ estimates in their originally published quantification. To
835 toggle view of individual proxy records in panels (a) and (c), please go to paleo-co2.org (Note:
836 panel (a) visualization will be published on the website after acceptance of the manuscript for
837 publication).

838



840 **Fig. 2. Category 1 paleo-CO₂ record compared to global climate signals.** The vertical dashed
841 line indicates the onset of continent-wide glaciation in Antarctica. **(a)** Atmospheric CO₂
842 estimates (symbols) and 500-kyr mean statistical reconstructions (median and 50 and 95%
843 credible intervals - dark and light-blue shading, respectively). Major climate events are
844 highlighted (K-PG - Cretaceous/Paleogene boundary, PETM - Paleocene Eocene Thermal
845 Maximum, EECO - Early Eocene Climatic Optimum, MECO - Middle Eocene Climatic
846 Optimum, EOT - Eocene/Oligocene Transition, MCO - Miocene Climatic Optimum, NHG -
847 onset of Northern Hemisphere Glaciation, MPT - Mid Pleistocene Transition). The 2022 annual
848 average atmospheric CO₂ of 419 ppm is indicated for reference. **(b)** Global mean surface
849 temperatures estimated from benthic $\delta^{18}\text{O}$ data after Westerhold et al. (43) (solid line, individual
850 proxy estimates as symbols, and statistically reconstructed 500-kyr mean values shown as the
851 continuous curve, with 50 and 95% credible intervals) and from surface temperature proxies (45)
852 (grey boxes). **(c)** Sea level after Ref. (66) with gray dots displaying raw data; the solid black line
853 reflects median sea level in a 1-Myr running window. High- and lowstands are defined within a
854 running 400-kyr window, with lower and upper bounds of highstands defined by the 75th and
855 95th percentiles, and lower and upper bounds of lowstands defined by the 5th and 25th
856 percentiles in each window. Globes depict select paleogeographic reconstructions and the
857 growing presence of ice sheets in polar latitudes from Ref. (116). **(d)** Crown ages show C₄
858 clades, with CCMs adapted to low CO₂, initially diversified in the early Miocene and then
859 rapidly radiated in the late Miocene (117). Flora transition from dominantly forested and
860 woodland to open grassland habitats based on fossil phytolith abundance data (96). North
861 American equids typify hoofed animal adaptations to new diet and environment (103), including
862 increasing tooth mesowear (black line, note inverted scale), hypsodonty (blue line), and body
863 size.
864



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866

867 **Fig. 3. Application of the Category 1 CO₂ record to determine ESS_[CO₂].** GMST deviation
868 (K) from preindustrial global average surface temperature of 14.15°C is displayed versus paleo-
869 CO₂ doublings relative to the preindustrial baseline of 280 ppm (upper x-axis) and paleo-CO₂
870 estimates on a log scale (lower x-axis). The slopes between two points in time reflect the average
871 ESS_[CO₂]. Circles reflect 500-kyr binned 'Category 1' CO₂ estimates paired with corresponding
872 GMST-means from Ref. (43), squares pair CO₂ and GMST means from compilations of sea
873 surface temperature (45) in seven coarsely resolved time intervals. Note that this figure omits the
874 Pliocene temperature estimate of (45) because it samples too short a time interval (cf. Fig. 2) to
875 be comparable with mean CO₂. Data from Cenozoic epochs are color coded and shift from red
876 (Paleocene) to yellow (Pleistocene); labels indicate specific age bins (Ma). Dashed lines indicate
877 reference ESS_[CO₂] lines of 8 and 5°C warming per doubling of CO₂.
878

