

1. Age model for site MD95-2037 over Termination II

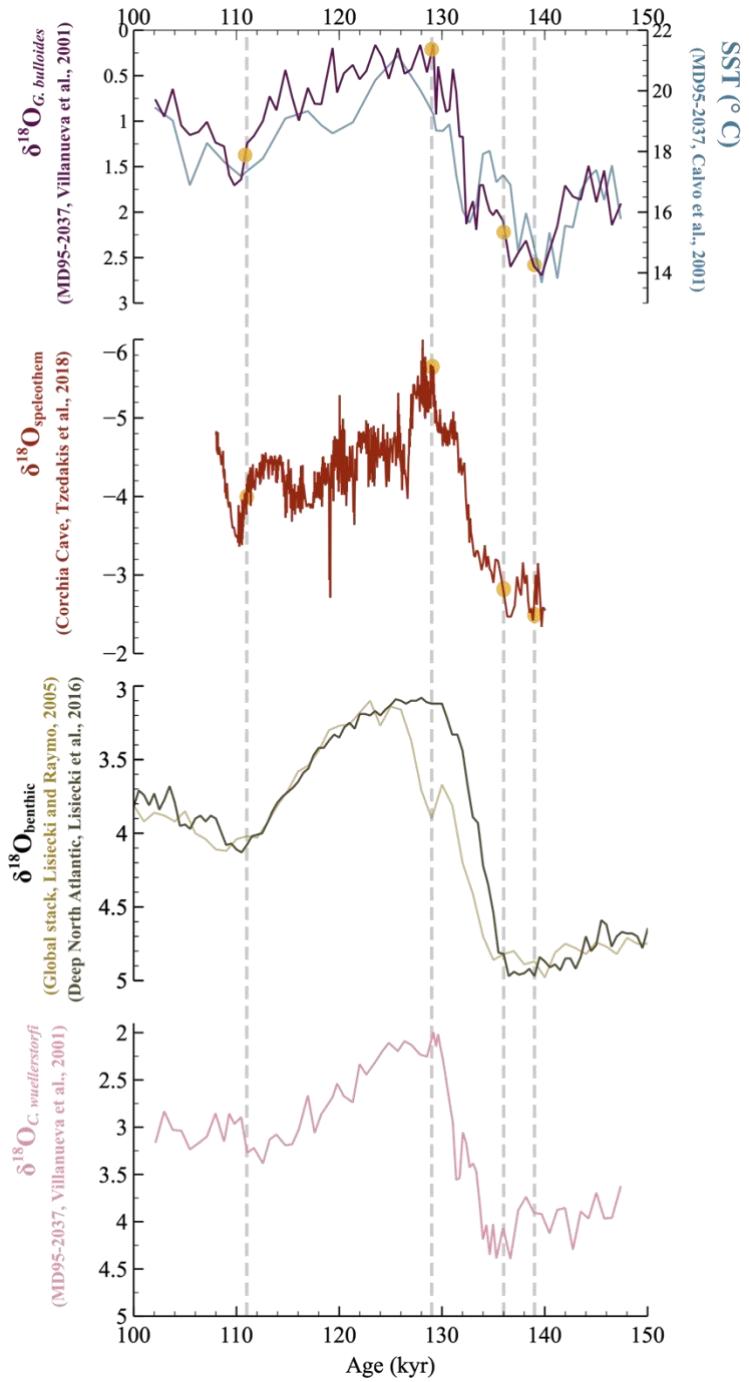


Figure S1: Synchronizing the MD95-2037 Termination II $\delta^{18}\text{O}_{\text{planktonic}}$ record with speleothem $\delta^{18}\text{O}$ records. We first aligned the *Cibidoides wullerstorfi* $\delta^{18}\text{O}$ signal of site MD95-2037 (Villanueva et al., 2001) to the Regional Deep North Atlantic $\delta^{18}\text{O}$ Benthic Stack (Lisiecki and Stern, 2016) for the first order trends. The age model around Termination II was refined by aligning the $\delta^{18}\text{O}_{\text{bulloides}}$ signal of site MD95-2037 (Villanueva et al., 2001) to the Corchia Cave composite speleothem $\delta^{18}\text{O}$ record (Tzedakis et al., 2018). Tie-points are represented as yellow dots. For comparison, we also included the Global Benthic $\delta^{18}\text{O}$ Stack (Lisiecki and Raymo, 2005) and SST records for the site (Calvo et al., 2001).

2. Fraction composition and preservation

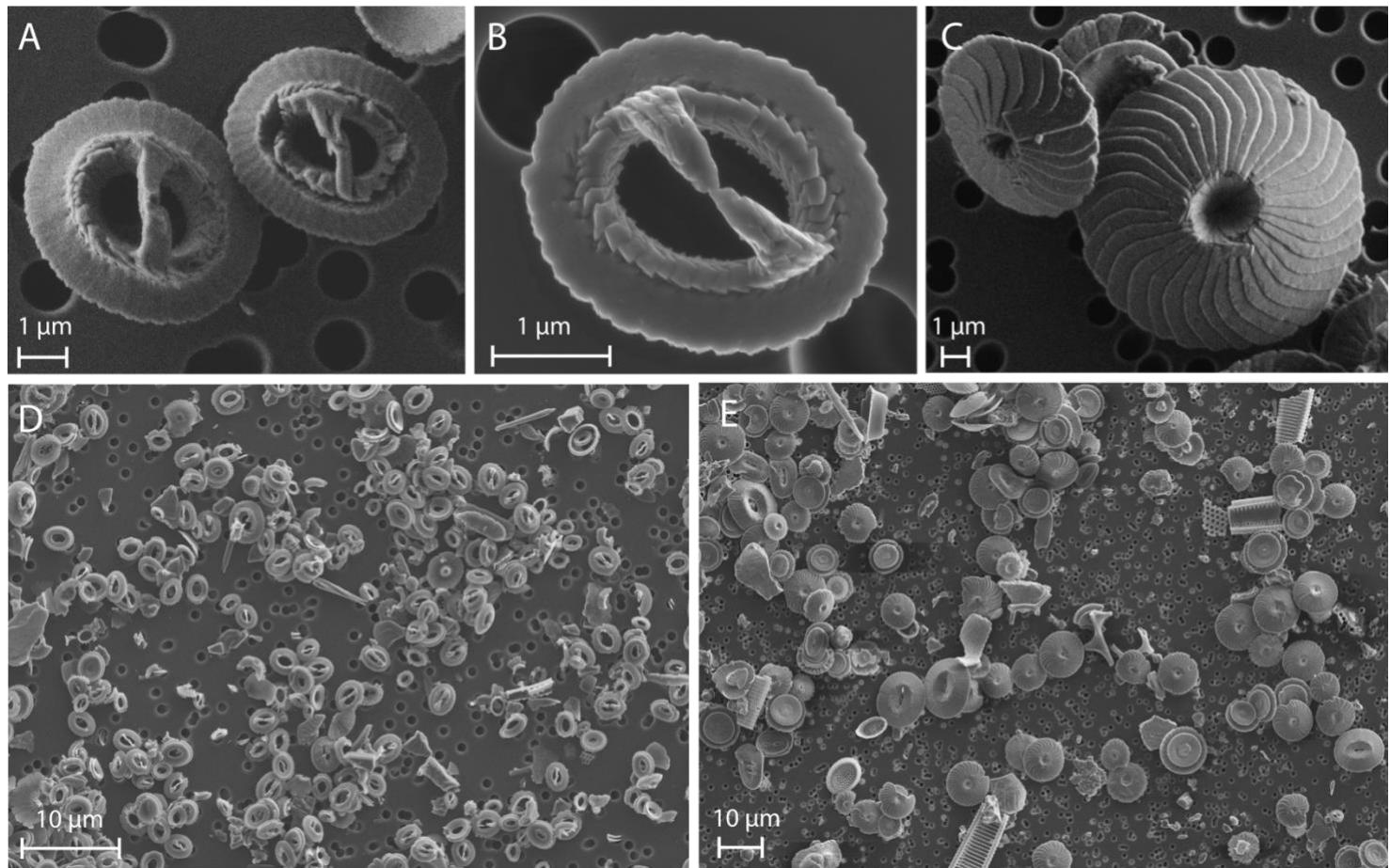


Figure S2: Coccolith close-ups and general views of the purified coccolith assemblages obtained for core MD95-2037 sediments. A: *Gephyrocapsa* sp. coccoliths at 123.5 kyrs (Interglacial). B: *Gephyrocapsa* sp. coccolith at 139.7 kyrs (Glacial). C: *Calcidiscus* sp. coccoliths at 123.5 kyrs (Interglacial). D: *Gephyrocapsa*-dominated 2-3 μm fraction at 110.5 kyrs (Last Glacial Inception). E: *Calcidiscus*-dominated 5-8 μm fraction at 135.9 kyrs (Glacial Maximum). Images were obtained using the SEM at the ISTeP Lab. The coccoliths exhibit slight etching, but no notable calcite overgrowth. The silicic material observed in the assemblages does not contribute to the isotopic signal measured on the sediment fractions.

3. Data used for the Miocene-to-present differential vital effect- CO_2 curve

Published literature provides other examples of the forcing of CO_2 on differential coccolith vital effects. We selected the datasets that provided a differential vital effect between coccolith size classes with similar diameters to those studied in this manuscript. These values are compared to the corresponding pCO_2 records available for the time period. These are translated, using the SST records available for the core location, and a salinity of 35 psu, into concentrations of aqueous CO_2 , via the “seacarb” package in R (<https://CRAN.R-project.org/package=seacarb>). Error estimates (1SD) for $[\text{CO}_2]$ were obtained by running 10,000 Monte Carlo simulations with the following uncertainties: For the Miocene/Pliocene pCO_2 record we took the uncertainties (1SD) of Rae et al., 2021, which enables to account for increasing uncertainties on pCO_2 reconstructions as age increases; We considered a ± 10 ppm uncertainty on pre-industrial pCO_2 to account for age uncertainties on core-top material; We considered a $\pm 1.5^\circ\text{C}$ uncertainty on Miocene/Pliocene SSTs (Conte et al., 2006), a conservative uncertainty of $\pm 1.0^\circ\text{C}$ on core-top SSTs, and a conservative estimate of salinity of ± 1 psu throughout. We listed below the published datasets used to study the response of coccolith differential vital effects to aqueous CO_2 changes since the Miocene:

Table S1: Datasets used to compare the evolution of coccolith differential vital effects and aqueous CO_2 changes since the Miocene.

Differential vital effect datasets	Fraction size and composition	Period, Site location for coccolith samples	pCO_2 data	SST data	Site location for SST data
Bolton et al., 2012	2-4 μm (<i>Reticulofenestra</i> sp.-rich) and 6-9 μm (<i>Helicosphaera</i> sp. and <i>Discoaster</i> sp.-rich)	Miocene-Pliocene, ODP site 999 (Caribbean)	Rae et al., 2021	Composite record from Rae et al., 2021 (largely based on Mg/Ca values)	Equatorial sites mainly
Bolton et al., 2012	2-5 μm (rich in small reticulofenestrids) and 7-9 μm (rich in reticulofenestrids)	Miocene-Pliocene, site 1088 (South Atlantic)	Rae et al., 2021	Herbert et al., 2016 Alkenone-Uk'	Same as core location
Candelier et al., 2013 (large fraction); Hermoso et al., 2015 (small fraction)	2-3 μm (<i>Gephyrocapsa</i> sp.-rich) and 5-8 μm (<i>Calcidiscus leptoporus</i> -rich)	Core-top (North Atlantic, Western Indian Ocean)	Pre-industrial 280 ppm value	Hermoso et al., 2015, from WOA13	Same as core location

4. Controls on coccolith differential vital effects

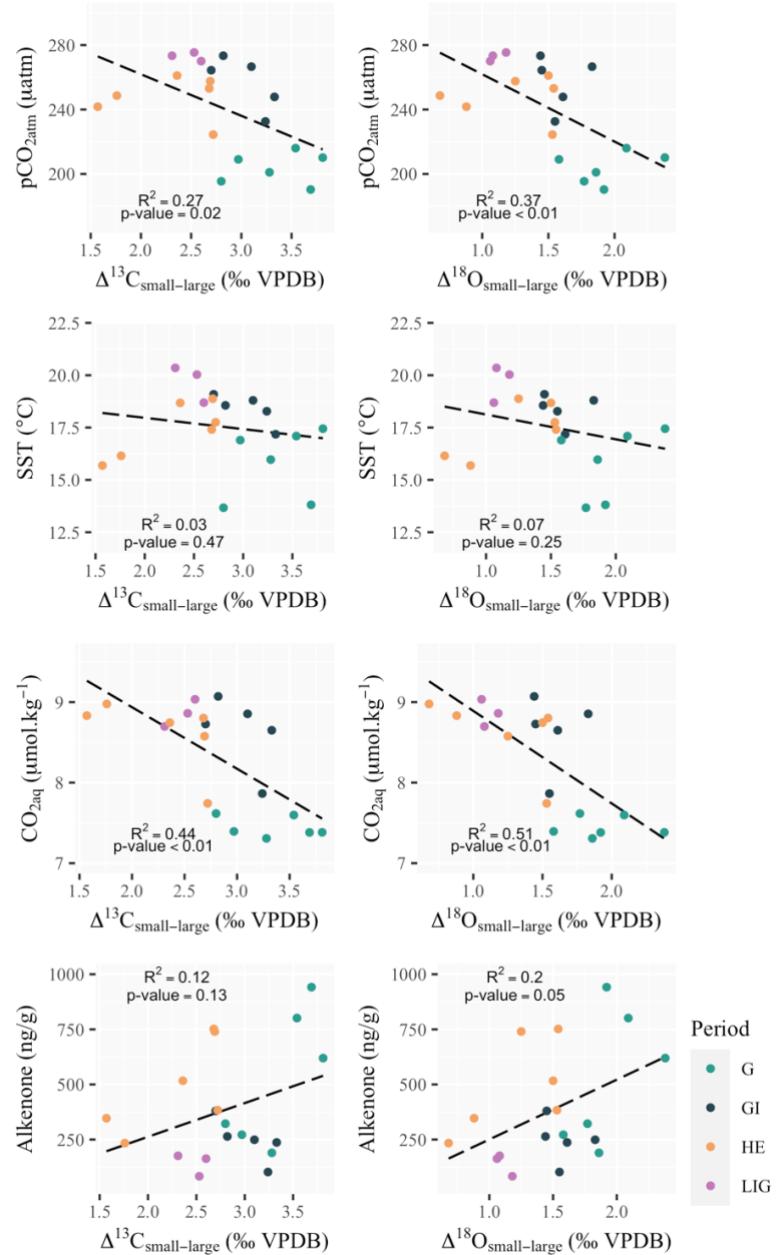


Figure S3: Environmental controls on coccolith differential vital effects. Data points are coloured according to the time period considered: The Glacial Maximum (green), Heinrich Event 11 (orange), the Last Interglacial (pink) and the Glacial Inception (dark blue). Ice core pCO_2 data is from Bereiter et al., 2015, Sea surface temperature (SST) data are from Calvo et al., 2001 for site MD95-2037, and alkenone concentrations are from Villanueva et al., 2001 for site MD95-2037.

Supplementary references

- Bereiter, B., Eggleston, S., Schmitt, J., Nehrbass-Ahles, C., Stocker, T. F., Fischer, H., Kipfstuhl, S., and Chappellaz, J.: Revision of the EPICA Dome C CO₂ record from 800 to 600 kyr before present, *Geophys. Res. Lett.*, 42, 542–549, <https://doi.org/10.1002/2014GL061957>, 2015.
- Bolton, C. T., Stoll, H. M., and Mendez-Vicente, A.: Vital effects in coccolith calcite: Cenozoic climate- pCO₂ drove the diversity of carbon acquisition strategies in coccolithophores?, *Paleoceanography*, 27, 1–16, <https://doi.org/10.1029/2012PA002339>, 2012.
- Calvo, E., Villanueva, J., Grimalt, J. O., Boelaert, A., and Labeyrie, L.: New insights into the glacial latitudinal temperature gradients in the North Atlantic. Results from U37K' sea surface temperatures and terrigenous inputs, *Earth Planet. Sci. Lett.*, 188, 509–519, [https://doi.org/10.1016/S0012-821X\(01\)00316-8](https://doi.org/10.1016/S0012-821X(01)00316-8), 2001.
- Candelier, Y., Minoletti, F., Probert, I., and Hermoso, M.: Temperature dependence of oxygen isotope fractionation in coccolith calcite: A culture and core top calibration of the genus *Calcidiscus*, *Geochim. Cosmochim. Acta*, 100, 264–281, <https://doi.org/10.1016/j.gca.2012.09.040>, 2013.
- Conte, M. H., Sicre, M. A., Röhleemann, C., Weber, J. C., Schulte, S., Schulz-Bull, D., and Blanz, T.: Global temperature calibration of the alkenone unsaturation index (U 37k) in surface waters and comparison with surface sediments, *Geochemistry, Geophys. Geosystems*, 7, <https://doi.org/10.1029/2005GC001054>, 2006.
- Herbert, T. D., Lawrence, K. T., Tzanova, A., Peterson, L. C., Caballero-Gill, R., and Kelly, C. S.: Late Miocene global cooling and the rise of modern ecosystems, *Nat. Geosci.*, 9, 843–847, <https://doi.org/10.1038/ngeo2813>, 2016.
- Hermoso, M., Candelier, Y., Browning, T. J., and Minoletti, F.: Environmental control of the isotopic composition of subfossil coccolith calcite: Are laboratory culture data transferable to the natural environment?, *GeoResJ*, 7, 35–42, <https://doi.org/10.1016/j.grj.2015.05.002>, 2015.
- Lisiecki, L. E. and Raymo, M. E.: A Pliocene-Pleistocene stack of 57 globally distributed benthic δ 18 O records, *Paleoceanography*, 20, 1–17, <https://doi.org/10.1029/2004PA001071>, 2005.
- Lisiecki, L. E. and Stern, J. V.: Regional and global benthic δ 18 O stacks for the last glacial cycle, *Paleoceanography*, 31, 1368–1394, <https://doi.org/10.1002/2016PA003002>, 2016.
- Rae, J. W. B., Zhang, Y. G., Liu, X., Foster, G. L., Stoll, H. M., and Whiteford, R. D. M.: Atmospheric CO₂ over the Past 66 Million Years from Marine Archives, *Annu. Rev. Earth Planet. Sci.*, 49, 609–641, <https://doi.org/10.1146/annurev-earth-082420-063026>, 2021.
- Tzedakis, P. C., Drysdale, R. N., Margari, V., Skinner, L. C., Menviel, L., Rhodes, R. H., Taschetto, A. S., Hodell, D. A., Crowhurst, S. J., Hellstrom, J. C., Fallick, A. E., Grimalt, J. O., McManus, J. F., Martrat, B., Mokeddem, Z., Parrenin, F., Regattieri, E., Roe, K., and Zanchetta, G.: Enhanced climate instability in the North Atlantic and southern Europe during the Last Interglacial, *Nat. Commun.*, 9, <https://doi.org/10.1038/s41467-018-06683-3>, 2018.

Villanueva, J., Calvo, E., Pelejero, C., Grimalt, J. O., Boelaert, A., and Labeyrie, L.: A latitudinal productivity band in the Central North Atlantic over the last 270 kyr: An alkylene perspective, *Paleoceanography*, 16, 617–626, <https://doi.org/10.1029/2000PA000543>, 2001.