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Carbon isotope fractionation during photosynthesis: New constraints on marine algal paleobarometry

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Better estimates of past carbon dioxide levels in the atmosphere (pCO_2) are critical to the advancement of climate science. While instrumental and ice-core records enable understanding of the current radiative forcing, geochemical archives offer insights into ancient pCO_2. A recent synthesis of Cenozoic proxy records both improved our understanding and exposed continuing uncertainties in these records[1]. Phytoplankton carbon isotope discrimination (p ≈ $\delta^{13}C_{CO2(aq)}$ – $\delta^{13}C_{phyto}$), i.e., algal paleobarometry, is one of the most frequently used proxy methods. Estimating CO_2 from p requires quantifying the relationship between the supply of carbon from seawater and the intracellular demand for that carbon, using the assumptions that p is set by the ratio of diffusive demand:supply, and that the fractionation primarily is governed by the kinetic isotope effect (KIE) of the carbon-fixing enzyme RuBisCO. The most widely used algal proxy uses the long-chain unsaturated ketones (“alkenones”) unique to the Noelaerhabdaceae, commonly represented by the widespread modern taxon, *Emiliania huxleyi*. Recent work on the KIE and kinetics of *E. huxleyi* RuBisCO[2] in tandem with environmental and laboratory studies[3-5] have significantly altered the standard view, suggesting that photosynthetic irradiance independently influences p, and/or that non-diffusive supply of CO_2 is common. These factors may explain why alkenone p does not accurately preserve Late Pleistocene glacial cycles[6-7]. Current efforts aim to address these discrepancies, both through models that incorporate additional intracellular compartmentalization of inorganic carbon[8], and through direct experiments with pure cultures (this work). Here we show that photon flux alone can cause significant changes in p in *E. huxleyi* grown under constant CO_2 conditions in a nutrient-limited chemostat. The results point toward future strategies to refine pCO_2 reconstructions and improve climate models.

[1] Hoenisch et al., (2023) *Science* 382:1136; [2] Boller et al., (2011) *GCA* 75:7200-7207; [3] Rost et al., (2002) *Limnol Oceanogr* 47:120–128; [4] Hernandez-Almeida et al., (2020) *G-cubed* 21:e2020GC009272; [5] Phelps et al. (2021) *G-cubed* 22:e2021GC009658; [6] Badger et al., (2019) *Clim Past* 15:539-554; [7] Zhang et al. (2019) *GCA* 260:177-191; [8] Wilkes and Pearson (2019) *GCA* 265, 163-181.