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Can stable isotope fractionation in diatom and coccolith biominerals elucidate the significance of carbon concentrating mechanisms (CCMs) in the past?

H. Stoll, C. Bolton, K. Isensee, A. Mendez-Vicente, M. Rubio-Ramos, and L.M. Mejia-Ramirez
University of Oviedo, Geology, Oviedo, Spain (hstoll@geol.uniovi.es)

Carbon isotopic fractionation in fossil algal biomarkers is typically interpreted to reflect atmospheric CO_2 changes assuming simple diffusive uptake of CO_2 by cells, however modern algae employ a diverse array of additional strategies to concentrate DIC inside the cell (CCMs). We previously hypothesized that the size-correlated range of vital effects in carbonate liths produced by different coccolithophore species was due to variable significance of CCMs in their C acquisition, and that an absence of interspecific vital effects may reflect a reduced importance of CCMs (or more similar CCMs employed). Here, we present stable isotope data from size-separated deep-sea sediments dominated by small, intermediate and large coccoliths from time slices throughout the Cenozoic. We show that the range of coccolith vital effects is distinct during several major Cenozoic proxy-inferred climate-CO_2 transitions, and where vital effects are significant their magnitude scales with cell size in the same sense as modern culture genera (increasing C and O isotope enrichment with decreasing coccolith size). Our new culture experiments with coccolithophorids reveal strong plasticity in the magnitude of stable carbon isotope vital effects in coccoliths of Calcidiscus leptoporus and Emiliania huxleyi with variable CO_2. At high CO_2 coccoliths of both species are more isotopically enriched, but the magnitude is greater in C. leptoporus leading to reduced interspecific offsets at high CO_2. In the case of E. huxleyi, higher CO_2 conditions resulted in significant reduction in the magnitude of DIC accumulation in the intracellular carbon pool, and more positive carbon isotopic values inside the particulate organic matter. A model of carbon acquisition incorporating both photosynthetic and carbonate production is used to explore mechanisms for these relationships. We also investigate fractionation in diatom organic matter and diatom biomineral-bound organic matter. While the carbon isotopic fractionation in Thalassiosira pseudonana follows the classic dependency on the ratio between carbon supply (CO_2) and demand (growth rate), fractionation in T. weissflogii does not, suggestive of different carbon acquisition strategies in this diatom as suggested by previous workers.