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The Cenozoic emergence of stable isotope 'vital effects' in coccolith calcite

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Long term adaptation of phytoplankton to decreasing carbon dioxide (CO_2) has resulted in modern algae capable of actively enhancing CO_2 at the site of photosynthesis. Coccolithophores, calcifying marine algae of the class Prymnesiophyceae, uniquely preserve the geological history of this adaptation because the stable carbon and oxygen isotopic compositions (δ18O and δ13C) of their calcite plates (coccoliths) are sensitive to active carbon uptake and transport by the cell. Cultures of coccolithophores grown under ambient, CO_2-limiting conditions show an unusually large array (up to 5 ‰ of size-correlated δ18O and δ13C compositions or 'vital effects', which are also evident in coccoliths from Plio-Pleistocene sediments. Conversely, the isotopic difference between small and large coccoliths diminishes in cultures grown at elevated CO_2 and is absent in fossil coccoliths from past Palaeocene greenhouse climates. Here, we constrain the detailed timing of the emergence of coccolith vital effects in the fossil record and its relationship to Cenozoic climate evolution and the long-term decrease in pCO_2. We present new records of δ18O and δ13C from size-separated fossil coccoliths over a number of key Cenozoic climate transitions and document a step increase in the range of vital effects between small and large coccoliths during an interval of significant global cooling at two widely spaced sites. Using a new cellular model, we show that the emergence of coccolith vital effects in the geological record could stem from a threshold adaptive response of cells to CO_2 limitation.