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Combined Stable Ca, Sr, and O Isotope Records Reveal Coccolithophore Calcification Stress Before and Across the PETM

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Rapid CO_2 release caused higher temperatures, ocean acidification (OA), and shoaling of the carbonate compensation depth during the Paleocene Eocene Thermal Maximum (PETM, ~56 Ma). Planktic foraminifera and coccolithophores also experienced turnover, migration, and impaired calcification. IODP Expedition 378 recently recovered a continuous succession of deep-sea pelagic facies spanning the PETM at Site U1553 on the Campbell Plateau in the South Pacific Ocean. The PETM interval comprises micritic limestone primarily derived from coccolith calcite. We generated high-precision stable calcium and strontium isotope ($\delta^{44/40}$Ca and $\delta^{88/86}$Sr) records for U1553 bulk carbonate, as well as δ^{13}C, δ^{18}O, Sr/Ca, and ^{87}Sr/^{86}Sr records to locate the Paleocene/Eocene boundary and investigate environmental change during the event. The novel "$\delta^{44/40}$Ca-$\delta^{88/86}$Sr multi-proxy" is sensitive to primary kinetic isotope effects, chemical weathering dynamics, and early diagenesis. Our data suggest that diagenesis occurred under rock-buffered conditions, thereby preserving primary signals. The $\delta^{44/40}$Ca record displays a distinctive positive excursion spanning the negative carbon isotope excursion, which agrees well with previously published foraminiferal $\delta^{44/40}$Ca records. $\delta^{44/40}$Ca and $\delta^{88/86}$Sr values negatively correlated with a slope of -0.32, which is close to the value expected for kinetic mass-dependent fractionation but opposite in sign. A positive correlation between $\delta^{88/86}$Sr and δ^{18}O values provides evidence for temperature-dependent Sr isotope fractionation. Both observations directly align with results from coccolithophore culturing studies. In combination, the data support an interpretation where warming and OA stimulated coccolithophore photosynthesis while impairing calcification. All proxy show shifts prior to the PETM onset. Our findings have numerous implications for understanding carbon-climate feedbacks during the PETM and for the first time, illustrate how the $\delta^{44/40}$Ca-$\delta^{88/86}$Sr multi-proxy can trace temperature, as well as calcification and photosynthetic growth rates, in coccolith-dominated facies.