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Triple oxygen isotope systematics of early Earth's carbonate record

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Chemical carbonate sediments throughout the Archaean show systematically lower oxygen isotope compositions by 10 to 15 ‰ in $\delta^{18}O$, compared to the Phanerozoic record [1]. There is still an ongoing debate about the actual mechanism behind the secular shift with three processes being suggested: i) reduced water-carbonate fractionation due to high ocean temperature, ii) an Archaean ocean that was considerably depleted in ^{18}O, and iii) diagenetic overprint and re-equilibration of pristine isotope signatures [2 and references therein].

In order to obtain further constraints on formation temperatures and diagenetic effects, we performed high-precision analyses of the second-order $\delta'^{17}O$ (= $\ln(\delta^{17}O+1) - 0.528$ Â· $\ln(\delta^{18}O+1)$) parameter. CO_2 was liberated from carbonate by orthophosphric acid digestion and measured for its triple oxygen isotope composition by recently developed fragment ion ($^{17}O^+/^{16}O^+$, $^{18}O^+/^{16}O^+$) analysis with a dual-inlet gas source HR-IRMS [3], as well as by a newly modified O_2-CO_2 equilibration technique [4].

Our results show that Archaean carbonates fall below the expected carbonate equilibrium between modern sea water and carbonate in the $\delta'^{17}O$ vs. $\delta^{18}O$ space, suggesting an early ocean that was significantly depleted in ^{18}O. This observation supports the concept of high CO_2 sequestration fluxes and enhanced silicification during the early Archaean [5].

[1] Shields and Veizer (2002), *Geochem. Geophys. Geosyst.*, *3*, 10.1029/2001GC000266 [2] Jaffrés et al. (2007), *Earth-Sci. Rev.*, *83*, 83-122 [3] Adnew et al. (2019), *Rapid Commun Mass Spectrom.*, 33, 1363-1380 [4] Jäger et al. (2021) Goldschmidt 2021 Abstract, 10.7185/gold2021.8081 [5] Herwartz, Pack & Nagel (2021), *PNAS*, *118*, 10.1073/pnas.2023617118