Evaluation of potential explanations for the offsets in Δ_{47} amongst various taxa of naturally occurring modern carbonate organisms

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For more than half a century, the $\delta^{18}O$ paleotemperature proxy has been applied with great success using marine carbonate fossils, yet progress has been limited because carbonate $\delta^{18}O$ is a function of both calcification temperature and the $\delta^{18}O$ of the water in which precipitation occurred, thereby requiring an assumption about past seawater $\delta^{18}O$. The recently developed carbonate "clumped isotope" paleothermometer (Δ_{47}) has the potential to circumvent this difficulty because the "clumping" of ^{13}C and ^{18}O atoms in a carbonate mineral lattice occurs independently of seawater $\delta^{18}O$ [1]. Furthermore, carbonate $\delta^{18}O$ and Δ_{47}-derived temperature are obtained simultaneously – in the same phase – allowing a calculation of seawater $\delta^{18}O$ using a single sample aliquot.

Initially, the relationship between Δ_{47} and calcification temperature was empirically derived using inorganic calcites that were grown at known temperatures [1]. Since that initial calibration study, several naturally occurring modern carbonates have been analyzed, and some yield Δ_{47} values that are consistent with the relationship derived for inorganic calcite (e.g., deep sea corals) [2], whereas others appear to be offset to values that are slightly higher (e.g. surface corals) [1] or slightly lower (e.g. otoliths) [3]. The differences observed between various taxa are not fully understood. Using both published and unpublished modern calibration data sets, several possible explanations for these offsets will be evaluated, including: "vital effects", mineralogical offsets, and differences in the sample preparation protocols of various Δ_{47} laboratories.

[1] Ghosh *et al.* (2006) *Geochim. Cosmochim. Acta* **70**, 1439-1456. [2] Thiagarajan *et al.* (2011) *Geochim. Cosmochim. Acta* **75**, 4416–4425. [3] Ghosh *et al.* (2007) *Geochim. Cosmochim. Acta* **71**, 2736-2744.

Nickel isotopic composition of modern seawater and rivers

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The oceans are the most important repositories for the fallout from continental activities, such as weathering, mediating its flux of elements through a variety of processes including biogeochemical cycling. The abundances and stable isotopes of transition metals have been key tools in understanding and deciphering the totality of these processes through time and, certainly, ongoing development of isotopic systems will continue to reveal new insights into Earth processes. Here we present the first dataset for the Ni isotopic compositions of seawater and rivers.

Dissolved Ni was analysed in several major river systems, including the Amazon and Chang Jiang. In general, rivers exhibit substantial variability in both Ni concentration (2.2-35 nmol kg^{-1}) and $\delta^{60}Ni$ (0.29-1.34‰). Most of the rivers are isotopically heavy with the exception being two rivers that have isotopic compositions that overlap the lithogenic range observed for silicate rocks and sediments [1]. The variability noted in these data may be attributable to isotopic fractionation during chemical weathering or partitioning between dissolved and suspended load, both of which have been put forward to explain similar features in other isotopic systems. However, further studies will be required to resolve the detailed processes.

The range of seawater Ni concentrations was 3.1 nmol kg^{-1} at the surface to 11.2 nmol kg^{-1} at depth, confirming the surface to depth partitioning seen previously, and correlating with nutrient elements such as phosphate and silica. Seawater Ni isotopic data from the Atlantic and Pacific oceans produced three conspicuous outcomes: (i) relative homogeneity of all samples, with an average $\delta^{60}Ni$ of 1.44 ± 0.15 (2σ); (ii) without exception, the Ni isotopic compositions are heavier than that of the dissolved riverine input; (iii) a missing light sink for Ni.

[1] Cameron *et al.* (2009) *PNAS* **106**, 10944-10948.

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