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Secular variation of lithium concentration and isotopic composition of Phanerozoic and Neoproterozoic seawater: Evidence from fluid inclusions in marine halite

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Changes in the global lithium cycle, as recorded in the seawater Li concentration ($[Li^+]_{SW}$) and isotopic composition (δ^7Li_{SW}), have emerged as a promising tracer for reconstructing the long-term controls of changes in seawater chemistry and Earth's geologic carbon cycle. Recent records of δ^7Li_{sw} derived from foraminifera [1], brachiopods [2,3], corals [4], and dolomite [5] show an ~8–9‰ increase over the past 60 million years (Ma) and shallow marine carbonates [6] show a substantial unidirectional increase of ~23‰ over the past 550 Ma. However, laboratory experiments [7, 8] and studies of drill cores from modern carbonate platforms [9] suggest that the δ^7Li values from carbonates are complicated by vital effects, diagenesis, and mineralogy (e.g., calcite vs. aragonite). Thus, other archives are needed to determine whether carbonate δ^7Li values indeed reflect secular changes in δ^7Li_{sw}. Recent experimental work shows the potential use of marine halites as archives of ancient seawater δ^7Li [10]. Here, we present δ^7Li and $[Li^+]$ of fluid inclusions in halite from a large suite of Neoproterozoic and Phanerozoic evaporite basins with marine $^{87}Sr/^{86}Sr$ values. These fluid inclusions were previously used to document the major and trace element composition of paleoseawater, including lithium concentrations [11,12,13]. $[Li^+]_{SW}$ varied twelve-fold and oscillated twice between high- and low-Li concentrations since 550 Ma, in rhythm with $[Ca^{2+}]_{SW,}$ $[Sr^{2+}]_{SW}$, modeled degassing rate and atmospheric pCO_2, aragonite-calcite-seas, KCl–$MgSO_4$ evaporites, and greenhouse-icehouse climates [11,12,13,14,15]. δ^7Li_{SW} varied ~17–19‰ over the past 550 Ma, and antiparallels the $[Li^+]_{SW}$, $[Ca^{2+}]_{SW}$, $[Sr^{2+}]_{SW}$, and parallels the Mg/Ca_{sw}. Secular variations in seawater chemistry point to the importance of plate tectonic activity and seafloor hydrothermal systems in regulating the composition of Earth's hydrosphere and atmosphere.

[1] Misra and Froelich, 2012; [2] Washington et al., 2020; [3] Gaspers et al., 2021; [4] Murphy et al., 2019; [5] Liu et al., 2023; [6] Kalderon-Asael et al., 2021; [7] Vigier et al., 2015; [8] Dellinger et al., 2018; [9] Murphy et al., 2022; [10] Lin et al., 2024; [11] Weldeghebriel et al., 2022; [12] Weldeghebriel et al., 2023; [13] Weldeghebriel and Lowenstein, 2023, [14] Mills et al., 2019; [15] Berner and Kothavala, 2001.