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# Decoding Earth's Lithospheric Redox Evolution through the Diversity and Distribution of Mineral-Forming Ions

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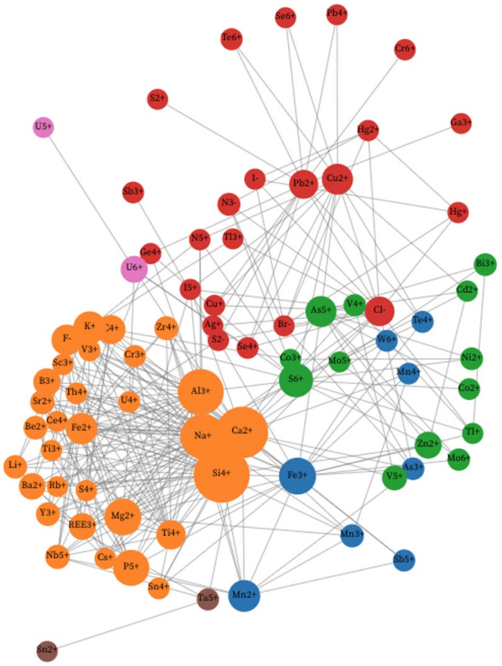

Crustal minerals record the evolution of near-surface environments through billions of years of Earth history. In particular, the deep-time distribution of elements and their redox-sensitive ions among minerals from different formational environments underscores a key theme of Victor Goldschmidt's [*1,2*] work on the geochemical classification of the elements. We analyzed distributions of 72 mineral-forming elements and 87 mineral-forming ions in 4834 oxygen- and/or halogen-bearing mineral species. We identified coexisting pairs of ions in those minerals and investigated unipartite networks with community detection and heatmap analysis with agglomerative clustering to discern patterns of element and ion associations and antipathies for different modes of mineral formation through Earth history. These analyses reveal striking deep-time changes in element and ion distributions, notably as a consequence of the co-evolving biosphere.

Most ions in oxygen-bearing minerals fall into two distinct communities/clusters. The larger group includes common ions in primary igneous and metamorphic minerals (e.g., Si$^{4+}$, Al$^{3+}$, Ca$^{2+}$, Mg$^{2+}$, K, and Na), ions characteristic of complex pegmatites (B$^{3+}$, Be$^{2+}$, Cs, Li), and highly differentiated alkaline igneous rocks (REE$^{3+}$, Ti$^{4+}$, Zr$^{4+}$, Ta$^{5+}$), with low average electronegativity. By contrast, a second community is associated with ions typical of near-surface environments, including brines (Ag$^{+}$, Br$^{-}$, Cu$^{+}$, Hg$^{+}$, I$^{-}$) or high field strength ions from weathered primary oxide or sulfide deposits (Cr$^{6+}$, Pb$^{4+}$, Mo$^{4+}$, Te$^{4+}$, Te$^{6+}$).

As Goldschmidt highlighted, many redox-sensitive elements occur in more than one category. For example, in crustal oxides and silicates, Fe$^{2+}$ usually occurs in lithophilic environments, whereas Fe$^{3+}$ is more strongly associated with weathering and aqueous alteration environments. At a planetary scale, an element's more oxidized state typically is lithophile. We find that a greater fraction of As$^{5+}$, Cr$^{6+}$, and Fe$^{3+}$ minerals bond to O$^{2-}$ compared to As$^{3+}$, Cr$^{3+}$, or Fe$^{2+}$ minerals. But in crustal environments, ions in their lower oxidation states typically group with unambiguously lithophile elements, whereas the more oxidized As$^{5+}$, Cr$^{6+}$, and Fe$^{3+}$ ions are most strongly linked to chalcophile elements. These trends underscore the importance of biological influences on the geochemical evolution of Earth's lithosphere.

[*1*] V.M. Goldschmidt (1937) *J. Chem. Soc*., 655-673

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