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The role of organosulfur in coupled Fe-S cycles in the deep terrestrial subsurface

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Despite being one of Earth's largest ecosystems, element cycling, especially coupled cycles, in the continental deep subsurface remains poorly constrained. Recent geochemical and metagenomic investigations of a 2.7 Ga banded iron formation within the Canadian Shield (Soudan Mine, MN) identified evidence of cryptic S cycling coupled to Fe cycling, with dissimilatory sulfur and iron reduction thought to facilitate the precipitation of iron sulfide minerals. Here, we expand on this framework by considering the potential for Fe and C interactions with the microbial cycling of organosulfur (OrgS), which have received little attention in the deep subsurface. In this study, we employ shotgun metagenomic sequencing of the continental deep biosphere to investigate the genetic potential for OrgS cycling and its impact on coupled elemental cycling. Our results demonstrate that a taxonomically diverse microbial community likely supports an active OrgS cycle, with dimethylsulfoniopropionate (DMSP) and taurine cycling genes identified in 95.4% and 64.6% of metagenome assembled genomes, respectively. Genetic evidence suggests an integral linkage between the organic and inorganic sulfur cycle, with the biosynthesis and subsequent cycling of OrgS compounds directly impacting the availability of redox reactive inorganic S species that support metabolism in this energy-limited setting. Thus, OrgS plays a vital role in fueling microbial life and acts as an important control on the S fluxes shown to mediate Fe cycling in this continental deep subsurface setting. The ubiquity of genes involved in organosulfur cycling and its tight coupling to inorganic sulfur and iron cycles necessitates consideration of OrgS when constraining the nature and extent of deep subsurface biogeochemical Fe-S cycling and their impact on biogeochemical cycling across Earth's Critical Zone.