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A modern sulfurous thermal spring with sulfur mass-independent fractionation

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The occurrence of sulfur mass-independent fractionations, typified by non-zero $\Delta^{33}S$ and $\Delta^{36}S$-values, is primarily restricted to Archean sediments (i.e. from -2 up to +15‰), the main mechanism being related to photochemical reactions involving SO_2 in an anoxic atmosphere [1]. Since the Proterozoic, mass-independent signatures remain rare but are common among tropospheric and stratospheric aerosols (i.e.from -1 to 2‰) and atmospheric-related deposits; these are, again, primarily related to photochemical reactions [2]. Other mass-independent sulfur signatures occurs through so-called mass-conservation effects and are quite well understood and relate to moderate magnitude anomalies (typically $\Delta^{33}S$ <0.2‰), as observed in sediments and microbiological culture experiments [3].

Here we report on the occurrence of a sulfurous thermal spring at Secours in the French Pyrenees that shows, over >3 years of sampling, significant and ubiquitous sulfur mass-independent fractionations. While dissolved sulfide is characterized by variable but always positive $\Delta^{33}S$-values, from ~ 0.2 to 0.9‰, dissolved sulfate is characterized by near constant $\Delta^{33}S$-values of -0.15‰ showing no relations with $\delta^{34}S$-covariations ($\Delta^{34}S_{sulfide\text{-}sulfate}$ from -27 to -11‰). There is no Archean rocks in the close vicinity pointing to a fractionation mechanism is at play. Both dissolved sulfide and sulfate show near zero $\Delta^{36}S$-values suggesting that a magnetic effect is at play.

Sulfide/sulfate ratios show significant variability over a factor 3 which suggests that some sulfur cycling operates through several oxidation/reduction steps. Anaerobic microbiological cultures conducted from water samples and supplemented with sulfate reveal too small isotope anomalies to account for the present isotope record ($\Delta^{33}S$ up to 0.2‰), although the anomalies are larger than those reported so far for sulfate-reducing bacteria. The study of taxonomic reveals that the microbial community is predominantly driven by the sulfur cycle, with a dominance of sulfur-oxidizing families including *Thiotrichaceae*, *Sulfurovaceae*, *Halothiobacillaceae* and *Acidithiobacillaceae*, along with few sulfate-reducers belonging to the *Desulfocapsaceae* family. We are presently determining the O-isotope compositions to investigate the scale of sulfide re-oxidation in the sulfate budget of the Secours waters.

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[3] Farquhar, Johnston, Wing (2007) GCA 71, 5862–5875.