

Environmental S isotope fractionations: An update

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The $\delta^{34}\text{S}$ values can be used to follow transport pathways of sulfur in ecosystems. However, if we want to ascribe a specific pollution source to sulfur found in damaged ecosystems, two difficulties may arise: (i) isotopic separation between potential S sources is too small ($<2\text{‰}$ CDT), and (ii) an isotope fractionation occurred. Isotope-selective processes can "smear out" the $\delta^{34}\text{S}$ signature of a sulfur source.

Until the early 1990's, bacterial sulfate reduction was viewed as the only process in the biosphere causing a large S isotope fractionation. Our data collected along two European transects (west-east, north-south) indicate that three more processes are associated with a systematic S isotope effect:

- atmospheric SO_2 -to- SO_4^{2-} conversion (isotopically heavier S in the product);
- assimilation of atmospheric S by plants (organic S lighter), and
- mineralization of organic S in soil (lighter S removed).

Air-borne S species, spruce canopy throughfall, forest floor moss, *Sphagnum* peat profiles, forest soil, lysimeter water and surface discharge were analysed for $\delta^{34}\text{S}$ over a period of 5 years ($n=800$). Study sites were located in Ireland, Scotland, England, Czech Republic, Sweden, France and Italy.

The "inverse" $\delta^{34}\text{S}$ fractionation (i.e., isotopically heavier product) found for oxidation of SO_2 in the air is explained by a large equilibrium effect of dissolution, with HSO_3^- enriched in ^{34}S . The $\delta^{34}\text{S}$ of sulfate aerosol was higher by 4.8‰ compared to SO_2 . Assimilation favours the lighter isotope ^{32}S on average by 2‰ . In contrast, mineralization of organic soil S leaves the residual S fraction on average by 1.7‰ heavier than fresh organic matter. Sulfate S in soil water taken by suction lysimeters was by 1.5‰ lighter than in atmospheric deposition, also documenting isotope selectiveness of S mineralization.