



Microbial oxidation of tetrathionate and elemental sulfur : Insights from S and O isotopes of sulfate

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In this study, we experimentally evaluate pH and sulfur chemistry associated with abiotic and microbial oxidation of tetrathionate and elemental sulfur under acidic aerobic conditions ($\text{pH} < 4$) and pure strain of *Acidithiobacillus thiooxidans* using S and O isotopes of sulfate. Experimental results of batch cultures showed active microbial sulfur processing of tetrathionate and elemental sulfur. Polythionates, principally thiosulfate, sulfate and elemental sulfur accumulated during oxidation of tetrathionate with decreasing pH (2.1) indicating disproportionation reactions. In contrast thiosulfate and sulfite was only determined at the early stage of elemental sulfur oxidation and disappeared with decreasing pH and sulfate became dominant sulfur species. Tetrathionate was not determined in biotic or abiotic elemental sulphur experiments. pH decreased in all incubation experiments from starting pH values of 4 to 2 or less for all two sulfur substrates. Abiotic control experiments run with either tetrathionate- or elemental sulphur as substrate did not produce significant amount of sulfate and thiosulfate and pH of solution remain static over the course of the experiments. In contrast to elemental sulfur in which the $[\text{U}+\text{F}064]$ $^{34}\text{S}/\text{SO}_4$ values showed insignificant sulfur isotope fractionation ($\epsilon_{\text{SO}_4-\text{S}_0} = -0.9\text{‰}$), *Acidithiobacillus thiooxidans* selectively fractionated ^{34}S during the oxidation of tetrathionate and produced sulfate enriched in $^{34}\text{S} \sim 6.4\text{‰}$ with an average value of $6.9 \pm 0.2\text{‰}$ after a month incubation. However, the range of fractionation and time course kinetics of enrichment varied. The contribution of water-derived O to sulfate by S_0 oxidation ranged from 58 % to 103 % and at the initial stage oxidation of elemental sulfur significant O_2 contribution into sulfate was estimated. At the end of a month incubation average $87 \pm 7.0\%$ of sulfate oxygen was derived from water. The contribution of water-derived O ranged from 53 % to 64 % during microbial oxidation of tetrathionate. The differences in both S and O isotopes values of sulfate obtained from elemental sulfur and tetrathionate experiments may indicate different sulfur processing pathways amongst phylogenetically same organisms.