

Supplementary Materials for

Large sulfur isotope fractionation by bacterial sulfide oxidation

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Table S1. Compilation of sulfur isotope enrichments during MSO used for the construction of Fig. 1 and additional information on growth conditions.

References (43–53)

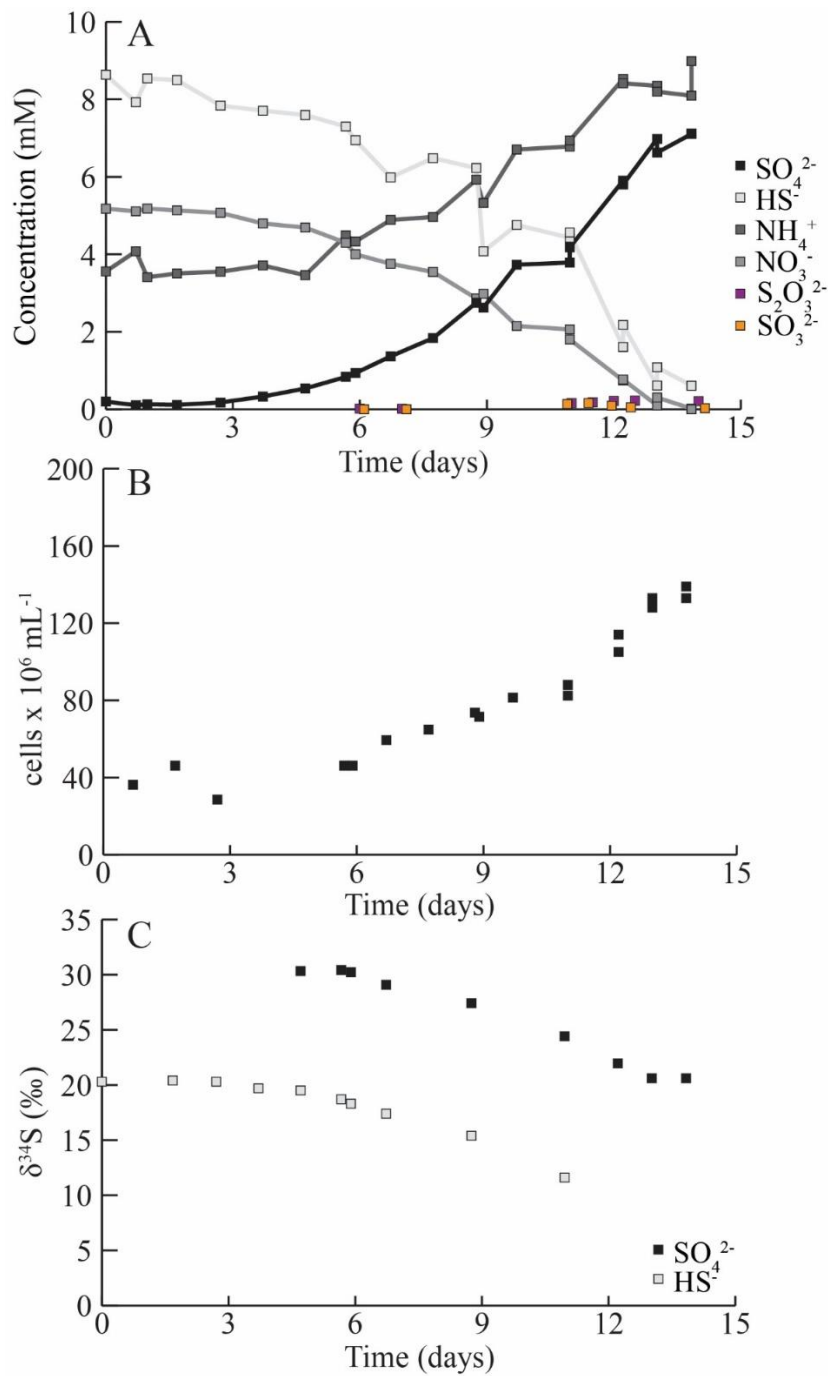


Fig. S1. Growth experiment 1. Panel A concentrations (in mM) of sulfide, sulfate ammonium, nitrate, thiosulfate and sulfite. Panel B cell density as a function of time, Panel C $\delta^{34}\text{S}$ of the sulfate and sulfide pools as a function of time. Sulfate $\delta^{34}\text{S}$ could not be measured prior to T= 5 days due to a lack of enough sulfate produced.

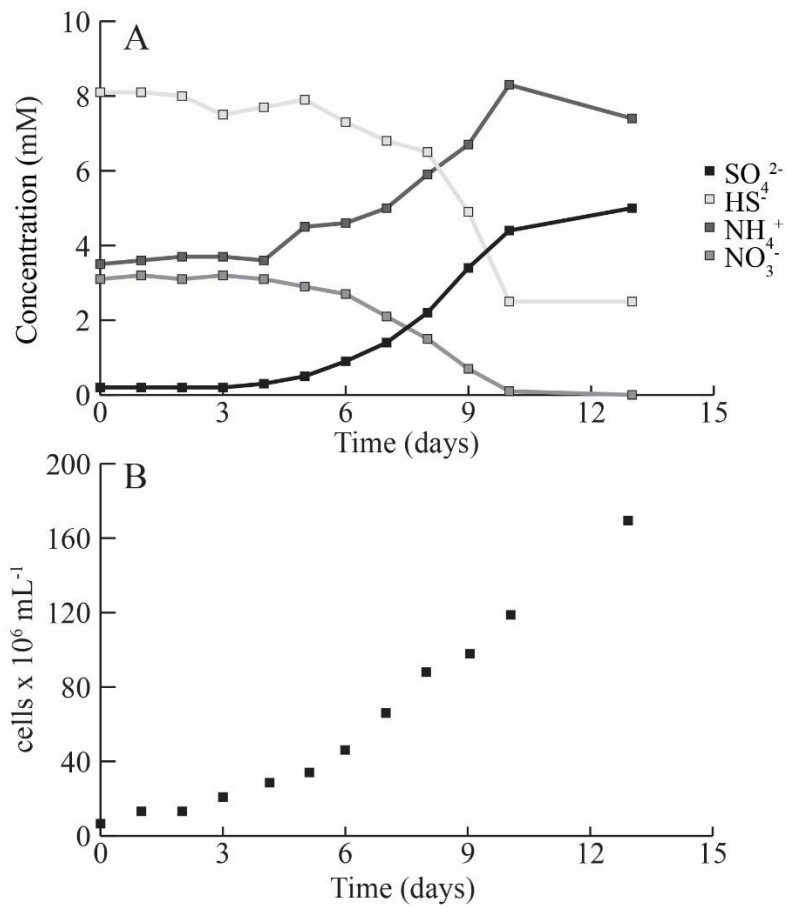


Fig. S2. Growth experiment 2. Panel A concentrations (in mM) of sulfide, sulfate ammonium, nitrate through the growth experiment, Panel B cell density as a function of time.

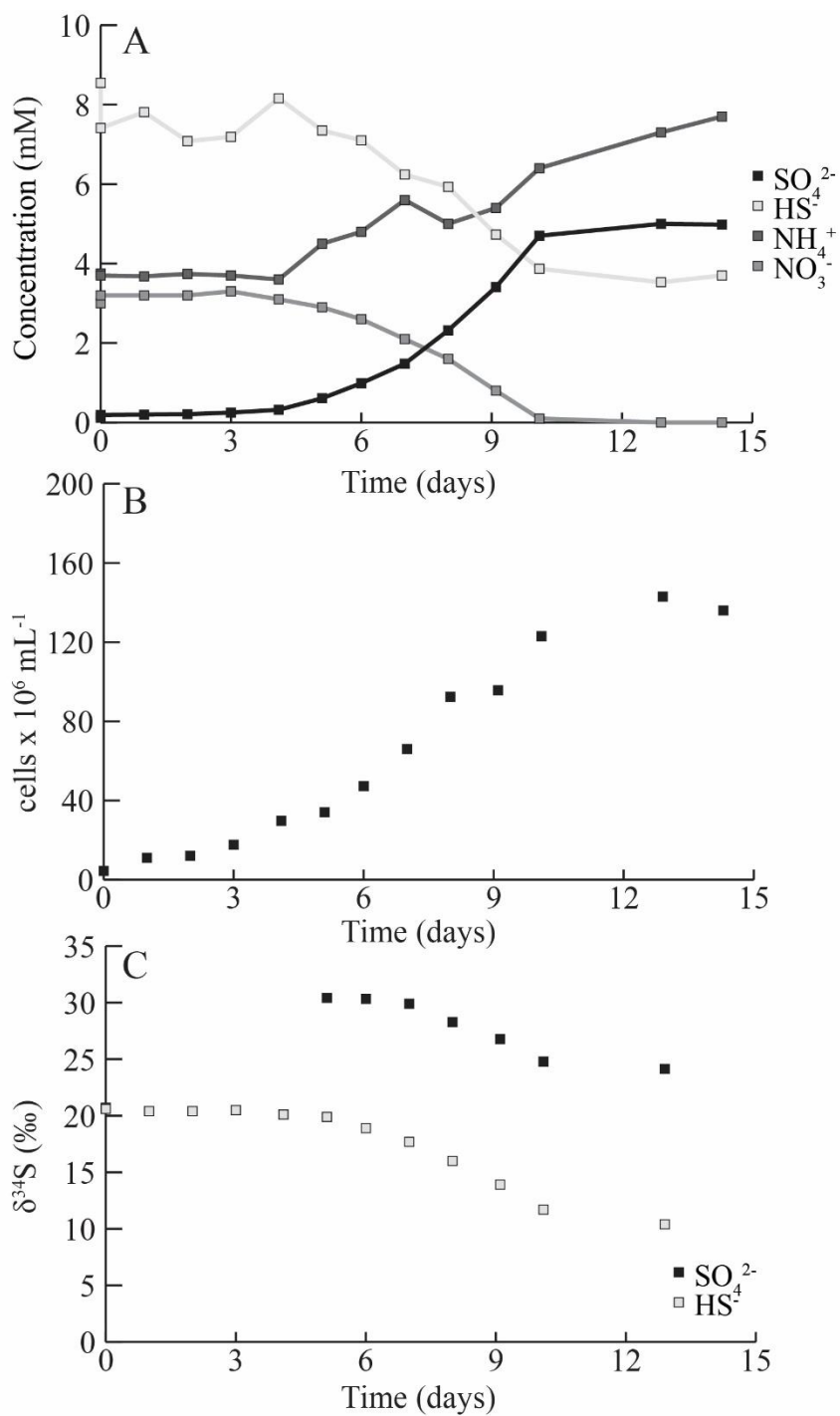
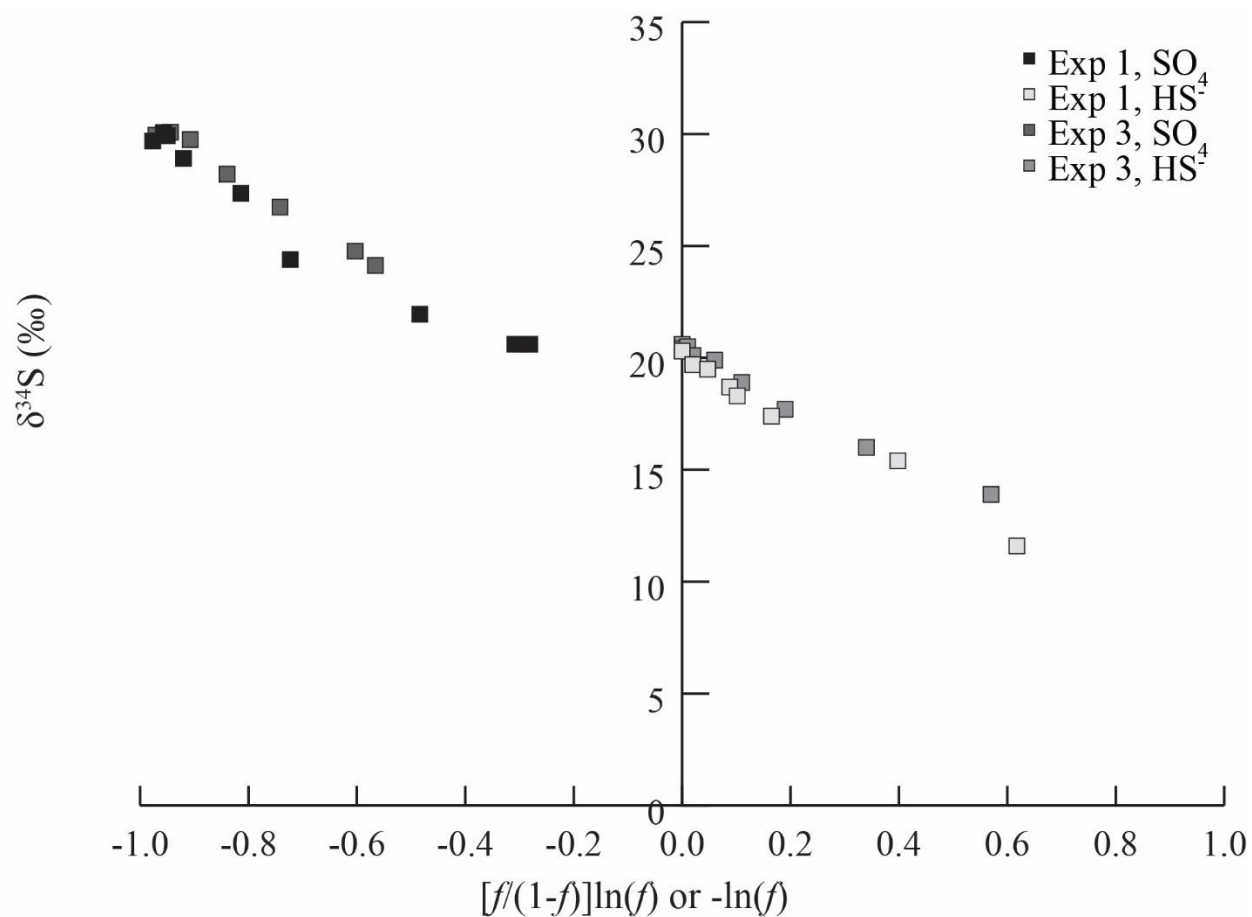


Fig. S3. Growth experiment 3. Panel A concentrations (in mM) of sulfide, sulfate ammonium, nitrate. Panel B cell density as a function of time, Panel C $\delta^{34}\text{S}$ of the sulfate and sulfide pools as a function of time. Sulfate $\delta^{34}\text{S}$ could not be measure prior to T= 5 days due to a lack of enough sulfate produced.



Experiment	HS^-		SO_4	
	$^{34}\epsilon$	σ	$^{34}\epsilon$	σ
1	13.1	0.7	14.1	1.1
2				
3	12.0	0.5	15.2	0.7

Fig. S4. Plot of $\delta^{34}\text{S}$ as a function of $\ln(f)$ for the sulfide or $[f/(1-f)]\ln(f)$ for the sulfate for experiments 1 and 3 (see Materials and Methods). The table shows the slope of the regression analysis and the uncertainty on the regression is reported as σ . Note that the estimates of $^{34}\epsilon$ are different between the sulfide and the sulfate. The lower estimates are reported in Table 1.

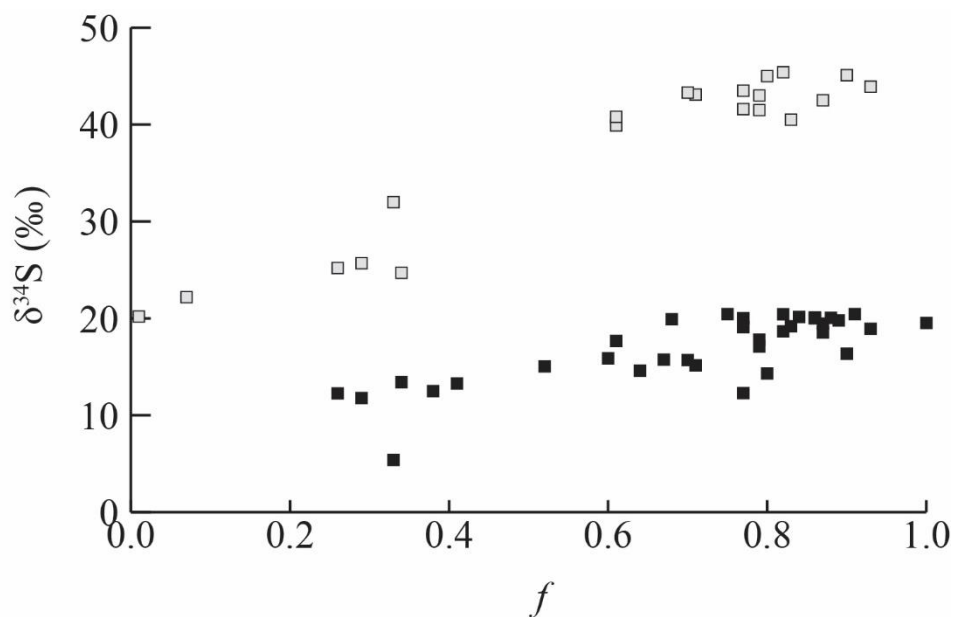


Fig. S5. Plot of $\delta^{34}\text{S}$ as a function of the sulfide consumed (f). Measurements for sulfide (black squares) and sulfate (grey squares) from when cells taken from the stationary phase are used as inoculum. The large difference between the $\delta^{34}\text{S}$ of sulfate and sulfide (up to +26 ‰) suggests that under different physiological conditions, sulfide oxidation by DA can produce larger sulfur isotope enrichments.

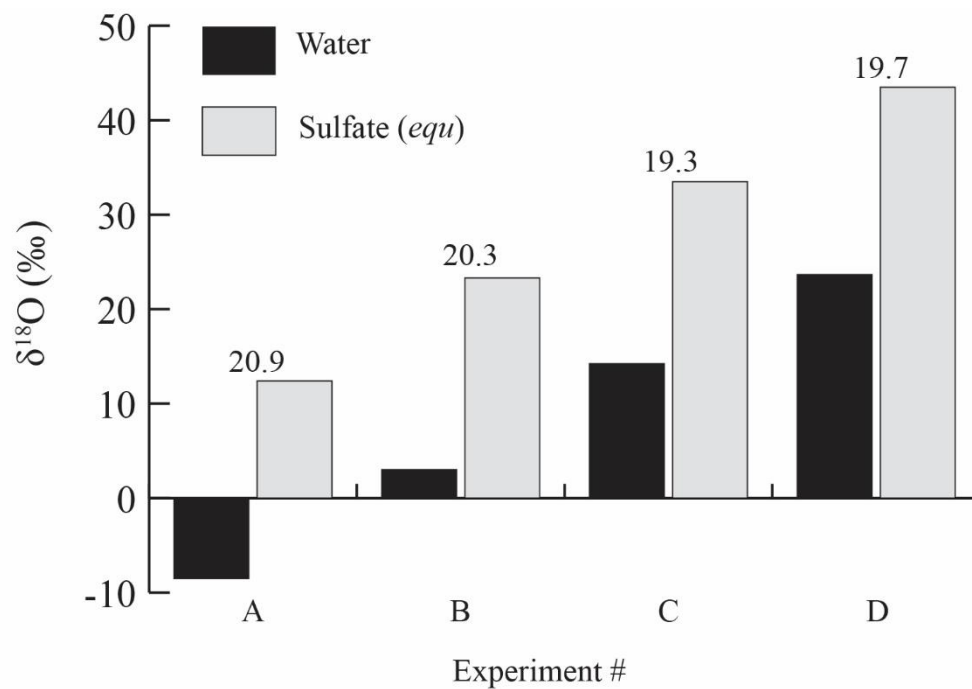


Fig. S6. Water (black bars) and equilibrium sulfate (grey bars) $\delta^{18}\text{O}$ in respective water enrichment experiments A to D. Numbers represent the difference between the sulfate and water with an average value of 20.1‰ .

Table S1. Compilation of sulfur isotope enrichments during MSO used for the construction of Fig. 1 and additional information on growth conditions. Top section compiles sulfur isotope enrichments between aqueous sulfide and sulfate during the microbial oxidation of sulfide while the middle section compiles the fractionation measured between the two major steps in sulfide oxidation specifically during phototrophic sulfide oxidation. The bottom section compiles fractionation measured with a mineral starting material. A large enrichment in ^{32}S is reported during oxidation of sulfide by *Chromatium sp.* (43). However, these result could not be replicated in subsequent studies (14). It has therefore not been included in this compilation. Number references in table are (24)¹, (44)², (45)³, This study⁴, (46)⁵, (47)⁶, (48)⁷, (49)⁸, (14)⁹, (4)¹⁰, (50)¹¹, (51)¹², (22)¹³, (52)¹⁴, (53)¹⁵

Substrate/product	Organism	Pathway/conditions	pH	T °C	ε _{P-R} (‰)		Ref.
					min	max	
H ₂ S/SO ₄ ²⁻	<i>Allochromatium vinosum</i>	Continuous sulfide oxidation	7	25		0.1	1
H ₂ S/SO ₄ ²⁻	<i>Chlorobium tepidum</i>	Two-step oxidation	7	45	-0.7		1
H ₂ S/SO ₄ ²⁻	<i>Chlorobium tepidum</i>	Same as above but early exponential phase	7	45	-2.3		1
H ₂ S/SO ₄ ²⁻	<i>Thiobacillus denitrificans</i>	Sulfide oxidation with NO ₃ ⁻	7	30	-2.9	-1.6	2
H ₂ S/SO ₄ ²⁻	<i>Sulfurimonas denitrificans</i>	Sulfide oxidation with NO ₃ ⁻	7	21	-4.3	-1.3	2
H ₂ S/SO ₄ ²⁻	<i>Acidithiobacillus thiooxidans</i>	Oxidation with O ₂				-0.9	3
H ₂ S/SO ₄ ²⁻	<i>Desulfurivibrio alkaliphilus</i>	DNRA-mediated sulfide oxidation	9.8	30		12.5	4
H ₂ S/S ⁰	<i>Chloropseudomonas ethylicum</i>	Green sulfur bacteria				1.5	5
H ₂ S/S ⁰	<i>Rhodopseudomonas sp.</i>	Purple sulfur bacteria				1.2	6
H ₂ S/S ⁰	<i>Ectothiorhodospira shaposhnikovii</i>	Purple sulfur bacteria	8.2	28		2.2	7
H ₂ S/S ⁰	<i>Chlorobium thiosulfatophilum</i>	Green sulfur bacteria				< 3	8
H ₂ S/S ⁰	<i>Chromatium vinosum</i>	Purple sulfur bacteria	8.1	35		2.4	9
H ₂ S/S ⁰	<i>Chlorobium tepidum</i>	Purple sulfur bacteria	7	48		1.8	10
S ⁰ /SO ₄ ²⁻	<i>Chlorobium tepidum</i>	Purple sulfur bacteria	7	48	-3.3	0	10
S ⁰ /SO ₄ ²⁻	<i>Chromatium vinosum</i>	Purple sulfur bacteria	8.1	35	0	0	9
S ⁰ /SO ₄ ²⁻	<i>Ectothiorhodospira shaposhnikovii.</i>	Purple sulfur bacteria			0	0	7
Chalcopyrite/SO ₄ ²⁻	<i>Acidithiobacillus ferrooxidans</i>	Oxidation by O ₂	3.4			-1.5	11
Chalcopyrite/SO ₄ ²⁻	<i>Acidithiobacillus ferrooxidans</i>	Oxidation by Fe(III)	2.2			-3.8	11
Sphalerite/SO ₄ ²⁻	<i>Acidithiobacillus ferrooxidans</i>	Oxidation by O ₂	2.9	25	0	0	12
Sphalerite/SO ₄ ²⁻	<i>Acidithiobacillus ferrooxidans</i>	Oxidation by Fe(III)	1.9	25	-2.6		12
Pyrite/SO ₄ ²⁻	<i>Acidithiobacillus ferrooxidans</i>	Oxidation by O ₂	2.3	25	-0.1		13
Pyrite/SO ₄ ²⁻	<i>Acidithiobacillus ferrooxidans</i>	Oxidation by O ₂	2.0	20	-1.7	3.5	14
Pyrite/SO ₄ ²⁻	<i>Acidithiobacillus ferrooxidans</i>	Oxidation by O ₂	1.6	30	-1.3	0.4	15