

# Supplementary Material: A novel approach to sulfate geoengineering with surface emissions of carbonyl sulfide

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## 1 Tables

<b>Experiment</b>	<b>Surface upward flux</b>	<b>Chemical production</b>	<b>Surface dry deposition</b>	<b>Chemical loss</b>	<b>Net [sources-sinks]</b>
<b>BG</b>	0.12±0.01	0.40±0.01	0.39±0.01	0.13±0.01	0.00±0.01
<b>SG-COS</b>	40.1±0.1	0.39±0.01	31.6±0.1	8.8±0.1	+0.1±0.1

S 1: Globally-annually averaged COS sources and sinks (Tg-S/yr) [years 2046-2055].

<b>COS sources</b>	<b>Gg-S/yr</b>	<b>%</b>
<b>Surface flux from oceans</b>	39.8	8.3
<b>Surface anthropogenic flux</b>	76.3	15.8
<b>Chem prod from CS<sub>2</sub> ocean sources</b>	76.8	15.9
<b>Chem prod from CS<sub>2</sub> anthropogenic sources</b>	117.6	24.4
<b>Chem prod from DMS ocean sources</b>	171.5	35.6

S 2: Globally-annually averaged COS sources for BG (Gg-S/yr and percent of the total direct surface upward flux and total atmospheric chemical production) [years 2046-2055].

<b>COS sink</b>	<b>Gg-S/yr</b>	<b>%</b>
<b>Dry deposition on soils</b>	125.6	24.1
<b>Surface dry dep on vegetation</b>	252.6	48.5
<b>Chemical loss by OH</b>	102.7	19.7
<b>Chemical loss by photolysis</b>	30.1	5.8
<b>Chemical loss by O</b>	9.7	1.9

S 3: Globally-annually averaged COS sinks for BG (Gg-S/yr and percent of the total surface dry deposition or total atmospheric chemical loss) [years 2046-2055].

<b>Experiment</b>	<b>DMS</b>	<b>SO<sub>2</sub></b>	<b>SO<sub>4</sub></b>	<b>COS</b>	<b>CS<sub>2</sub></b>	<b>H<sub>2</sub>S</b>	<b>Total</b>
<b>BG</b>	25.9	63.7	1.41	0.12	0.88	4.0	96.0
<b>SG-SO2</b>	25.9	71.7	1.41	0.12	0.88	4.0	104.0
<b>SG-COS</b>	25.9	63.7	1.41	40.1	0.88	4.0	136.0

S 4: Globally-annually averaged sources of sulfur species (Tg-S/yr) [years 2046-2055].

<b>SG-COS - BG: Sulfate aerosols RF (W/m<sup>2</sup>)</b>	<b>SW</b>	<b>LW</b>	<b>NET</b>
<b>Clear sky</b>	-2.49	+0.48	-2.01
<b>Cloud adjustment [background clouds]</b>	+0.80	-0.06	+0.74
<b>Cloud adjustment [cirrus ice thinning]</b>	+0.27	-0.52	-0.25
<b>Cloud adjusted</b>	-1.42±0.12	-0.10±0.25	-1.52±0.13

S 5: Temperature-adjusted tropopause RF of sulfate aerosols in the SG-COS case with respect to BG (shortwave, longwave and net) (W/m<sup>2</sup>) [years 2046-2055]. First row shows RFs under clear sky conditions. Second and third rows present the cloud adjustment of RFs, separately for the mere presence of background clouds and for the cirrus ice thinning produced in SG conditions (see Kuebbeler et al., 2012; Visioni et al., 2018).

<b>SG-SO2 - BG: Sulfate aerosols RF (W/m<sup>2</sup>)</b>	<b>SW</b>	<b>LW</b>	<b>NET</b>
<b>Clear sky</b>	-2.75	+0.52	-2.23
<b>Cloud adjustment [background clouds]</b>	+0.81	-0.06	+0.75
<b>Cloud adjustment [cirrus ice thinning]</b>	+0.38	-0.73	-0.35
<b>Cloud adjusted</b>	-1.56±0.10	-0.27±0.23	-1.83±0.13

S 6: As in Table 5, but for the SG-SO2 case.

<b>SG-COS - BG: Greenhouse gases RF (W/m<sup>2</sup>)</b>	<b>SW</b>	<b>LW</b>	<b>NET</b>
<b>COS</b>	0.00	+0.17±0.02	+0.17±0.02
<b>CH<sub>4</sub></b>	0.00	+0.12±0.01	+0.12±0.01
<b>H<sub>2</sub>O [stratosphere]</b>	0.00	-0.041±0.004	-0.041±0.004
<b>O<sub>3</sub> [stratosphere]</b>	-0.051±0.005	+0.010±0.001	-0.041±0.005
<b>O<sub>3</sub> [troposphere]</b>	0.00±0.01	+0.02±0.01	+0.02±0.01
<b>Total</b>	-0.05±0.01	+0.28±0.03	+0.23±0.03

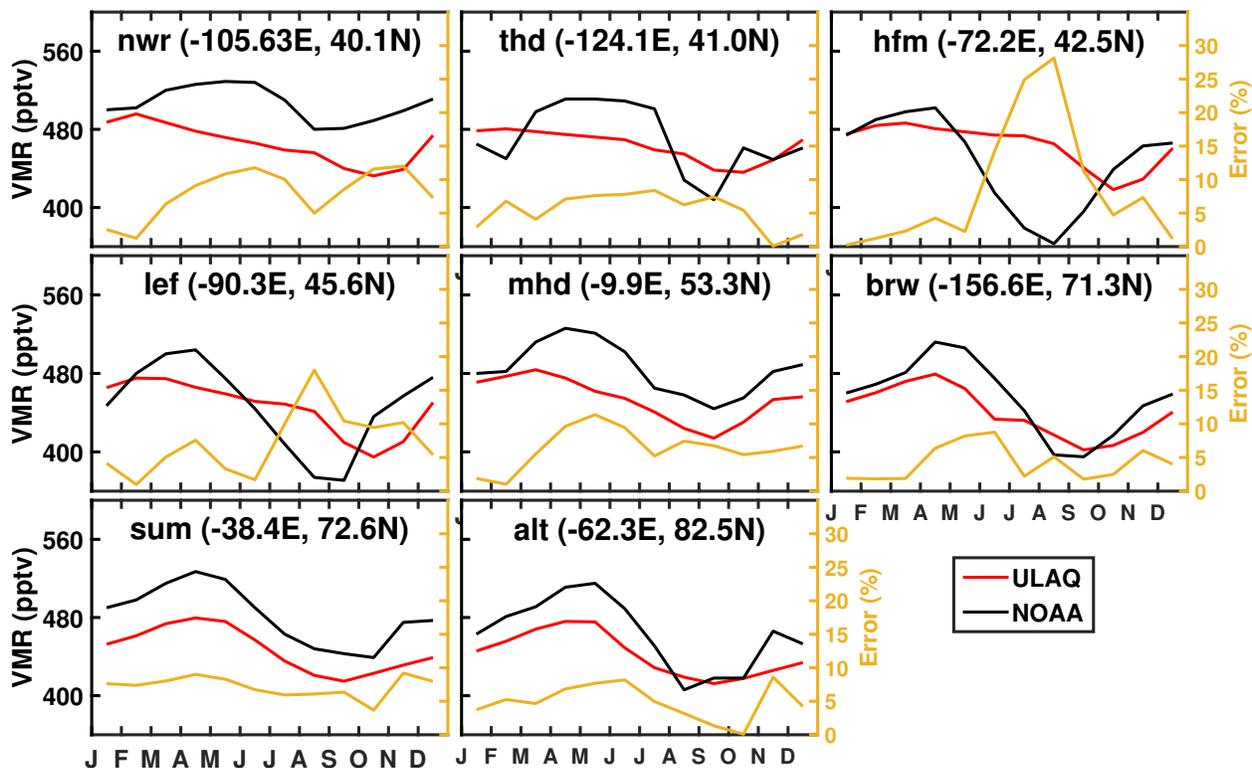
S 7: Temperature-adjusted tropopause RF of greenhouse gases in the SG-COS case with respect to BG (shortwave, longwave and net) (W/m<sup>2</sup>) [years 2046-2055]. First five rows present the RF contributions of specific greenhouse gases affected directly and indirectly by SG (*i.e.*, COS, CH<sub>4</sub>, stratospheric H<sub>2</sub>O, stratospheric and tropospheric O<sub>3</sub>). Last row shows the gas net total RF.

<b>SG-SO2 - BG: Greenhouse gases RF (W/m<sup>2</sup>)</b>	<b>SW</b>	<b>LW</b>	<b>NET</b>
<b>COS</b>	0.00	0.00	0.00
<b>CH<sub>4</sub></b>	0.00	+0.11±0.02	+0.11±0.02
<b>H<sub>2</sub>O [stratosphere]</b>	0.00	+0.005±0.001	+0.005±0.001
<b>O<sub>3</sub> [stratosphere]</b>	-0.01±0.01	0.00±0.01	-0.01±0.01
<b>O<sub>3</sub> [troposphere]</b>	0.00±0.01	+0.02±0.01	+0.02±0.01
<b>Total</b>	-0.01±0.01	+0.14±0.02	+0.13±0.02

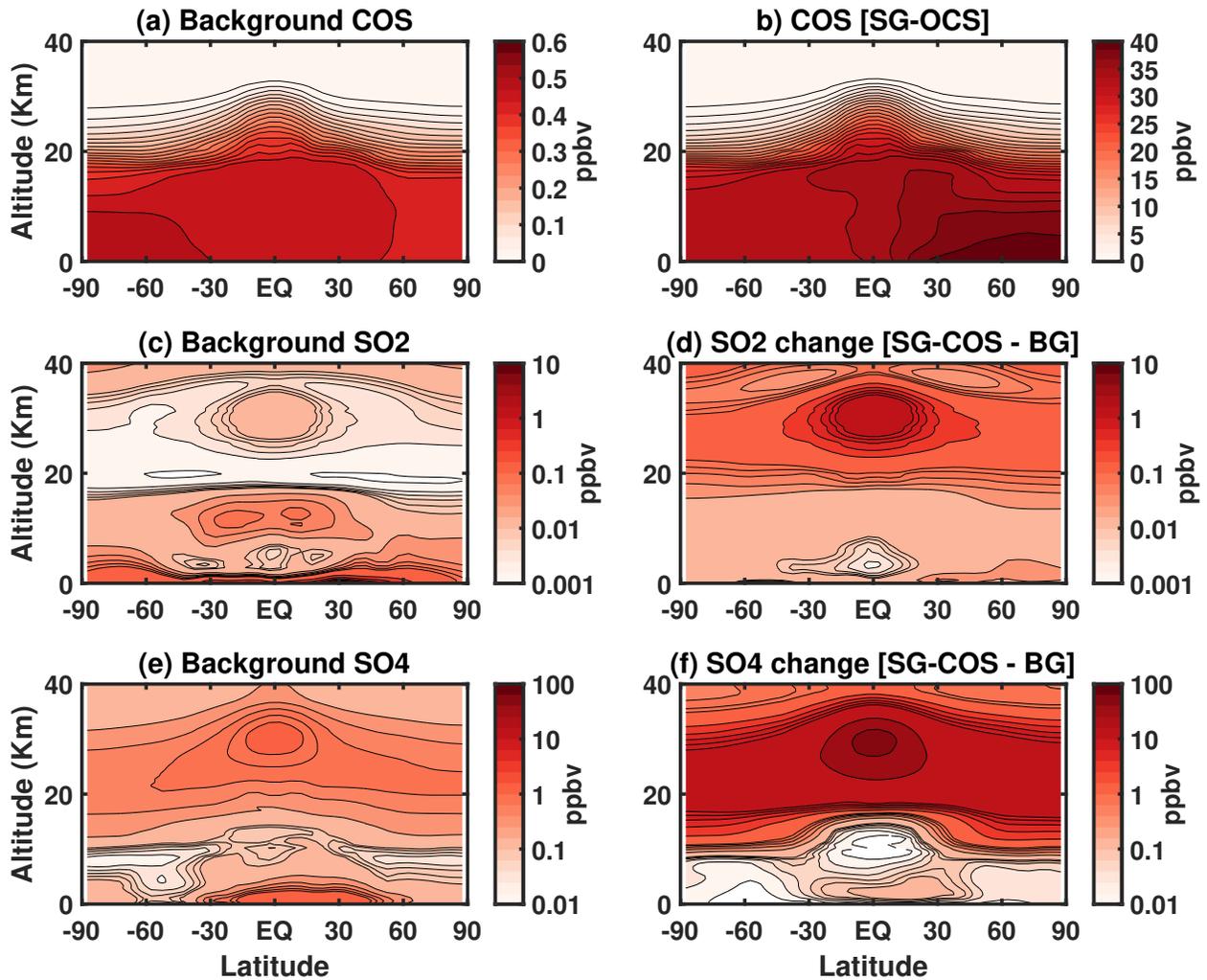
S 8: As in Table 7, but for the SG-SO2 case.

## 2 Figures

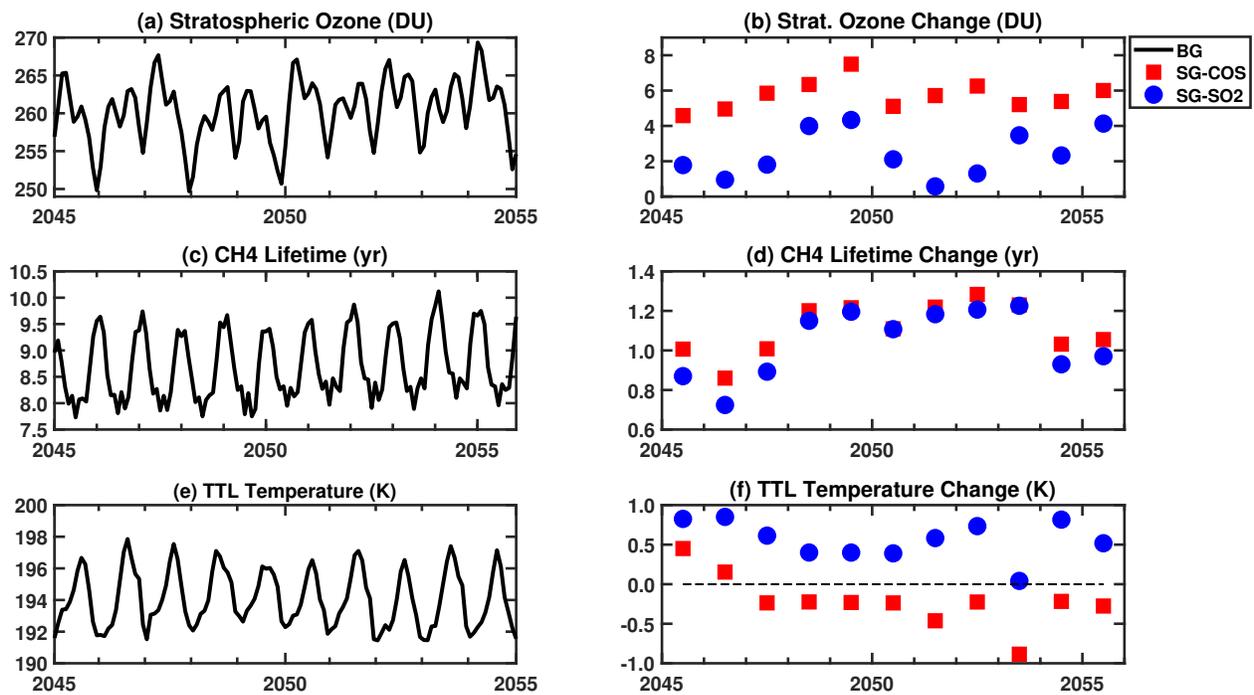
### COS Surface Mixing Ratio



S 1: COS surface mixing ratio (in pptv, left axis) evaluation at NOAA stations: NWR: Niwot Ridge, United States; THD: Trinidad Head, United States; HFM: Harvard Forest, United States; LEF: Wisconsin, United States; MHD: Mace Head, Ireland; BRW: Barrow, United States; SUM: Summit, Greenland; ALT: Alert, Canada. Red line is the COS surface mixing ratio averaged over 2046-2055 as calculated by ULAQ-CCM, black line is the NOAA observations in 2006. On the right axis, we report the absolute difference (in %) between the two lines.

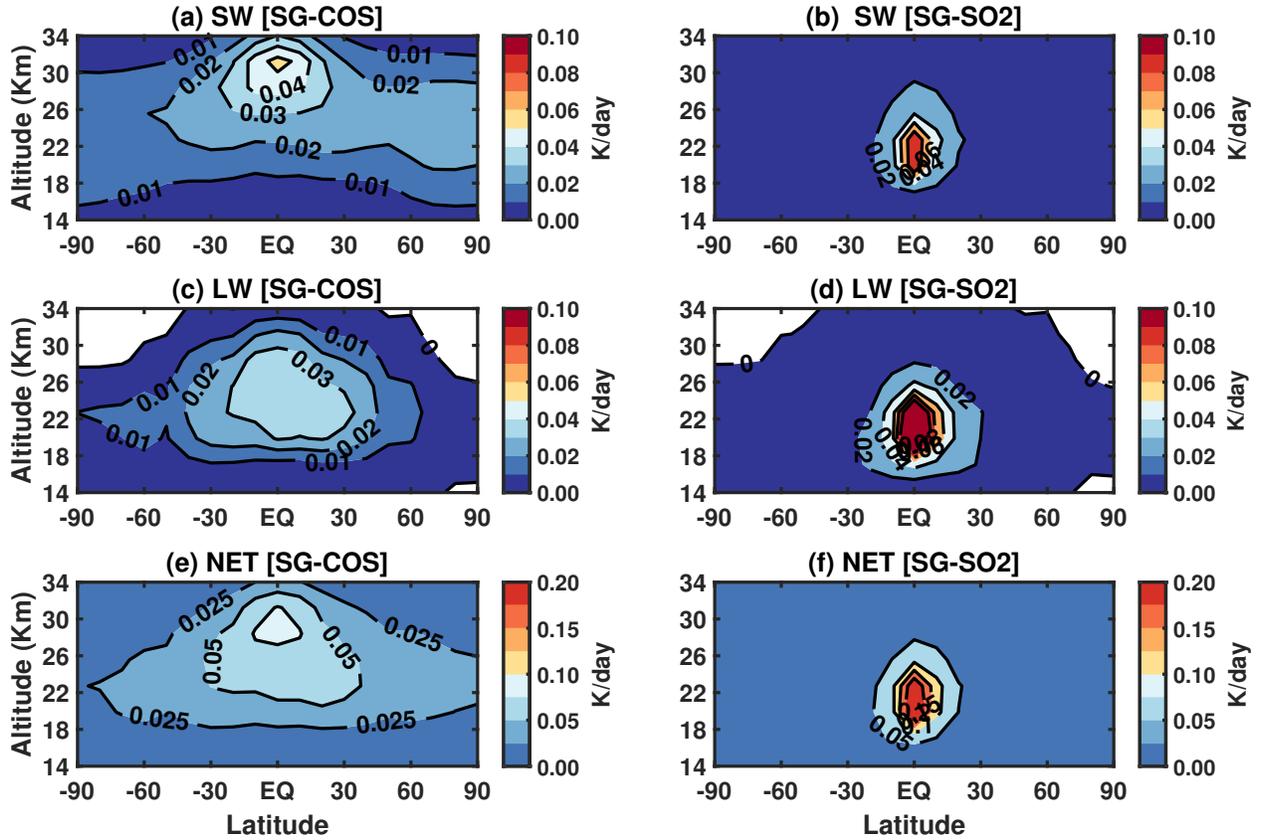


S 2: Mean zonal values of volume mixing ration (in ppbv) in BG and change in SG-COS of COS (a and b, respectively),  $\text{SO}_2$  (c and d) and  $\text{SO}_4$  (c and d). All quantities are annually averaged over the years 2046-2055.

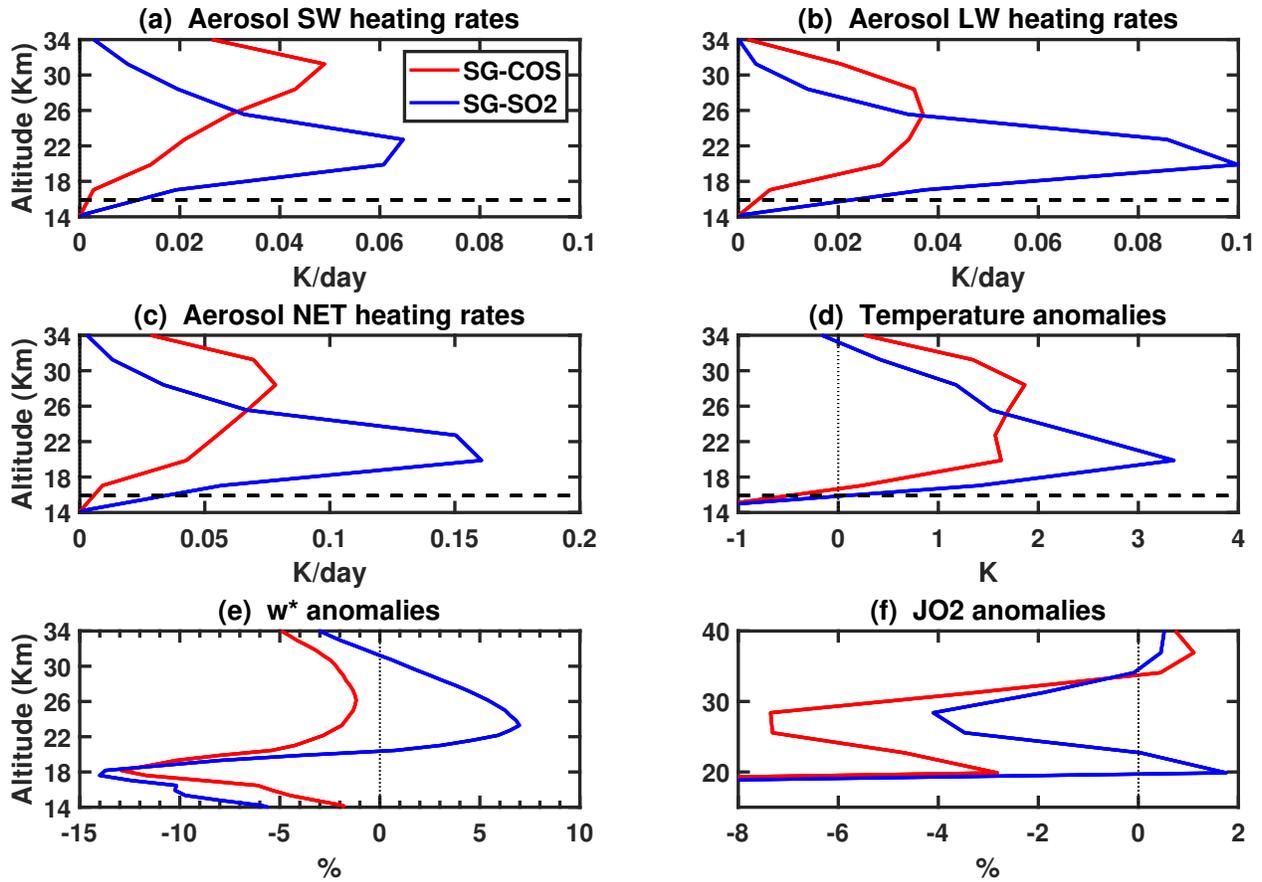


S 3: Monthly values and annually averaged changes [years 2046-2055] with respect to BG of atmospheric stratospheric ozone column (in DU) (panels a and b, respectively), methane lifetime (in yr) (panels c and d), and TTL temperature (in K) (panels e and f).

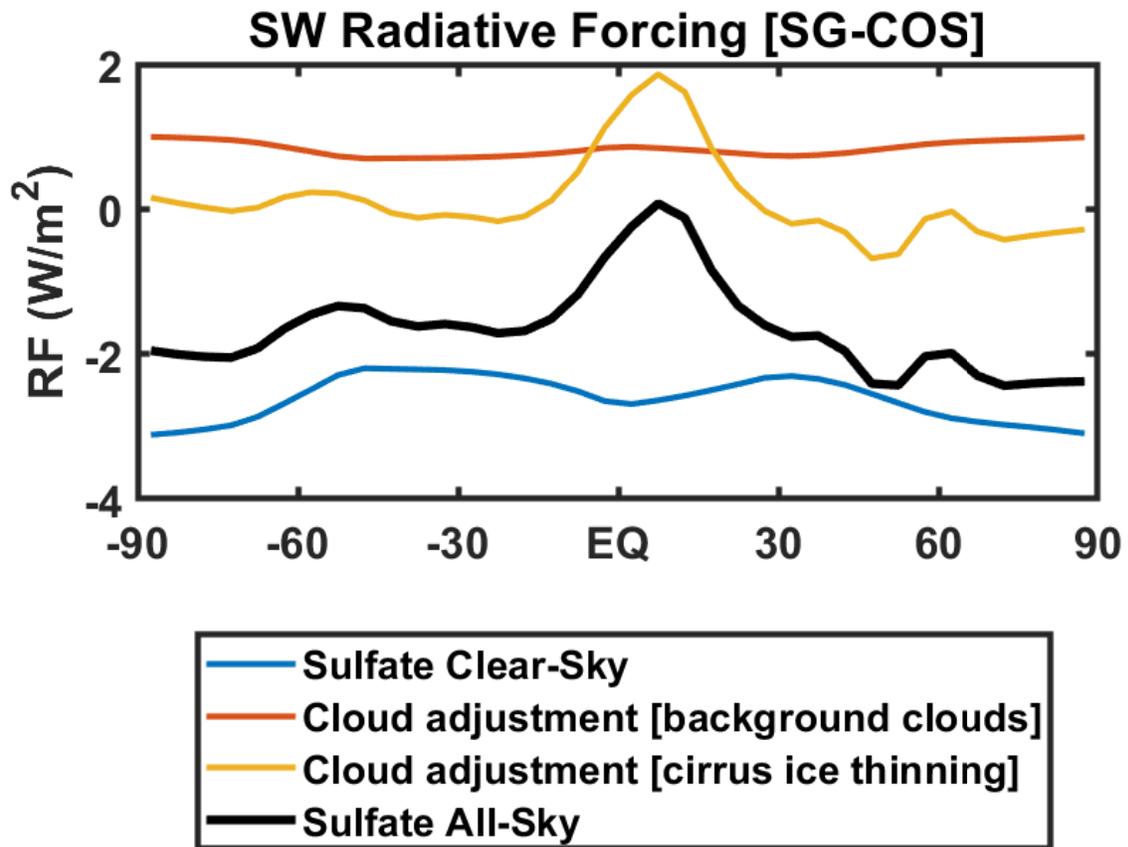
### Aerosol heating rates



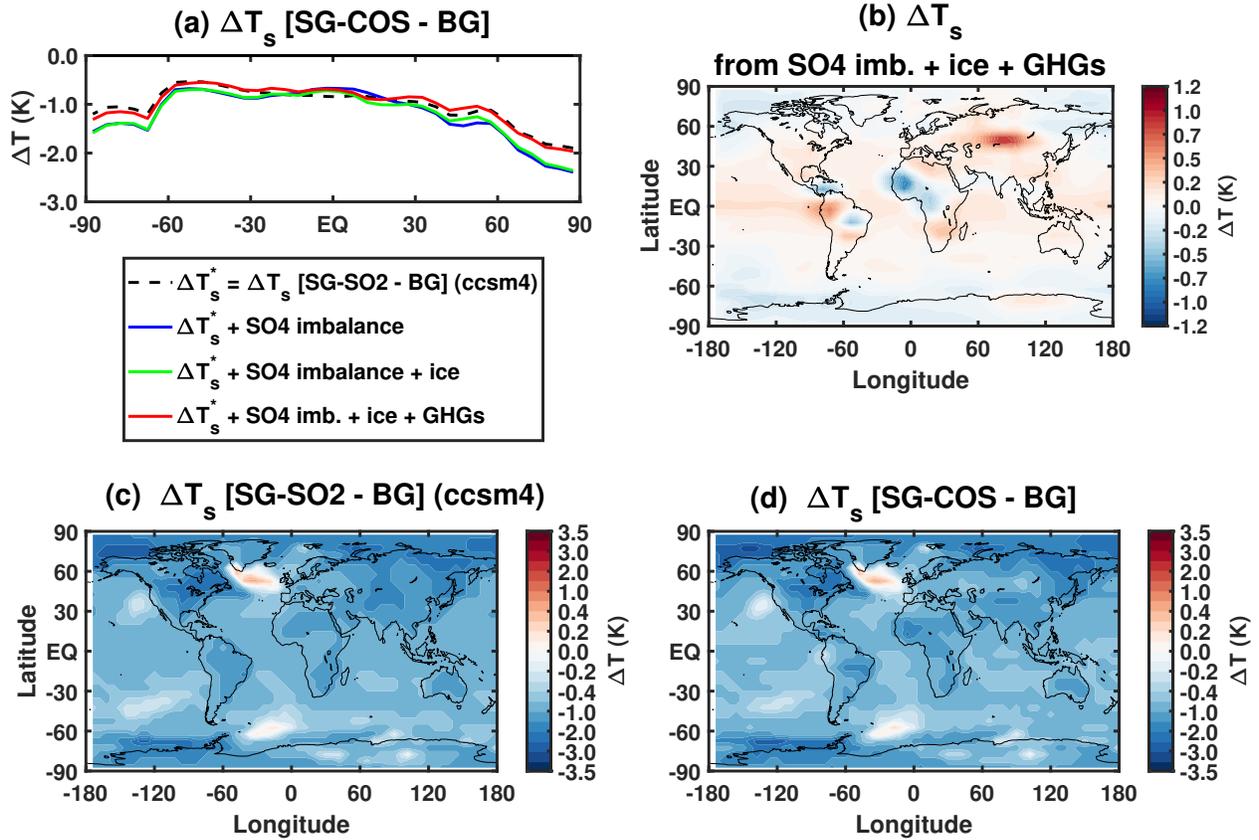
S 4: Mean zonal values of aerosol heating rates for shortwave (SW) and longwave (LW) wavelengths, and net (NET) in SG-COS (a, c and d, respectively), and SG-SO2 (b, d and f). All quantities are annually averaged over the years 2046-2055.



S 5: Tropical vertical profiles of the aerosol shortwave (SW), longwave (LW) and net (NET) heating rates (a, b, c) anomalies (in K/day), temperature anomaly (in K), and residual vertical velocity (d, e) and  $O_2$  photodissociation coefficient percentage anomalies (f). All quantities are annually averaged over the years 2046-2055.



S 6: Latitudinal distribution of zonal mean values of single contribution to shortwave RF (in  $\text{W}/\text{m}^2$ ) in SG-COS: sulfate aerosols in Clear-sky condition (blue), cloud adjustment for presence of background clouds (orange) and for the cirrus ice thinning produced (yellow). Sulfate All-Sky RF in black is the sum of all previous contributions.



S 7: (a) Zonally averaged surface temperature ( $T_s$ , in K) anomalies between SG-COS and BG under different conditions:  $T_s$  anomalies between SG-SO<sub>2</sub> and BG (black dashed line); as above, but adding the  $T_s$  anomalies due to the SO<sub>4</sub> imbalance between SG-COS and SG-SO<sub>2</sub> (blue line); as above, but adding the  $T_s$  anomalies due to cirrus ice changes (green line); as above, but also adding the  $T_s$  anomalies due to GHG changes (red line). (b) Lat-long distribution of the  $T_s$  anomalies calculated online in the ULAQ-CCM model considering cirrus ice changes, the SO<sub>4</sub> imbalance between SG-COS and SG-SO<sub>2</sub> and GHG changes. (c) Lat-long distribution of the  $T_s$  anomalies between SG-SO<sub>2</sub> and BG. (d) Lat-long distribution of the  $T_s$  anomalies between SG-COS and BG.