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An approach to sulfate geoengineering with surface emissions of carbonyl sulfide

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1 Tables

Experiment	Surface upward flux	Chemical production	Surface dry deposition	Chemical loss	Net [sources-sinks]
BG	0.12 ±0.01	0.40±0.01	0.39±0.01	0.13±0.01	0.00±0.01
SG-COS-SRF	40.1 ±0.1	0.39±0.01	31.6 ±0.1	8.8 ±0.1	+0.1 ±0.1
SG-COS-TTL	(0.12+6.0*)±0.1	0.39±0.01	3.5 ±0.1	3.1 ±0.1	-0.1 ±0.1

S 1: Globally-annually averaged COS sources and sinks (Tg-S/yr) [years 2046-2055]. *Additional flux of COS injected in the tropical upper troposphere.

COS sources	Gg-S/yr	%
Surface flux from oceans	39.8	8.3
Surface anthropogenic flux	76.3	15.8
Chem prod from CS2 ocean sources	76.8	15.9
Chem prod from CS2 anthropogenic sources	117.6	24.4
Chem prod from DMS ocean sources	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].

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

Experiment	DMS	SO_2	SO_4	COS	CS_2	H_2S	Total
BG	25.9	63.7	1.41	0.12	0.88	4.0	96.0
SG-COS-SRF	25.9	63.7	1.41	40.1	0.88	4.0	136.0
SG-COS-TTL	25.9	63.7	1.41	6.1	0.88	4.0	102.0
SG-SO2	25.9	67.7	1.41	0.12	0.88	4.0	100.0

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

SG-COS-SRF - BG: Sulfate aerosols RF (W/m^2)	SW	LW	NET
Clear sky	-2.49	+0.48	-2.01
Cloud adjustment [background clouds]	+0.81	-0.06	+0.75
Cloud adjustment [cirrus ice thinning]	+0.26	-0.51	-0.25
Cloud adjusted	-1.42±0.12	-0.09±0.25	-1.51±0.13

S 5: Temperature-adjusted tropopause RF of sulfate aerosols in the SG-COS-SRF case with respect to BG (shortwave, longwave and net) (W/m^2) [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).

SG-COS-TTL - BG: Sulfate aerosols RF (W/m^2)	SW	LW	NET
Clear sky	-2.49	+0.48	-2.01
Cloud adjustment [background clouds]	+0.80	-0.06	+0.74
Cloud adjustment [cirrus ice thinning]	+0.33	-0.64	-0.31
Cloud adjusted	-1.36±0.16	-0.22±0.26	-1.58±0.13

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

SG-SO2 - BG: Sulfate aerosols RF (W/m^2)	**SW**	**LW**	**NET**
Clear sky	-2.72	+0.52	-2.20
Cloud adjustment [background clouds]	+0.80	-0.06	+0.74
Cloud adjustment [cirrus ice thinning]	+0.36	-0.71	-0.35
Cloud adjusted	-1.56±0.10	-0.25±0.23	-1.81±0.13

S 7: As in Table 5, but for the SG-SO2 case.

SG-COS-SRF - BG: Greenhouse gases RF (W/m^2)	**SW**	**LW**	**NET**
COS	0.00	+0.17±0.02	+0.17±0.02
CH_4	0.00	+0.12±0.01	+0.12±0.01
H_2O [stratosphere]	0.00	-0.024±0.004	-0.024±0.004
O_3 [stratosphere]	-0.048±0.005	+0.010±0.001	-0.038±0.005
O_3 [troposphere]	0.00±0.01	+0.02±0.01	+0.02±0.01
Total	-0.05±0.01	+0.30±0.03	+0.25±0.03

S 8: Temperature-adjusted tropopause RF of greenhouse gases in the SG-COS-SRF case with respect to BG (shortwave, longwave and net) (W/m^2) [years 2046-2055]. First five rows present the RF contributions of specific greenhouse gases affected directly and indirectly by SG (*i.e.*, COS, CH_4, stratospheric H_2O, stratospheric and tropospheric O_3. Last row shows the gas net total RF.

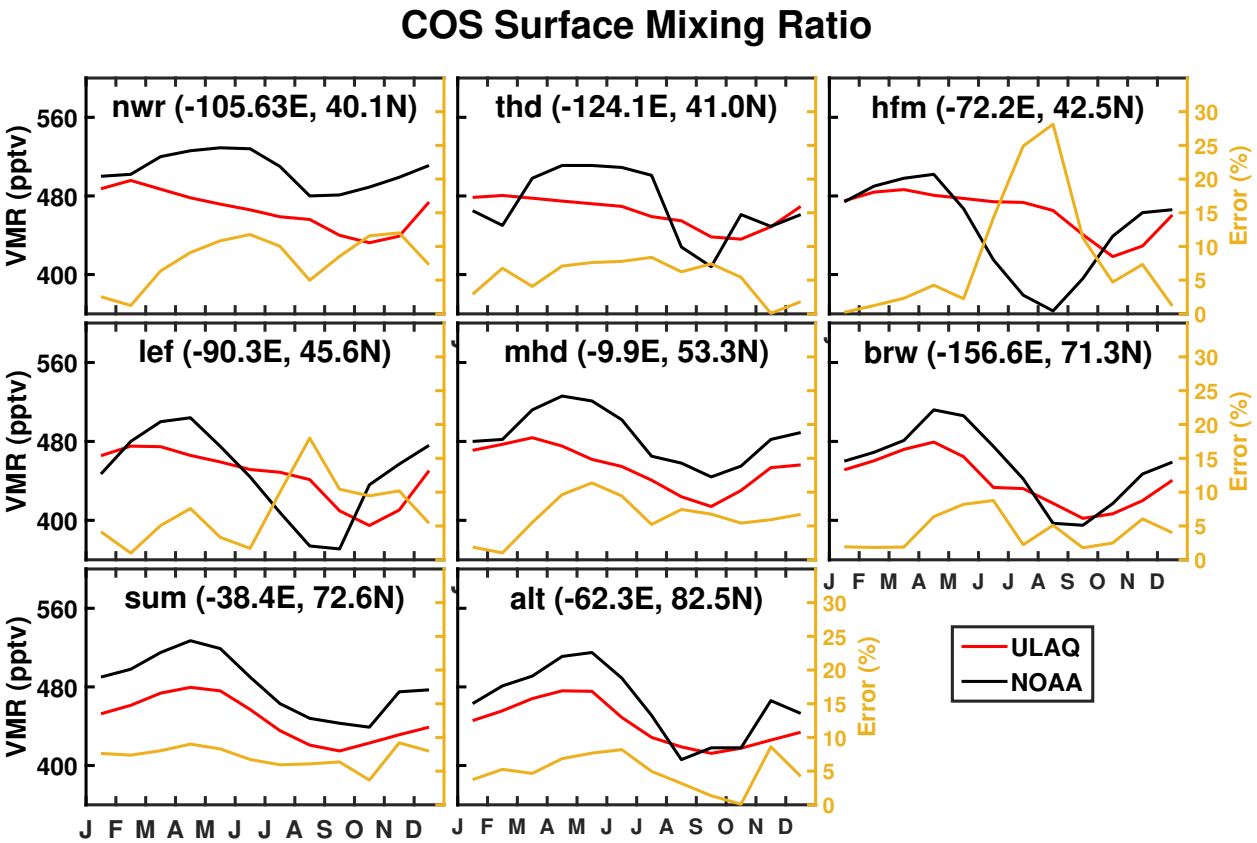
SG-COS-TTL - BG: Greenhouse gases RF (W/m^2)	**SW**	**LW**	**NET**
COS	0.00	0.03±0.01	0.03±0.01
CH_4	0.00	+0.12±0.02	+0.12±0.02
H_2O [stratosphere]	0.00	-0.024±0.004	-0.024±0.004
O_3 [stratosphere]	-0.048±0.005	+0.010±0.001	-0.038±0.005
O_3 [troposphere]	0.00±0.01	+0.02±0.01	+0.02±0.01
Total	-0.05±0.01	+0.16±0.03	+0.11±0.03

S 9: As in Table 8, but for the SG-SO2 case.

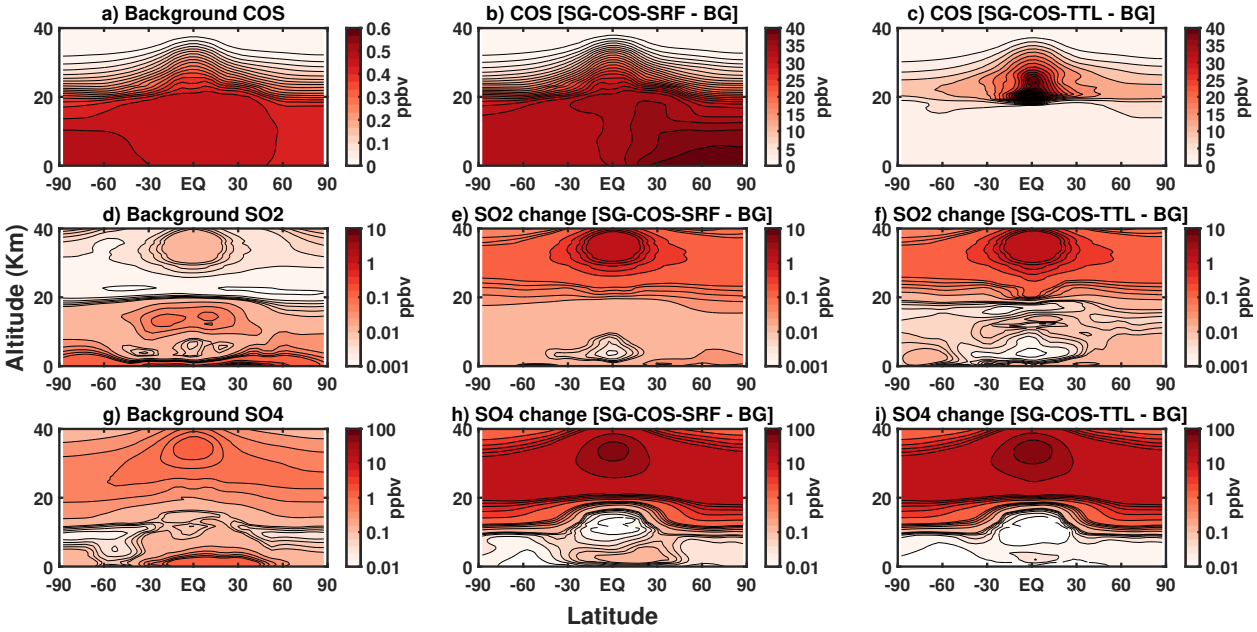
SG-SO2 - BG: Greenhouse gases RF (W/m^2)	SW	LW	NET
COS	0.00	0.00	0.00
CH_4	0.00	+0.11±0.02	+0.11±0.02
H_2O [stratosphere]	0.00	+0.009±0.001	+0.009±0.001
O_3 [stratosphere]	-0.015±0.01	0.00±0.01	-0.015±0.01
O_3 [troposphere]	0.00±0.01	+0.02±0.01	+0.02±0.01
Total	-0.02±0.01	+0.14±0.02	+0.12±0.02

S 10: As in Table 8, but for the SG-SO2 case.

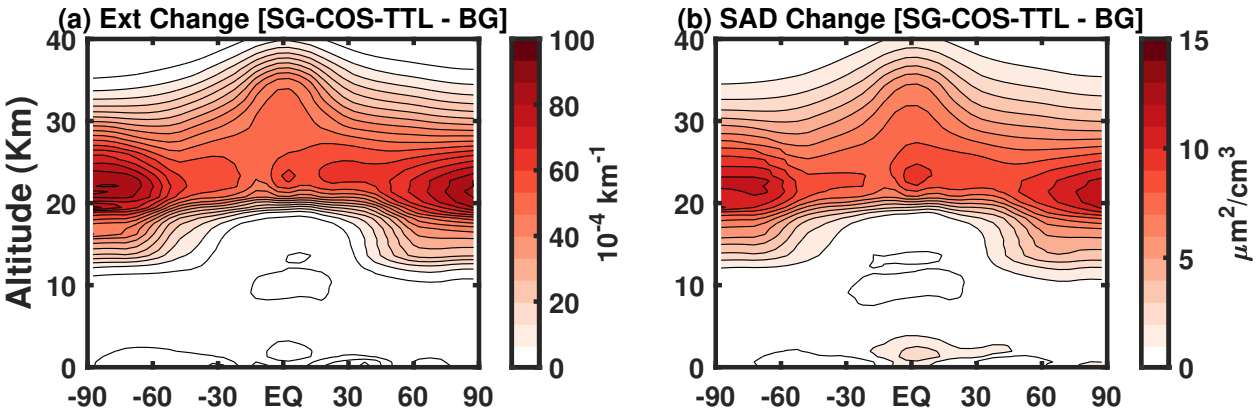
2 Figures



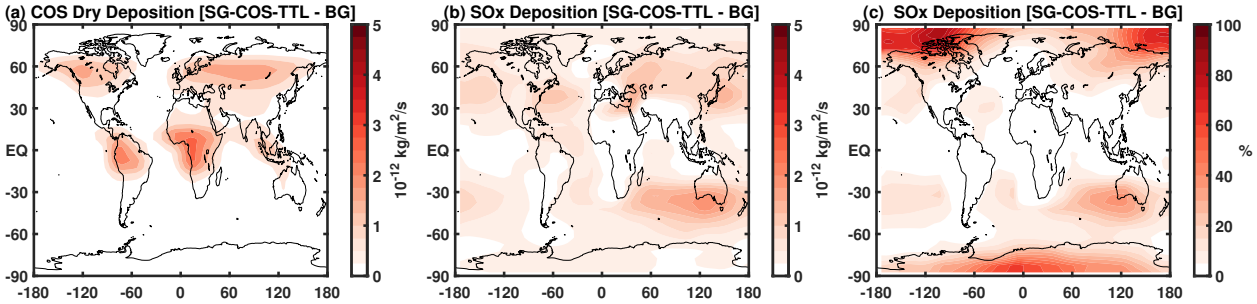
S 1: COS surface mixing ratio (in pptv, left axis) evaluation at NOAA stations: NWR: Niwot Ridge, United States; THD: Trinidad Head, United States; HFM: Harvard Forest, United States; LEF: Wisconsin, United States; MHD: Mace Head, Ireland; BRW: Barrow, United States; SUM: Summit, Greenland; ALT: Alert, 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.



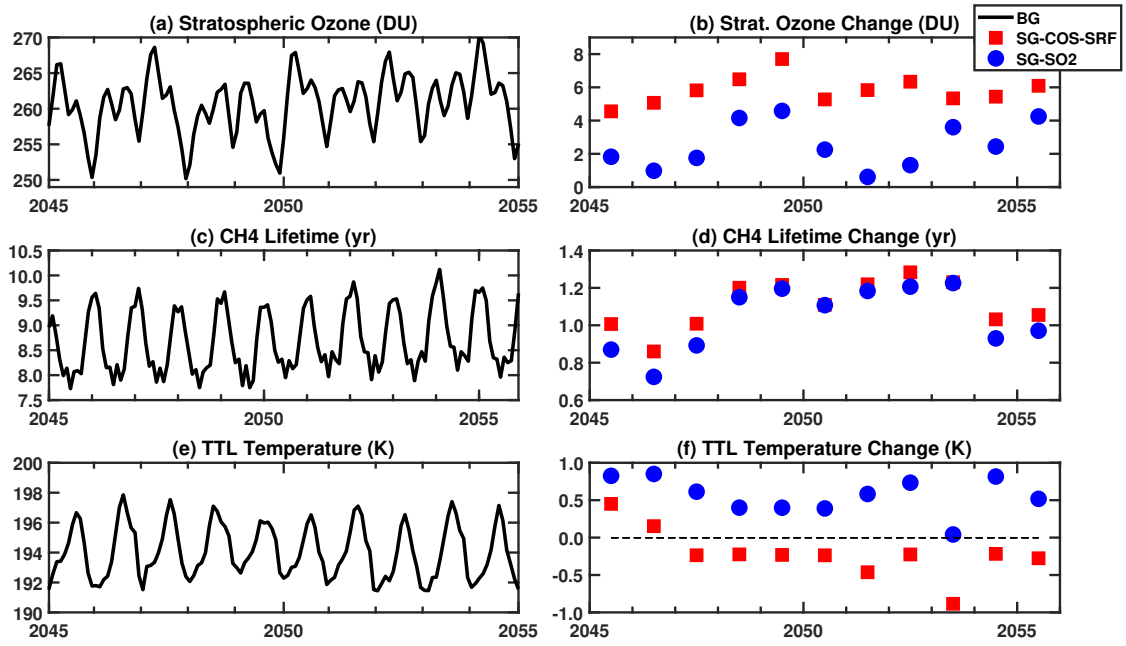
S 2: Mean zonal values of volume mixing ration (in ppbv) in BG and changes in both SG-COS experiments of COS (a, b and c, respectively), SO_2 (d, e and f) and SO_4 (g, h and i) . All quantities are annually averaged over the years 2046-2055.



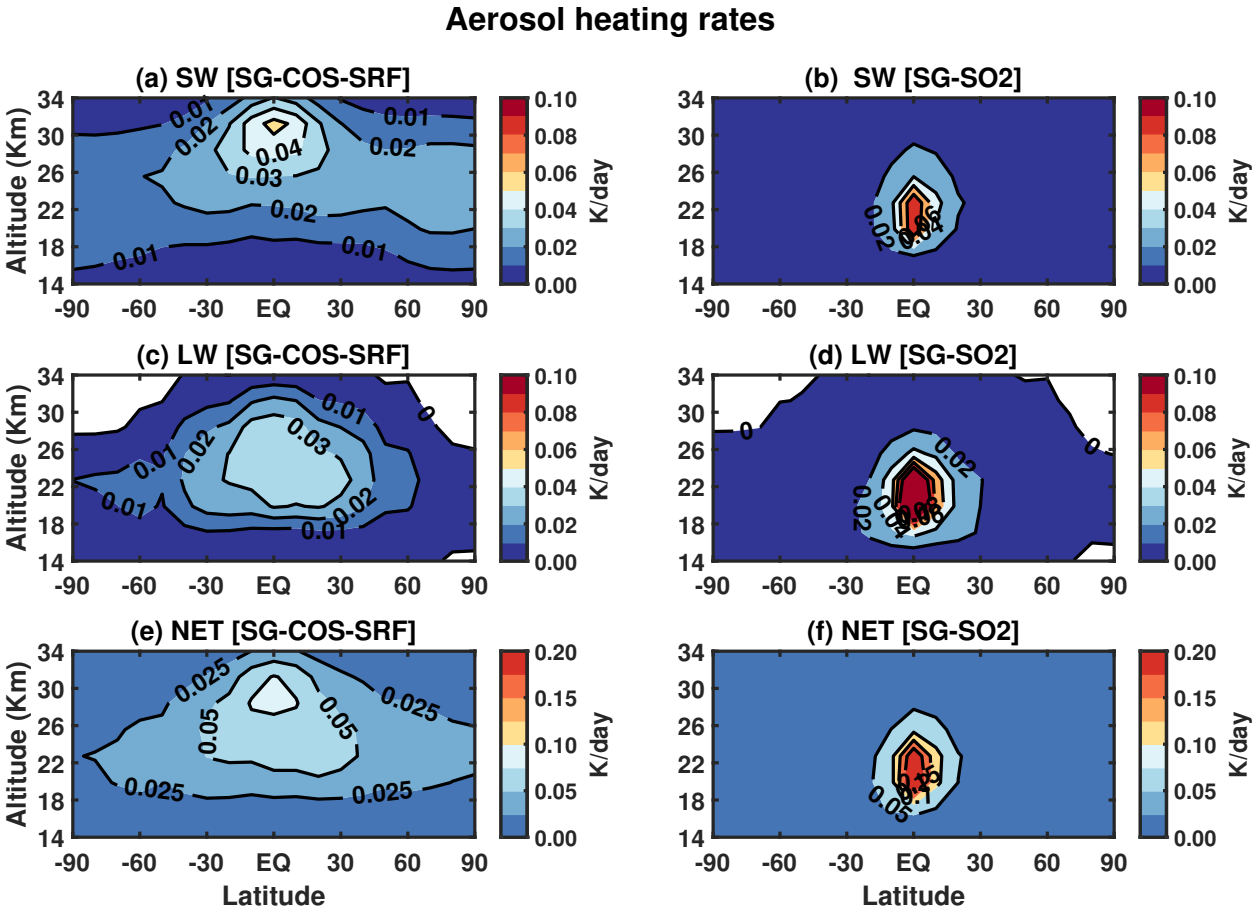
S 3: Zonal mean values of sulfate extinction (in 10^{-4} km^{-1}) and SAD change (in $\mu m^2/cm^3$) in SG-COS-TTL with respect to the background. All quantities are annually averaged over the years 2046-2055.



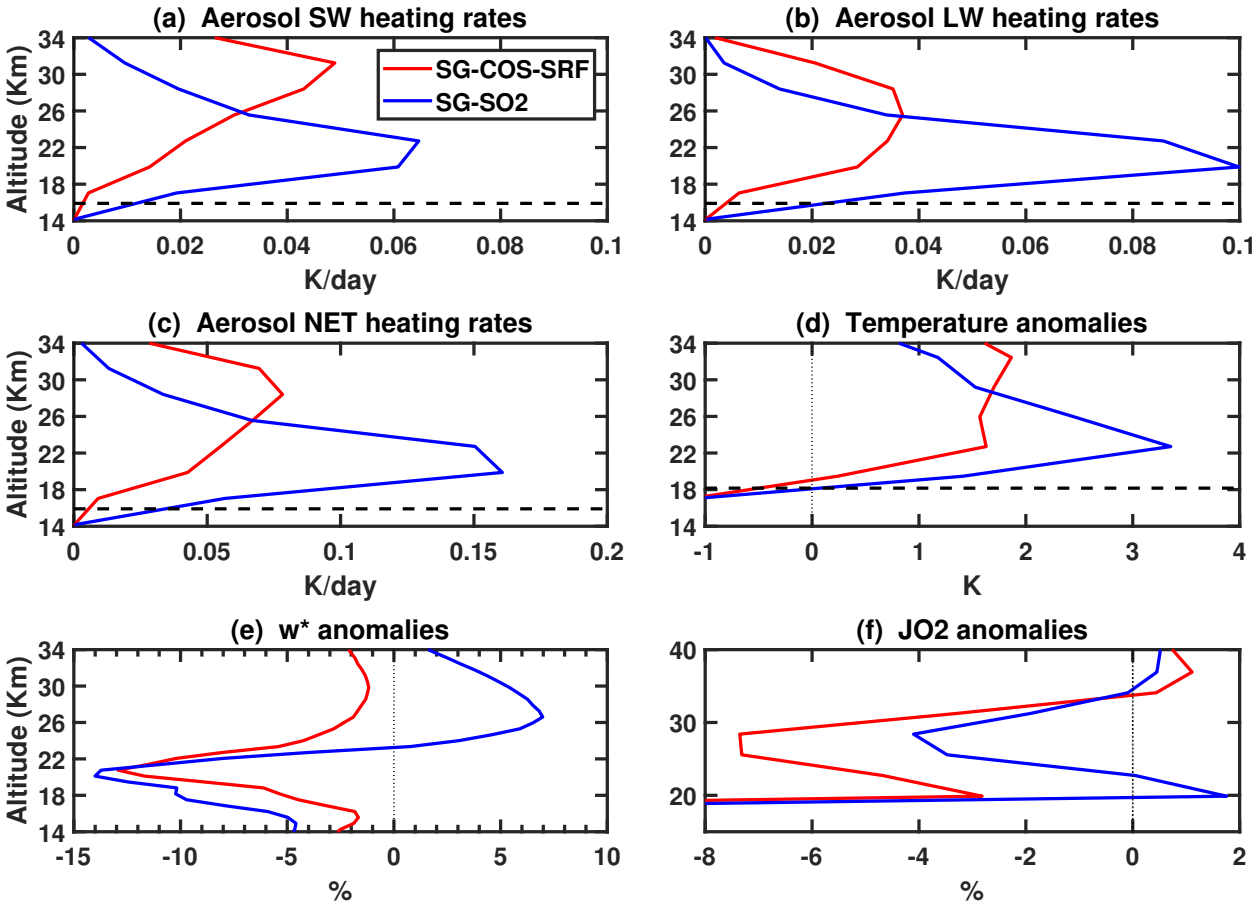
S 4: a) Change in COS dry deposition fluxes in SG-COS-TTL compared to the background. b) Change in SO_x total deposition fluxes in SG-COS-TTL compared to the background. c) as b) but in % of the background values.



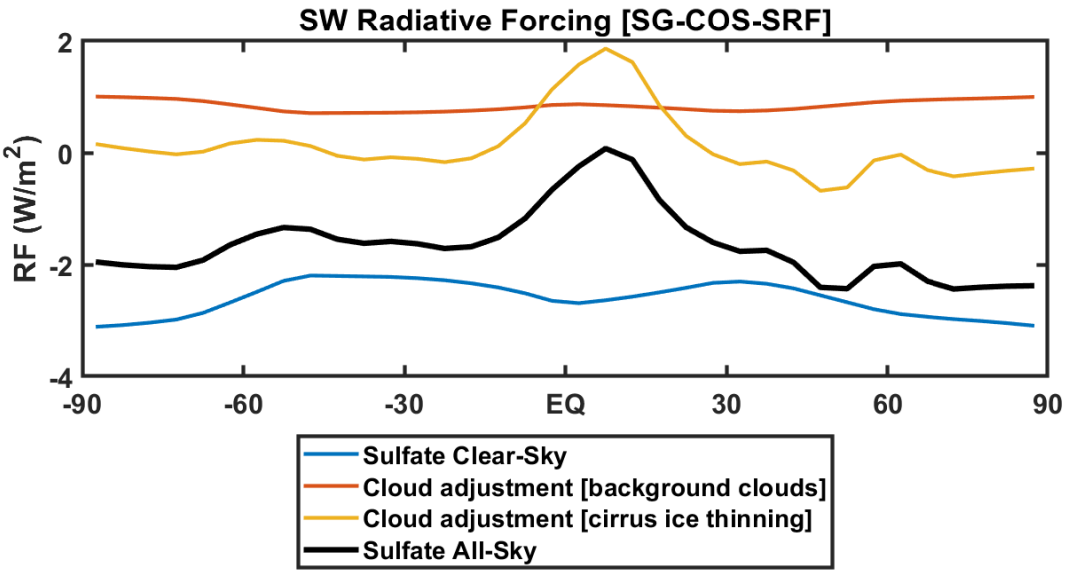
S 5: Monthly values and annually averaged changes in SG-COS-SRF (red) and SG-SO2 (blue) [years 2046-2055] with respect to BG of atmospheric stratospheric ozone column (in DU) (panels a and b, respectively), methane lifetime (in yr) (panels c and d), and TTL temperature (in K) (panels e and f).



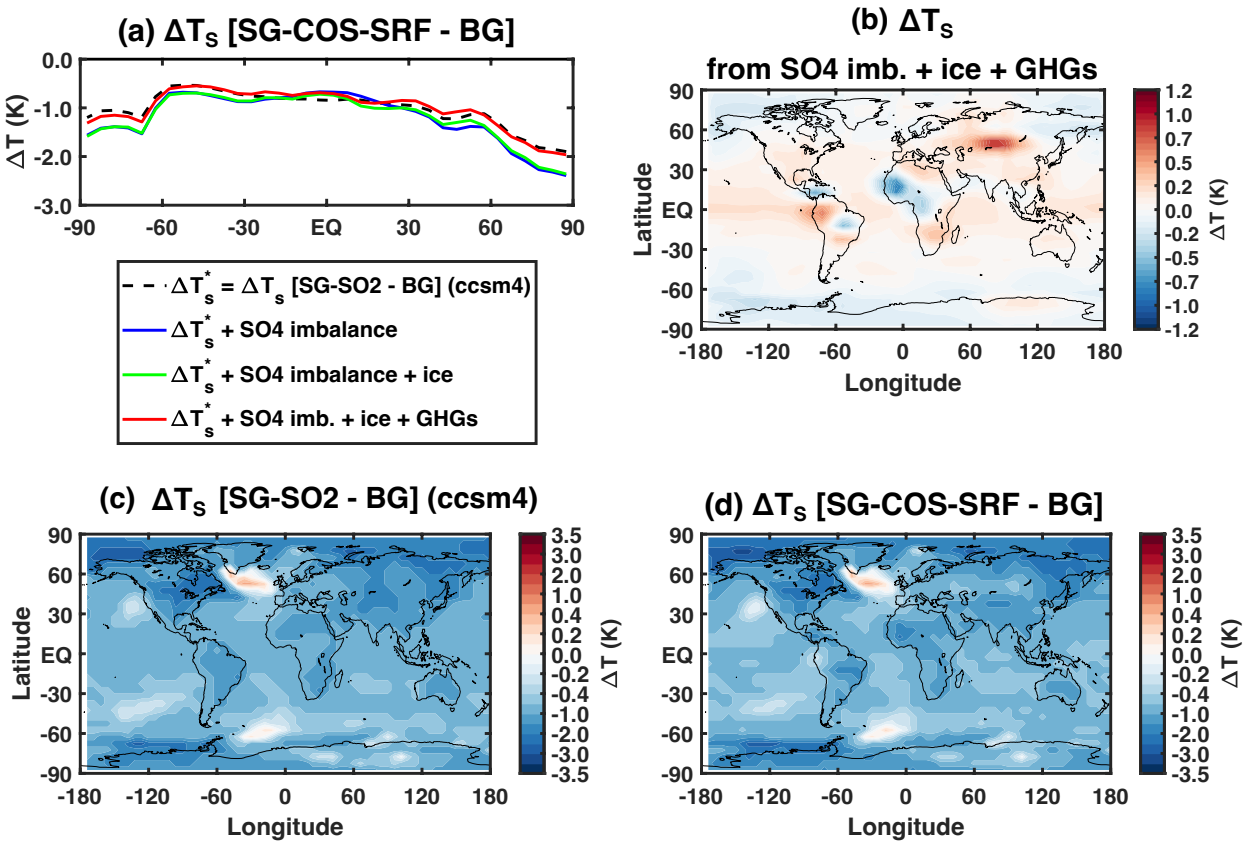
S 6: Mean zonal values of aerosol heating rates for shortwave (SW) and longwave (LW) wavelengths, and net (NET) in SG-COS-SRF (a, c and e, respectively), and SG-SO2 (b, d and f). All quantities are annually averaged over the years 2046-2055.



S 7: 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, d), and residual vertical velocity (in %, e) and O_2 photodissociation coefficient percentage anomalies (f). All quantities are annually averaged over the years 2046-2055.



S 8: Latitudinal distribution of zonal mean values of single contribution to shortwave RF (in W/m^2) in SG-COS-SRF: sulfate aerosols in Clear-1sky condition (blue), cloud adjustment for the 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 9: (a) Zonally averaged surface temperature (T_s, in K) anomalies between SG-COS-SRF and BG under different conditions: T_s anomalies between SG-SO2 and BG (black dashed line); as above, but adding the T_s anomalies due to the SO_4 imbalance between SG-COS-SRF and SG-SO2 (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–lon distribution of the T_s anomalies calculated online in the ULAQ-CCM model considering cirrus ice changes, the SO_4 imbalance between SG-COS-SRF and SG-SO2 and GHG changes. (c) Lat–lon distribution of the T_s anomalies between SG-SO2 and BG. (d) Lat–lon distribution of the T_s anomalies between SG-COS-SRF and BG.