



## Supplement of

## Impacts of stratospheric sulfate geoengineering on tropospheric ozone

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11	CESM1 (CAM-Chem) has been evaluated for the troposphere (Tilmes et al., 2016b) and
12	has also been used for studies in the stratosphere (Fernandez et al., 2017). Here, we use the same
13	model setup but for a higher horizontal resolution of $0.9^{\circ}$ x $1.25^{\circ}$ (called here the $1^{\circ}$ version)
14	instead of $1.9^{\circ} \ge 2.5^{\circ}$ (2° version), which is the version that is participating in the Chemistry
15	Climate Model Initiative. Some differences in stratospheric column ozone between model
16	versions occur (Fig. S1), likely due to slight differences in the stratospheric dynamics, for
17	instance as result of differences in gravity waves. The 1° model shows some improvement in
18	stratospheric column ozone in high northern latitudes in winter and spring as well as in summer
19	in the high southern latitudes compared to the 2° version with regard to a present-day ozone
20	climatology based on Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder
21	(MLS) satellite observations between 2004 and 2010, compiled by Ziemke et al. (2011).
22	However, it also indicates an overestimation of the Antarctic ozone hole in October. Besides
23	these differences, both versions reproduce observed column ozone very well.
24	Tropospheric ozone and other tracers (not shown) in both the 1° and the 2° model versions
25	are very similar (Fig. S2) and are therefore not further discussed. A detailed description of the
26	performance of the 2° simulation is given in Tilmes et al. (2016b).
27	
28	Reference:

Tilmes, S., Lamarque, J.-F., Emmons, L. K., Conley, A., Schultz, M. G., Saunois, M., Thouret, V.,
Thompson, A. M., Oltmans, S. J., Johnson, B., and Tarasick, D.: Technical Note: Ozonesonde
climatology between 1995 and 2011: description, evaluation and application, Atmos. Chem.
Phys., 12, 7475-7497, doi:10.5194/acp-12-7475-2012, 2012.

Ziemke, J. R., Chandra, S., Labow, G. J., Bhartia, P. K., Froidevaux, L., and Witte, J. C.: A global
 climatology of tropospheric and stratospheric ozone derived from Aura OMI and MLS
 measurements, Atmos. Chem. Phys., 11, 9237-9251, doi:10.5194/acp-11-9237-2011, 2011.





Figure S1. Monthly and zonally averaged stratospheric ozone column (in DU) comparison 38 between the 10°N to 10°S gridded present day MLS/OMI satellite product (Ziemke et al., 2011) 39 (black), CAM-Chem 1° simulation (red), and CAM-Chem 2° simulation (blue) for 2004-2010, 40 shown for four months (different panels). The model tropopause to derive the stratospheric 41 column is defined as the 150 ppb ozone level, while the climatological tropopause uses the 42 World Meteorological Organization definition from the National Centers for Environmental 43 Prediction. This may lead to small differences between observations and model simulations, but 44 not between model experiments themselves. Model results are interpolated to the same grid as 45 46 the observations and error bars indicate the  $\pm 1$  standard deviation of the interannual variability for each latitude interval. 47



51 Figure S2. Regionally aggregated seasonal cycle comparisons of vertical measurements from

- 52 ozone soundings (in ppb) averaged between 1995 and 2010 (black lines) (Tilmes et al., 2012), and
- 53 CAM-Chem 1° results (red), and CAM-Chem 2° results (blue) averaged between 2005 and 2010,
- 54 interpolated to 900 mb (top) and 500 mb (bottom).



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**Figure S3**. Global map of seasonal surface temperature differences (K) between G4SSA and RCP6.0 (left column), G4SSA-S and RCP6.0 (middle column) and G4SSA and G4SSA-S (right column) for 2030-2069. Hatched regions are areas with p > 0.05 (where changes are not statistically significant based on a paired *t*-test).



**Figure S4**. Surface water flux differences, shown as G4SSA minus RCP6.0 and G4SSA-S minus

64 RCP6.0 for 2030-2069. P is precipitation. E is total evaporation. GE is ground evaporation,

65 which is the sum of soil evaporation, snow evaporation, soil sublimation, and snow sublimation

66 minus dew. CE is canopy evaporation and T is transpiration. For P, positive is downward, and

67 for all the other fluxes, positive is upward.



**Figure S5**. Global averaged annual total net flux on the top of the model. Lines are the average

of three ensemble members. Geoengineering starts at 1 January 2020 and ends at 1 January2070.



**Figure S6.** Global map of surface ozone concentration (ppb) in (a) RCP6.0, (b) G4SSA and (c)

<sup>77</sup> G4SSA-S for 2030-2069.



- 81 RCP6.0, (b) G4SSA-S and RCP6.0 and (c) G4SSA and G4SSA-S for 2030-2069. Hatched
- 82 regions are areas with p > 0.05.

**Figure S7.** Global map of surface ozone concentration change percentage (%) in (a) G4SSA and



**Figure S8.** Global map of seasonal surface ozone concentration differences (ppb) between

- 87 G4SSA and RCP6.0 (left column), G4SSA-S and RCP6.0 (middle column) and G4SSA and
- 64SSA-S (right column) for 2030-2069. Hatched regions are areas with p > 0.05.



Figure S9. Zonal mean water vapor mixing ratio differences (g kg<sup>-1</sup>) in the geoengineering
experiments (a) G4SSA minus RCP6.0, (b) G4SSA-S minus RCP6.0, and (c) G4SSA minus

94 G4SSA-S. These are averaged for three ensemble members for years 2030-2069. Hatched

95 regions are areas with p > 0.05.

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**Figure S10.** Global map of differences of column NO produced by lightning (Tg N yr<sup>-1</sup>)

between (a), G4SSA and RCP6.0, (b) G4SSA-S and RCP6.0, and (c) G4SSA and G4SSA-S for 2030-2069. Hatched regions are areas with p > 0.05.



Figure S11. Global map of surface water vapor mixing ratio difference (g kg<sup>-1</sup>) between G4SSA and G4SSA-S over the period of years 2030-2069. Hatched regions are areas with p > 0.05.

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Figure S12. Global map of surface NO<sub>x</sub> concentration (ppb) in (a) RCP6.0, (b) G4SSA and (c)
 G4SSA-S for 2030-2069.



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114 Figure S13. Global map of seasonal surface bio-emitted isoprene concentration differences

115 (ppb) between G4SSA and RCP6.0 (left column), G4SSA-S and RCP6.0 (middle column) and

- 116 G4SSA and G4SSA-S (right column) for 2030-2069. Hatched regions are areas with p > 0.05.
- 117





**Figure S14**. Zonal mean seasonal ozone concentration differences (ppb) between G4SSA and

- 120 RCP6.0 (top row), G4SSA-S and RCP6.0 (middle row) and G4SSA and G4SSA-S (bottom row)
- 121 for 2030-2069.



Figure S15. Global map of surface O<sub>3</sub><sup>Strat</sup> differences (ppb) between (a), G4SSA and RCP6.0, 124 (b) G4SSA-S and RCP6.0, and (c) G4SSA and G4SSA-S for 2030-2069. Hatched regions are 125 areas with p > 0.05. There is much less  $O_3^{Strat}$  at the surface in G4SSA relative to RCP6.0 as 126 well as G4SSA-S. Changes in O3<sup>Strat</sup> at the surface are on the one hand due to reduced ozone in 127 the stratosphere, and on the other hand due to to changes in the rate of STE. Although the 128 absolute value of O3<sup>Strat</sup> is overestimated, because of a missing loss process via dry depostion in 129 the version of the model, it can be qualitatively used to compare the two scenarios, since dry 130 deposion is not expected to change significantly. 131