

9 CESM1 (CAM-Chem) has been evaluated for the troposphere (Tilmes et al., 2016b) and
10 has also been used for studies in the stratosphere (Fernandez et al., 2017). Here, we use the same
11 model setup but for a higher horizontal resolution of $0.9^\circ \times 1.25^\circ$ (called here the 1° version)
12 instead of $1.9^\circ \times 2.5^\circ$ (2° version), which is the version that is participating in the Chemistry
13 Climate Model Initiative. Some differences in stratospheric column ozone between model
14 versions occur (Fig. S1), likely due to slight differences in the stratospheric dynamics, for
15 instance as result of differences in gravity waves. The 1° model shows some improvement in
16 stratospheric column ozone in high northern latitudes in winter and spring as well as in summer
17 in the high southern latitudes compared to the 2° version with regard to a present-day ozone
18 climatology based on Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder
19 (MLS) satellite observations between 2004 and 2010, compiled by Ziemke et al. (2011).
20 However, it also indicates an overestimation of the Antarctic ozone hole in October. Besides
21 these differences, both versions reproduce observed column ozone very well.

22 Tropospheric ozone and other tracers (not shown) in both the 1° and the 2° model versions
23 are very similar (Fig. S2) and are therefore not further discussed. A detailed description of the
24 performance of the 2° simulation is given in Tilmes et al. (2016b).

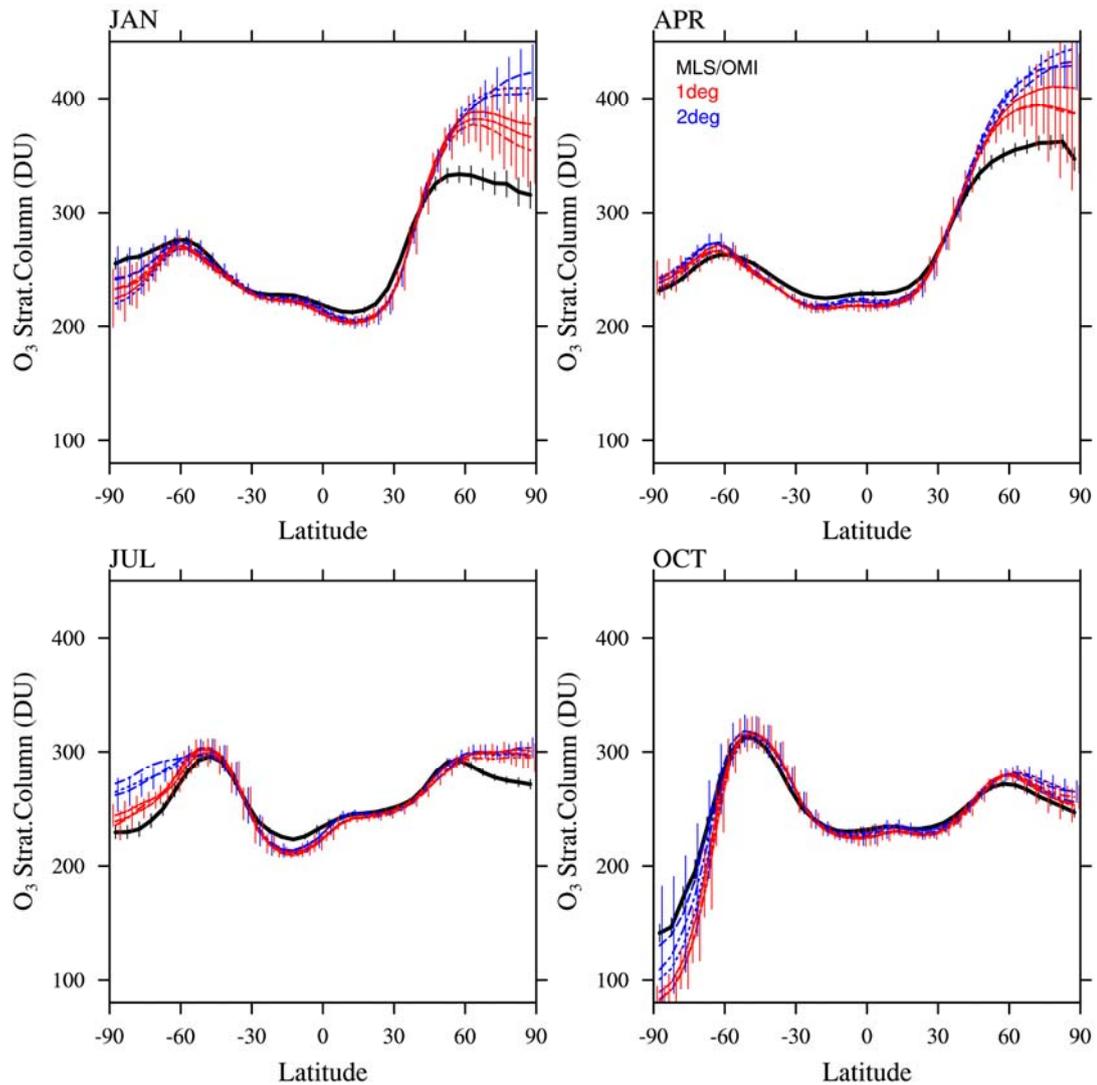
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26 Reference:

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

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

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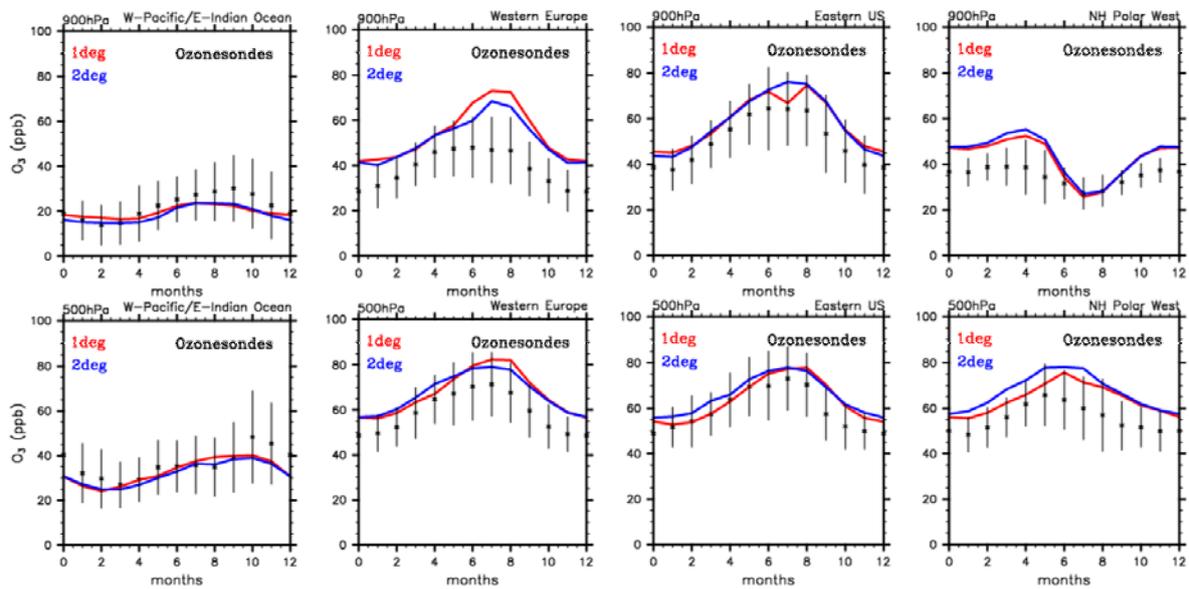


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36 **Figure S1.** Monthly and zonally averaged stratospheric ozone column (in DU) comparison
 37 between the 10°N to 10°S gridded present day MLS/OMI satellite product (Ziemke et al., 2011)
 38 (black), CAM-Chem 1° simulation (red), and CAM-Chem 2°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

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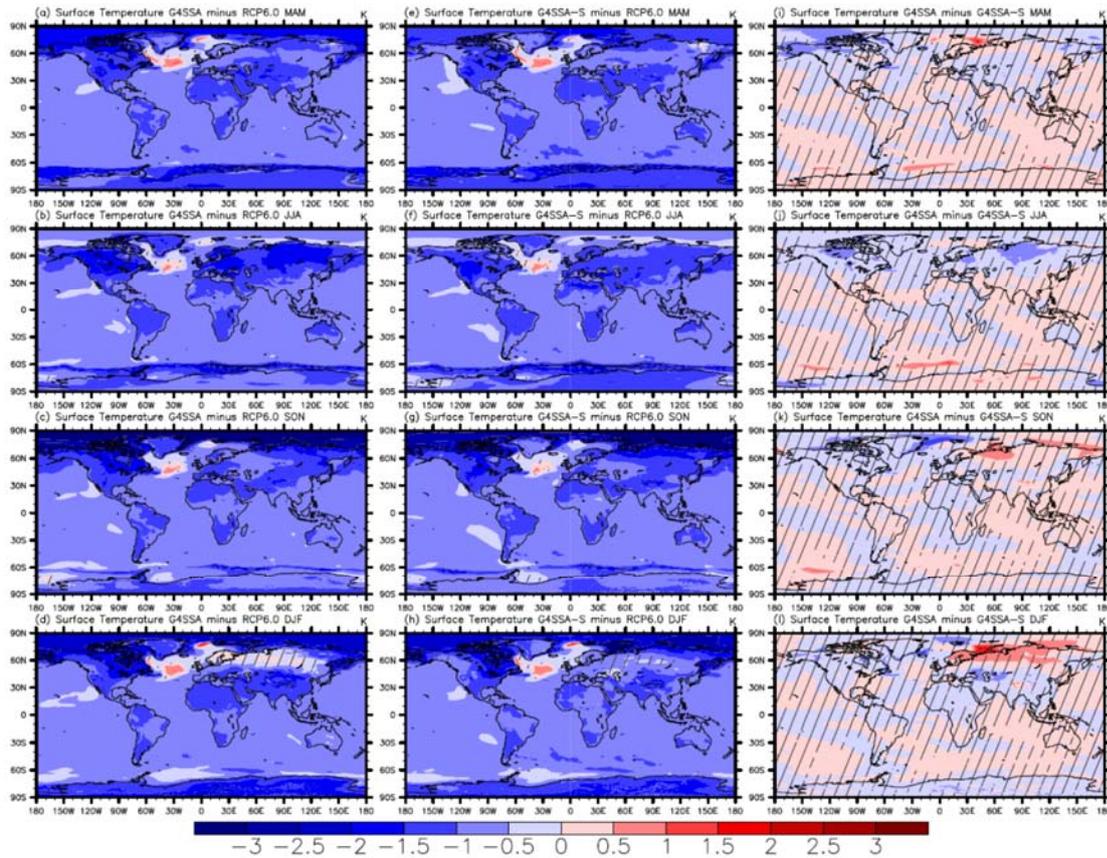
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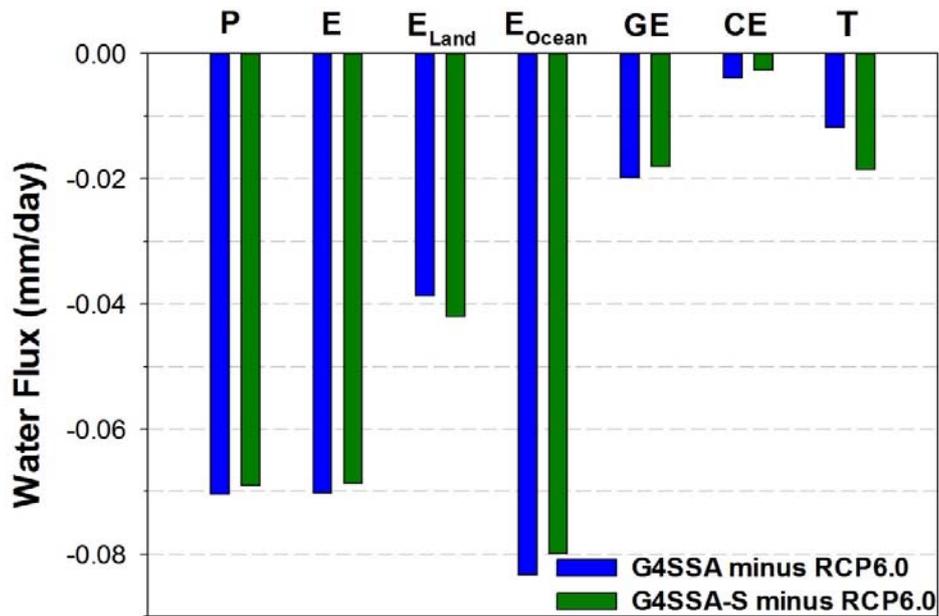
49 **Figure S2.** Regionally aggregated seasonal cycle comparisons of vertical measurements from
 50 ozone soundings (in ppb) averaged between 1995 and 2010 (black lines) (Tilmes et al., 2012), and
 51 CAM-Chem 1° results (red), and CAM-Chem 2° results (blue) averaged between 2005 and 2010,
 52 interpolated to 900 mb (top) and 500 mb (bottom).

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54

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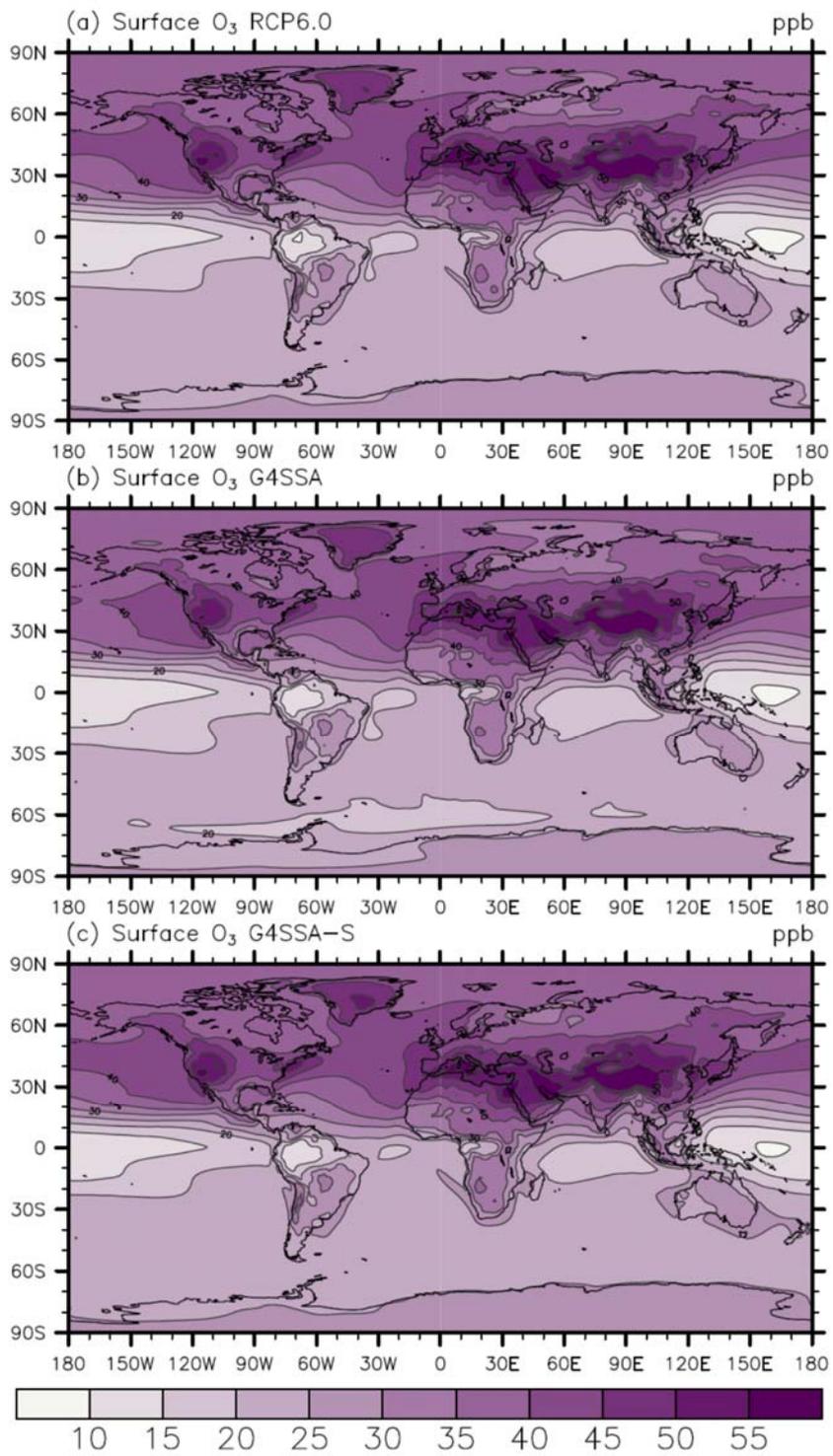


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61 **Figure S4.** Surface water flux differences, shown as G4SSA minus RCP6.0 and G4SSA-S minus
 62 RCP6.0 for 2030-2069. P is precipitation. E is total evaporation. GE is ground evaporation,
 63 which is the sum of soil evaporation, snow evaporation, soil sublimation, and snow sublimation
 64 minus dew. CE is canopy evaporation and T is transpiration. For P, positive is downward, and
 65 for all the other fluxes, positive is upward.

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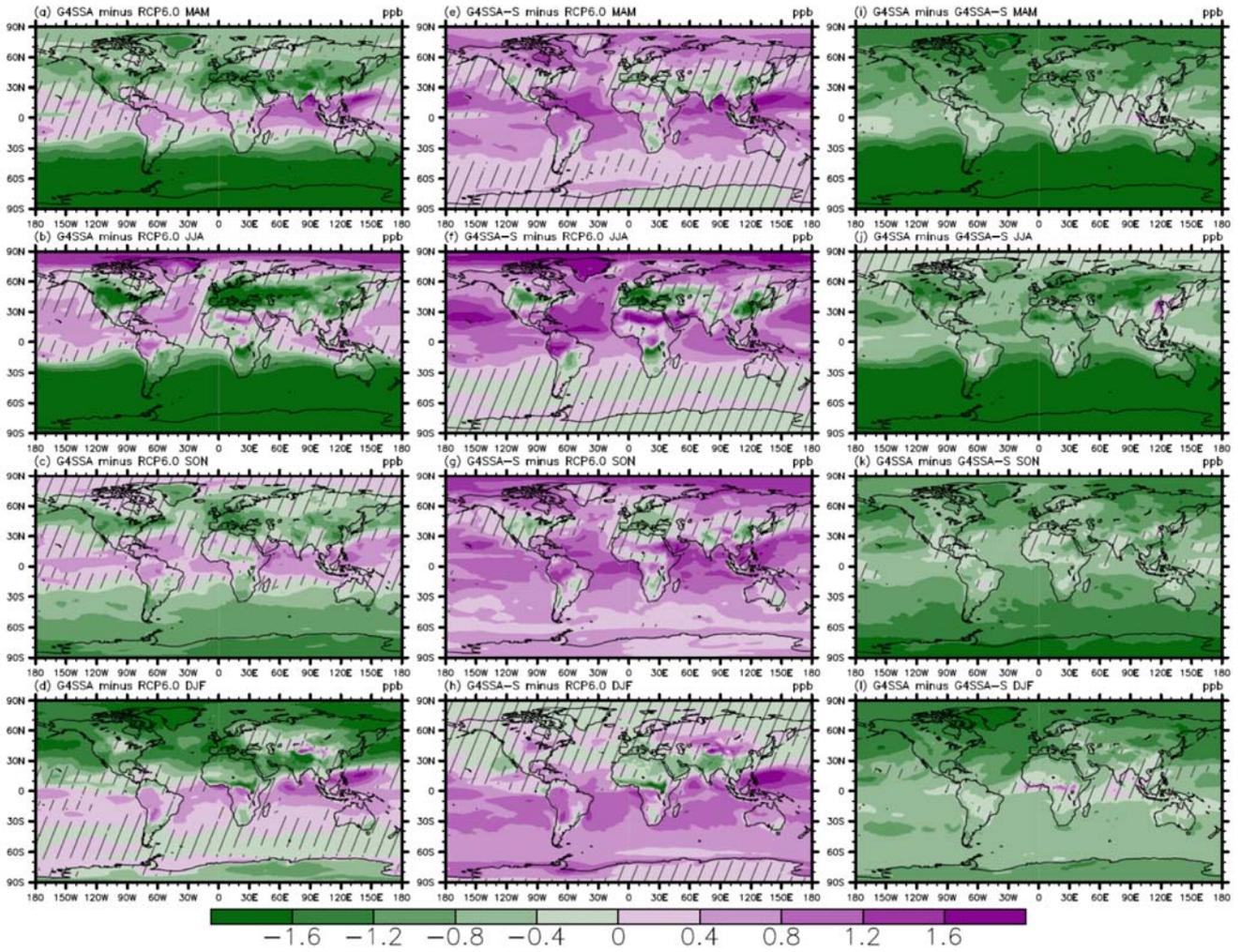


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69 **Figure S5.** Global map of surface ozone concentration (ppb) in (a) RCP6.0, (b) G4SSA and (c)
 70 G4SSA-S for 2030-2069.

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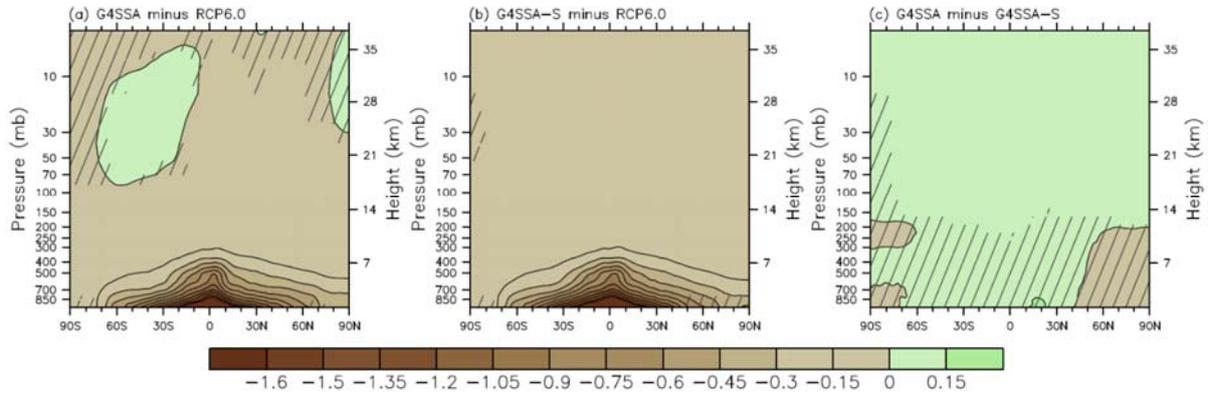


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74 **Figure S6.** Global map of seasonal surface ozone concentration differences (ppb) between
75 G4SSA and RCP6.0 (left column), G4SSA-S and RCP6.0 (middle column) and
76 G4SSA and G4SSA-S (right column) for 2030-2069. Hatched regions are areas with $p > 0.05$.

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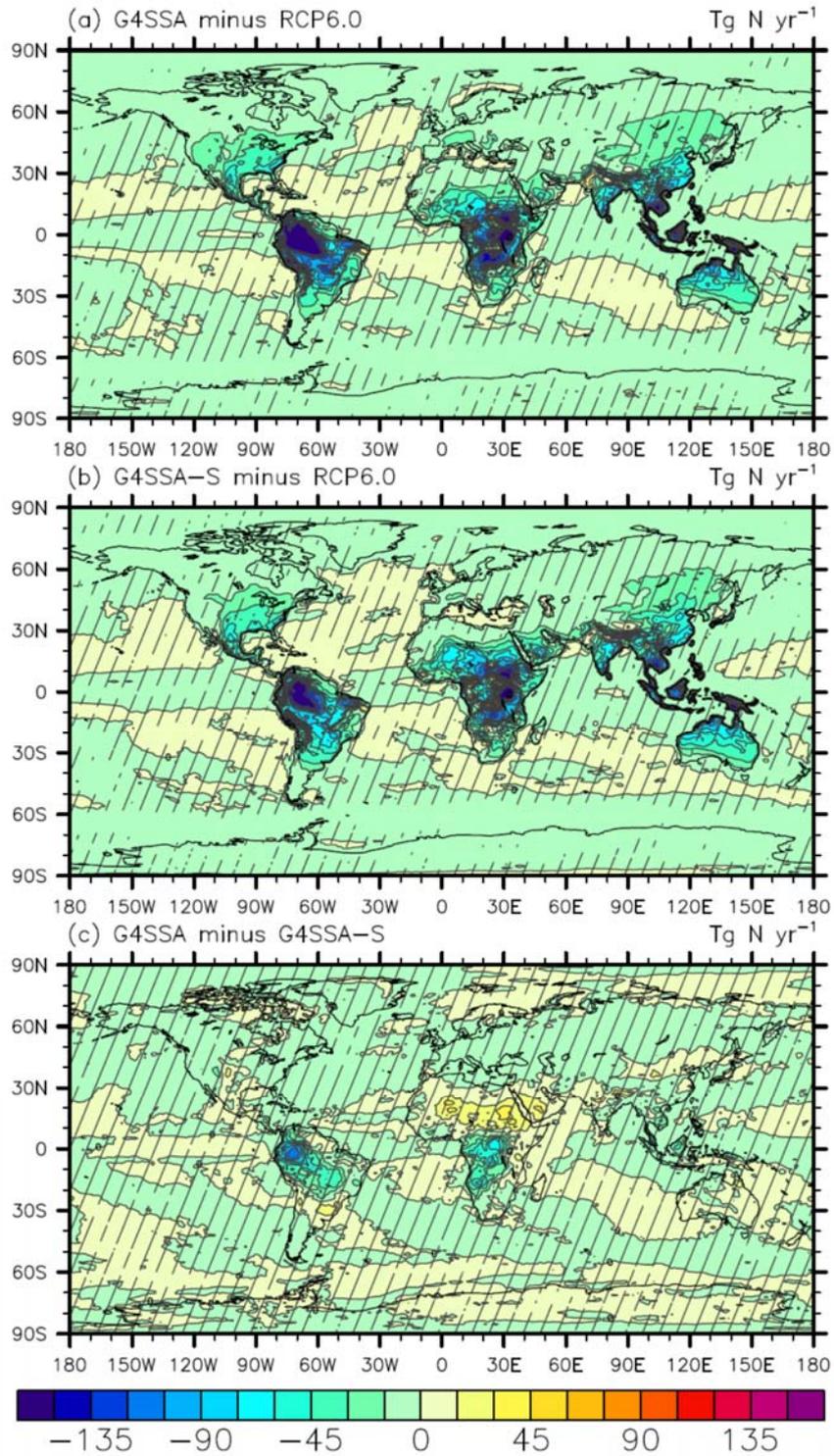


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80 **Figure S7.** Zonal mean water vapor mixing ratio differences (g kg^{-1}) in the geoengineering
 81 experiments (a) G4SSA minus RCP6.0, (b) G4SSA-S minus RCP6.0, and (c) G4SSA minus
 82 G4SSA-S. These are averaged for three ensemble members for years 2030-2069. Hatched
 83 regions are areas with $p > 0.05$.

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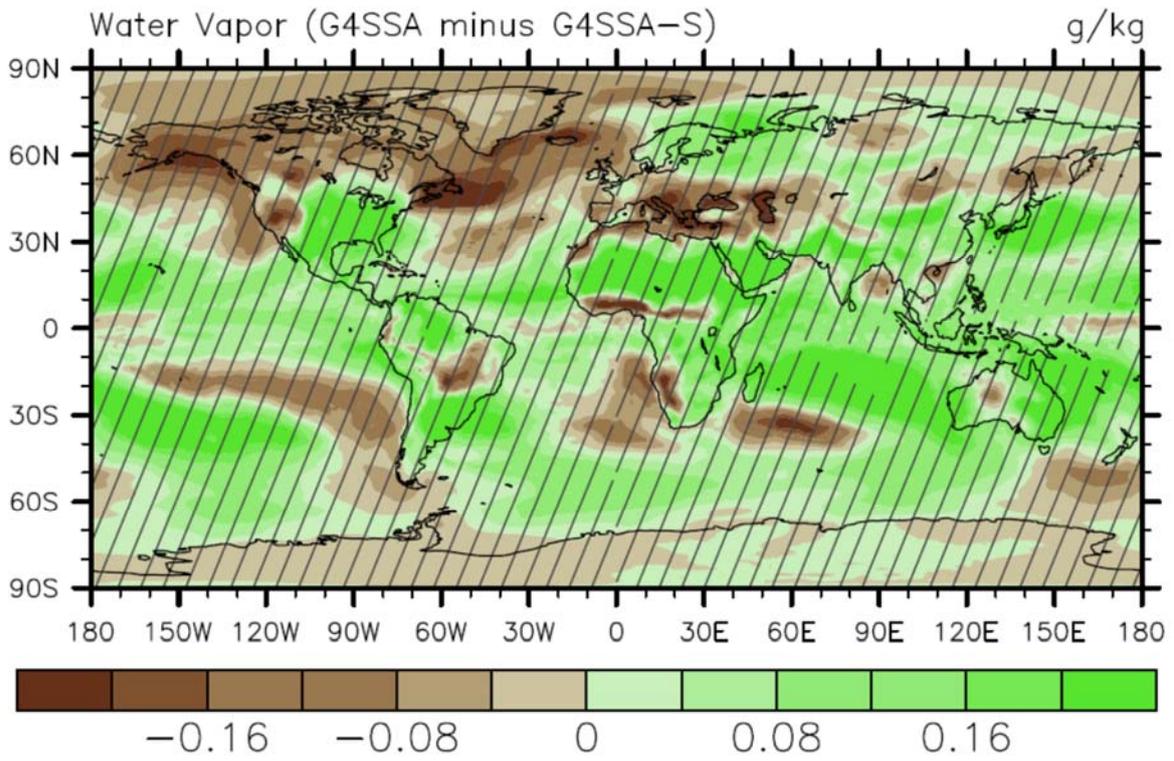
87 **Figure S8.** Global map of differences of column NO produced by lightning (Tg N yr⁻¹) between
 88 (a), G4SSA and RCP6.0, (b) G4SSA-S and RCP6.0, and (c) G4SSA and G4SSA-S for 2030-
 89 2069. Hatched regions are areas with $p > 0.05$.

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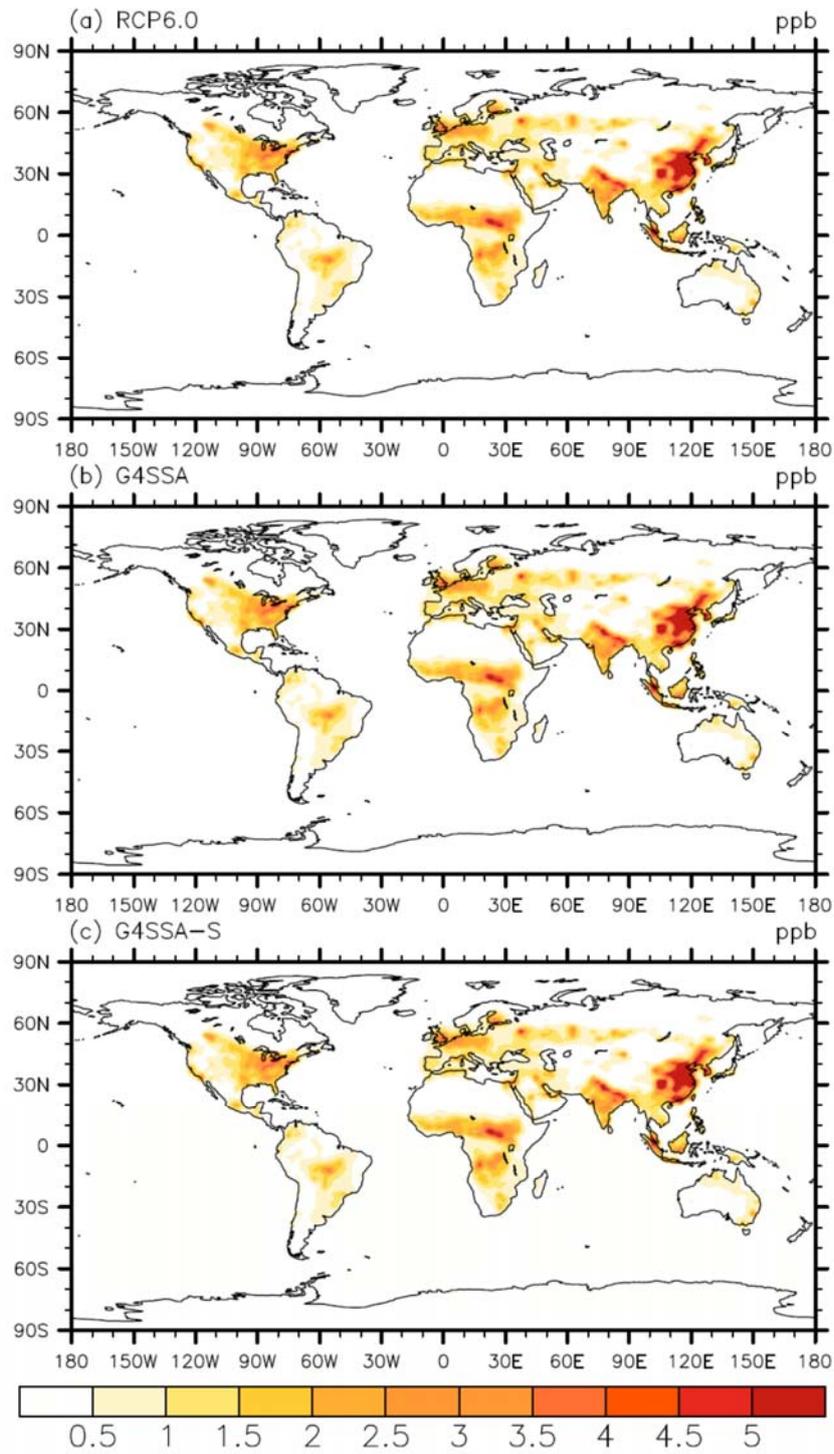


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95 **Figure S9.** Global map of surface water vapor mixing ratio difference (g kg^{-1}) between G4SSA
96 and G4SSA-S over the period of years 2030-2069. Hatched regions are areas with $p > 0.05$.

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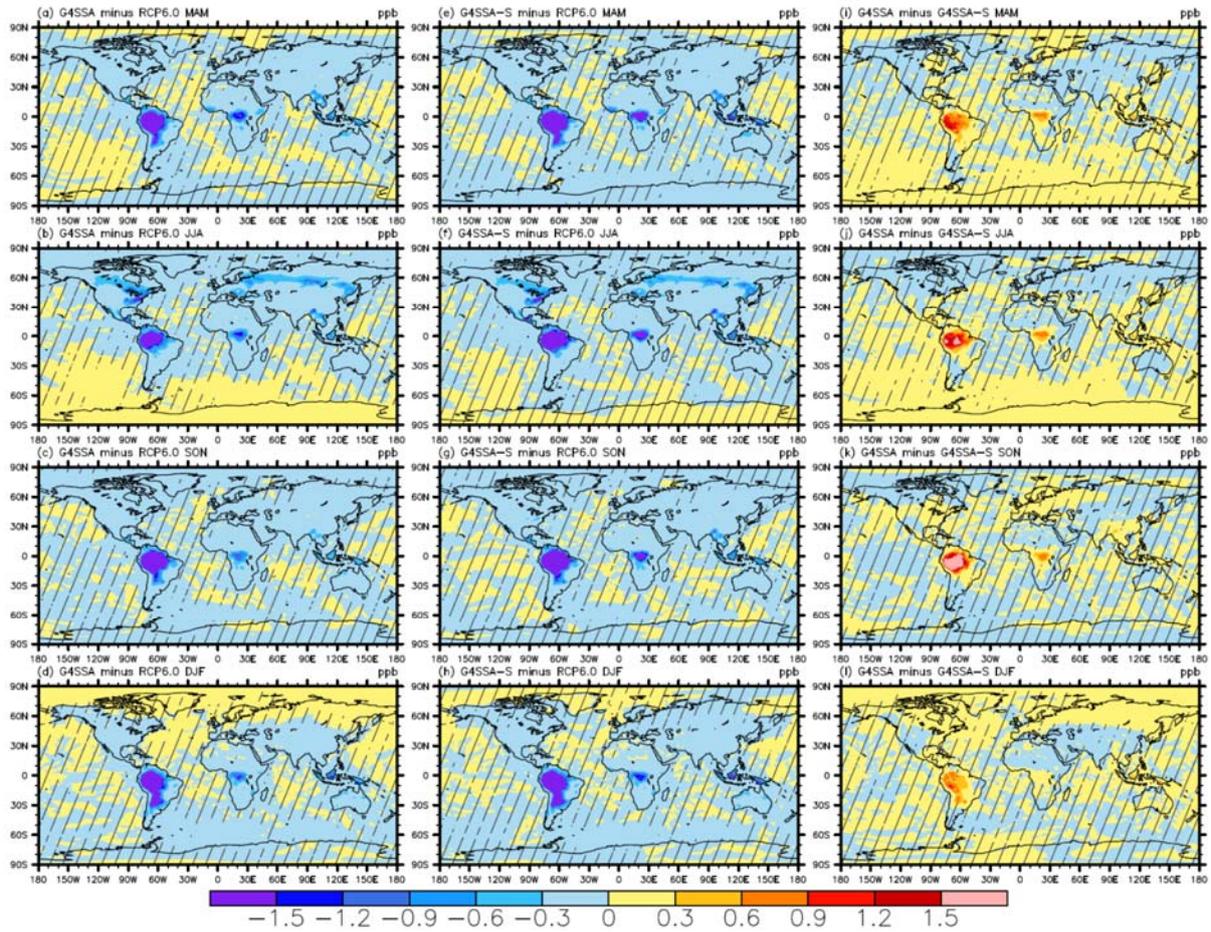


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100 **Figure S10.** Global map of surface NO_x concentration (ppb) in (a) RCP6.0, (b) G4SSA and (c)
 101 G4SSA-S for 2030-2069.

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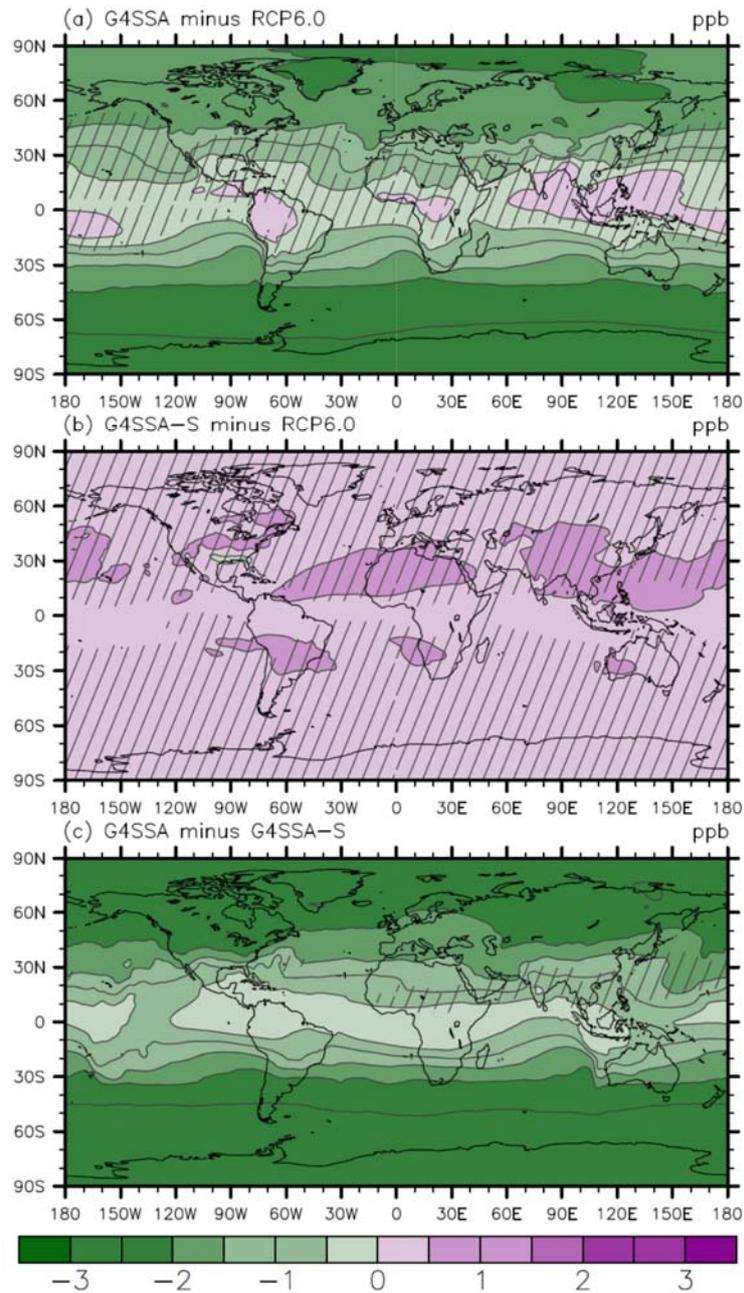


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105 **Figure S11.** Global map of seasonal surface bio-emitted isoprene concentration differences
106 (ppb) between G4SSA and RCP6.0 (left column), G4SSA-S and RCP6.0 (middle column) and
107 G4SSA and G4SSA-S (right column) for 2030-2069. Hatched regions are areas with $p > 0.05$.

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111 **Figure S12.** Global map of surface O_3^{Strat} differences (ppb) between (a), G4SSA and RCP6.0,
 112 (b) G4SSA-S and RCP6.0, and (c) G4SSA and G4SSA-S for 2030-2069. Hatched regions are
 113 areas with $p > 0.05$. There is much less O_3^{Strat} at the surface in G4SSA relative to RCP6.0 as
 114 well as G4SSA-S. Changes in O_3^{Strat} at the surface are on the one hand due to reduced ozone in
 115 the stratosphere, and on the other hand due to changes in the rate of STE. Although the
 116 absolute value of O_3^{Strat} is overestimated, because of a missing loss process via dry deposition in
 117 the version of the model, it can be qualitatively used to compare the two scenarios, since dry
 118 deposition is not expected to change significantly.

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