Supplementary material:

Tropospheric ozone changes, radiative forcing and attribution to emissions in the Atmospheric Chemistry and Climate Model Inter-comparison Project (ACCMIP)

D.S. Stevenson[1], P.J. Young[2,3,*], V. Naik[4], J.-F. Lamarque[5], D.T. Shindell[6], A. Voulgarakis[7], R.B. Skeie[8], S.B. Dalsoren[8], G. Myhre[8], T.K. Berntsen[8], G.A. Folberth[9], S.T. Rumbold[9], W.J. Collins[9,], I.A. MacKenzie[1], R.M. Doherty[1], G. Zeng[10], T.P.C. van Noije[11], A. Strunk[11], D. Bergmann[12], P. Cameron-Smith[12], D.A. Plummer[13], S.A. Strode[14], L. Horowitz[15], Y.H. Lee[6], S. Szopa[16], K. Sudo[17], T. Nagashima[18], B. Josse[19], I. Cionni[20], M. Righi[21], V. Eyring[21], A. Conley[5], K.W. Bowman[22], O. Wild[23], A. Archibald[24]**

This supplementary material comprises:

Figure S1 shows equivalent results to Figure 1 of the main text, but for all models.

Figure S2 shows equivalent results to Figure 3 of the main text, but for all models.

Figure S3 shows equivalent results to Figure 3a-c, but for the IGAC/SPARC ozone dataset.

Figure S4 shows attribution experiment ozone results for Model B.

Figure S5 shows attribution experiment radiative forcing results for Model B.

Figure S6 shows radiative forcing results for all available models and timeslice experiments.

Figure S7 shows equivalent results to Figure 5 of the main text, but for ten models.

A section describing the changes in CO_2 due to emissions of CH_4, CO and NMVOCs.

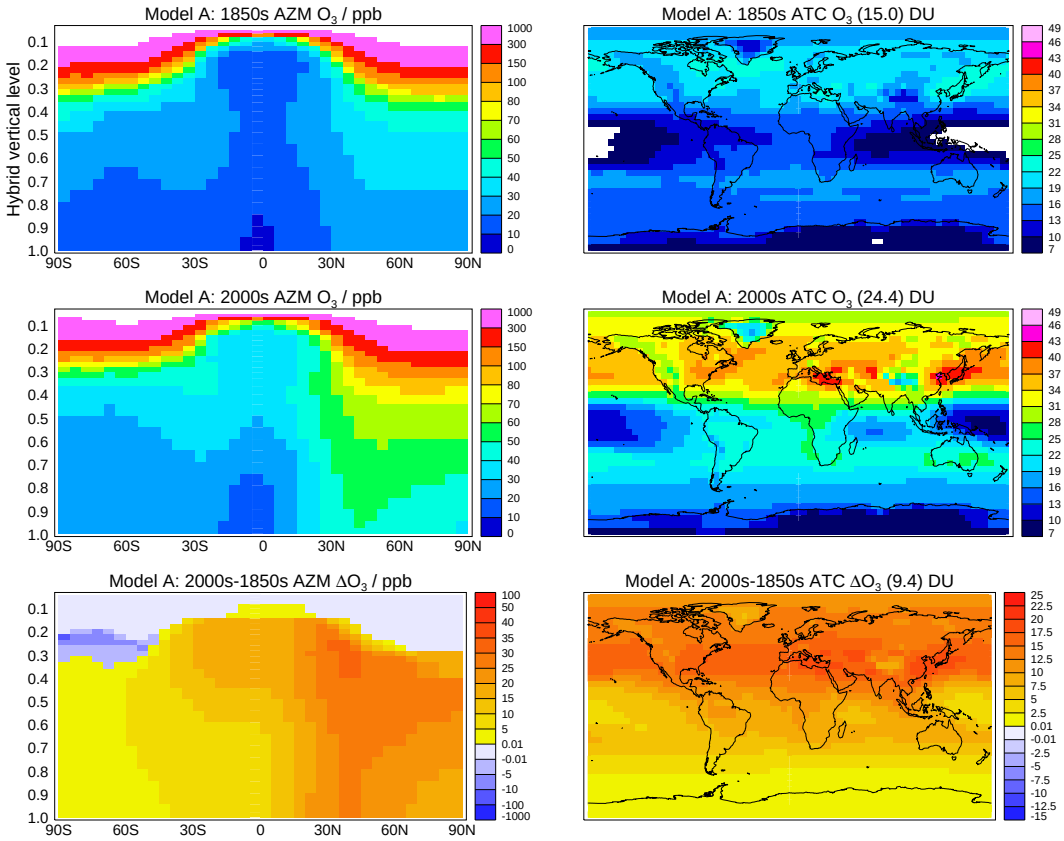
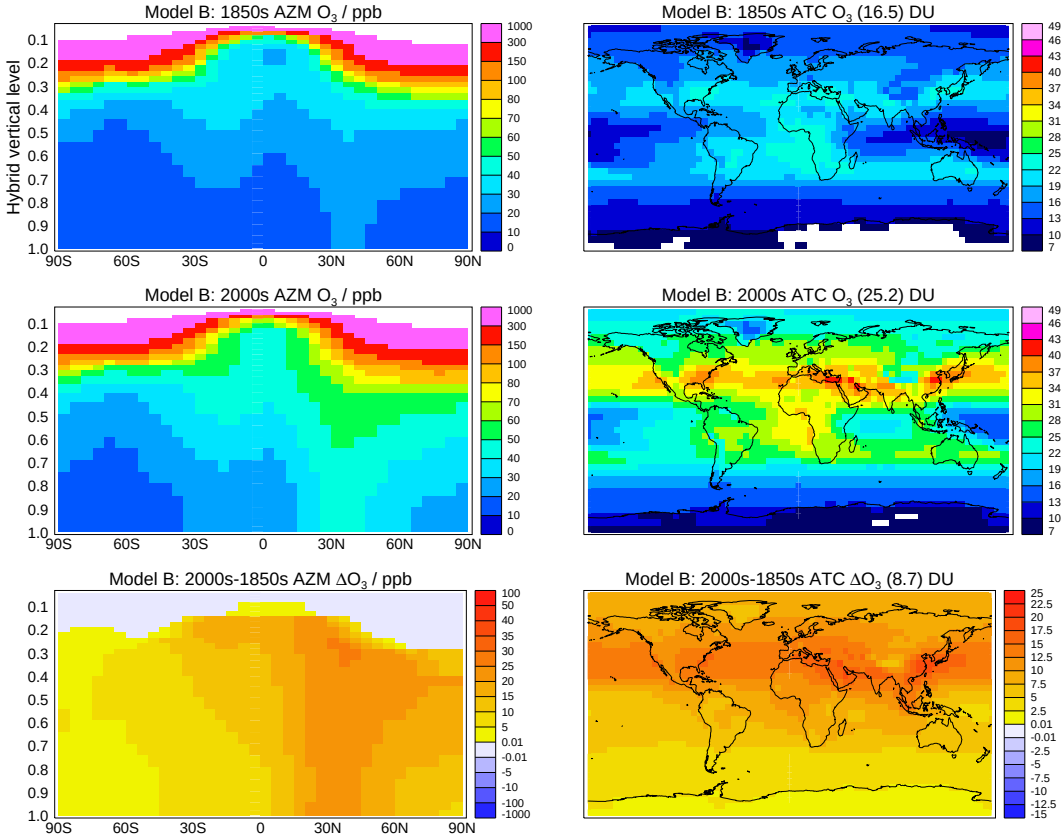


Figure S1: Same as Figure 1 from the main text, but for all individual models.



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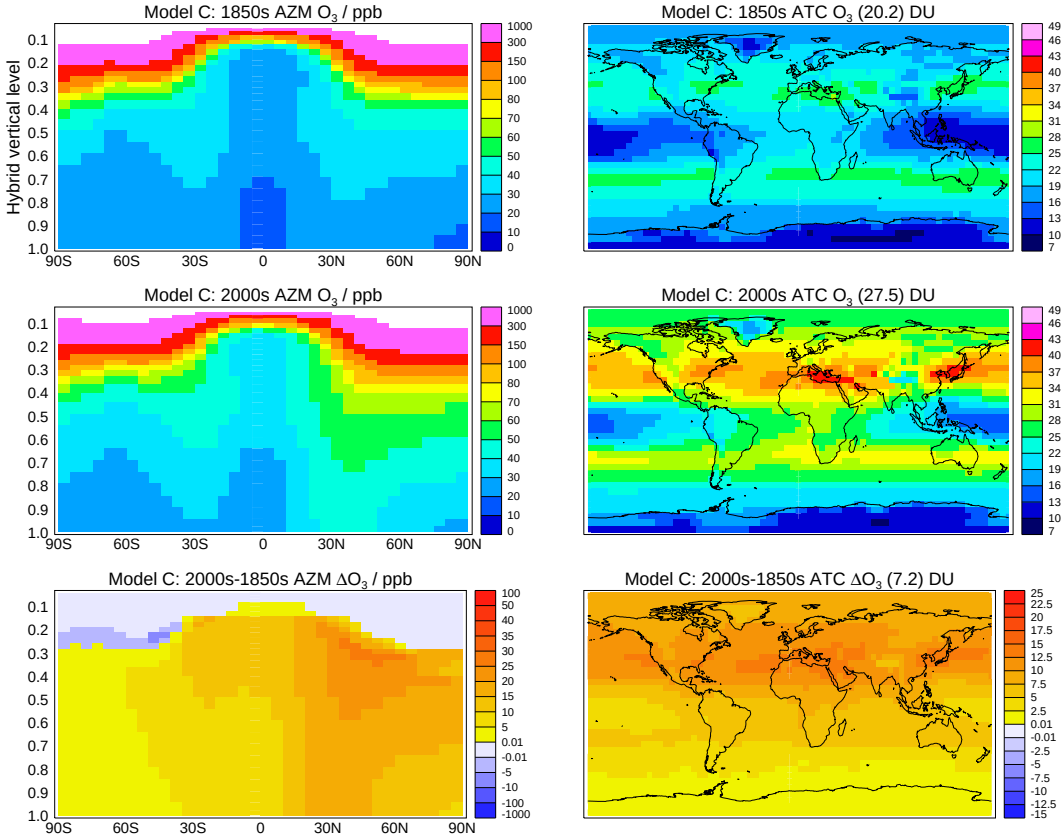

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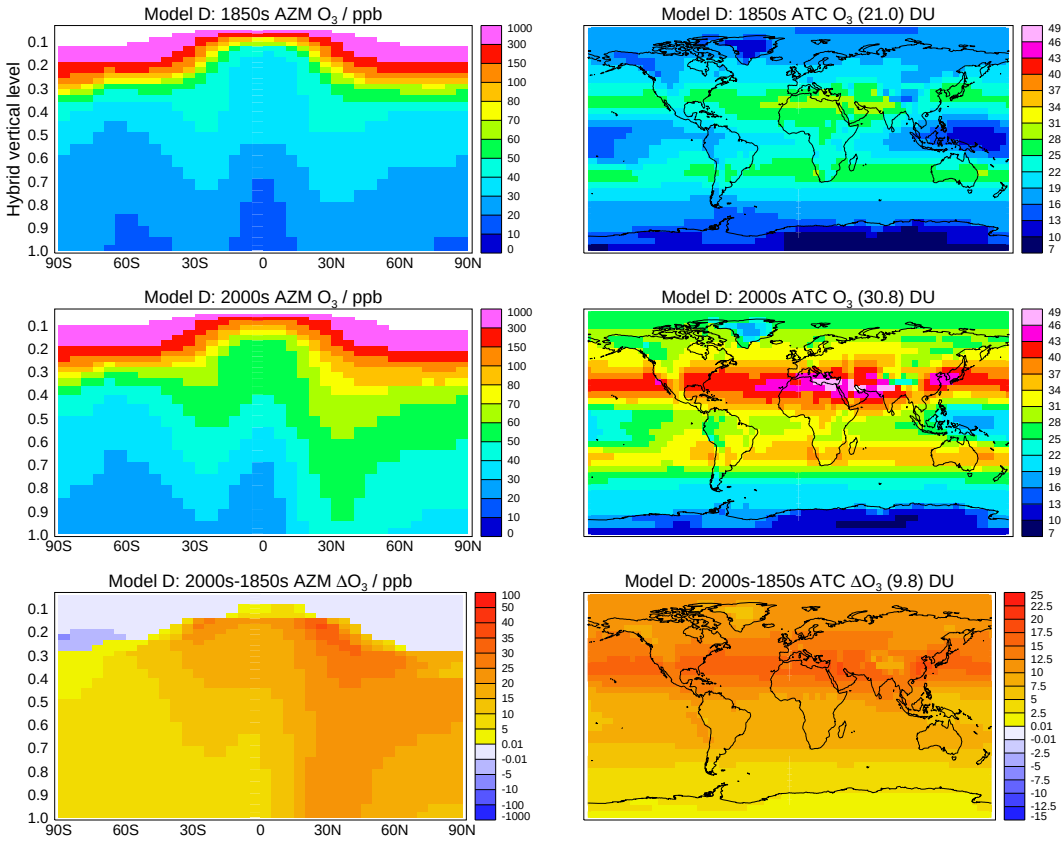

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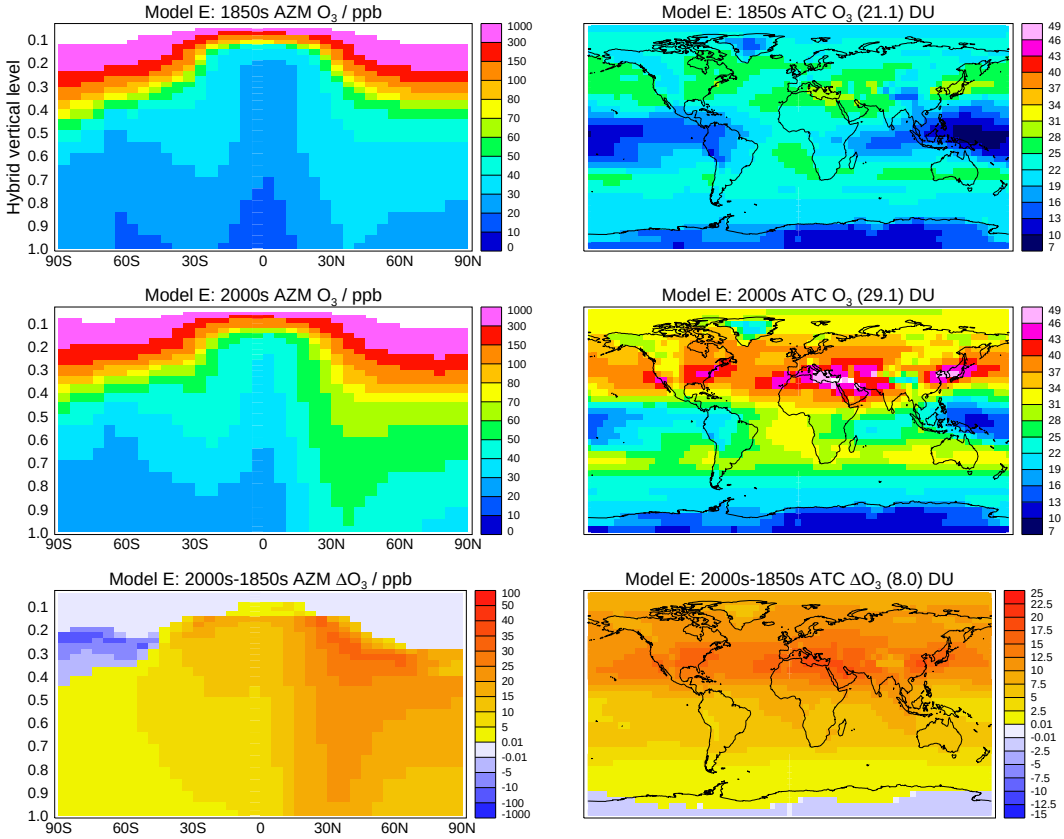

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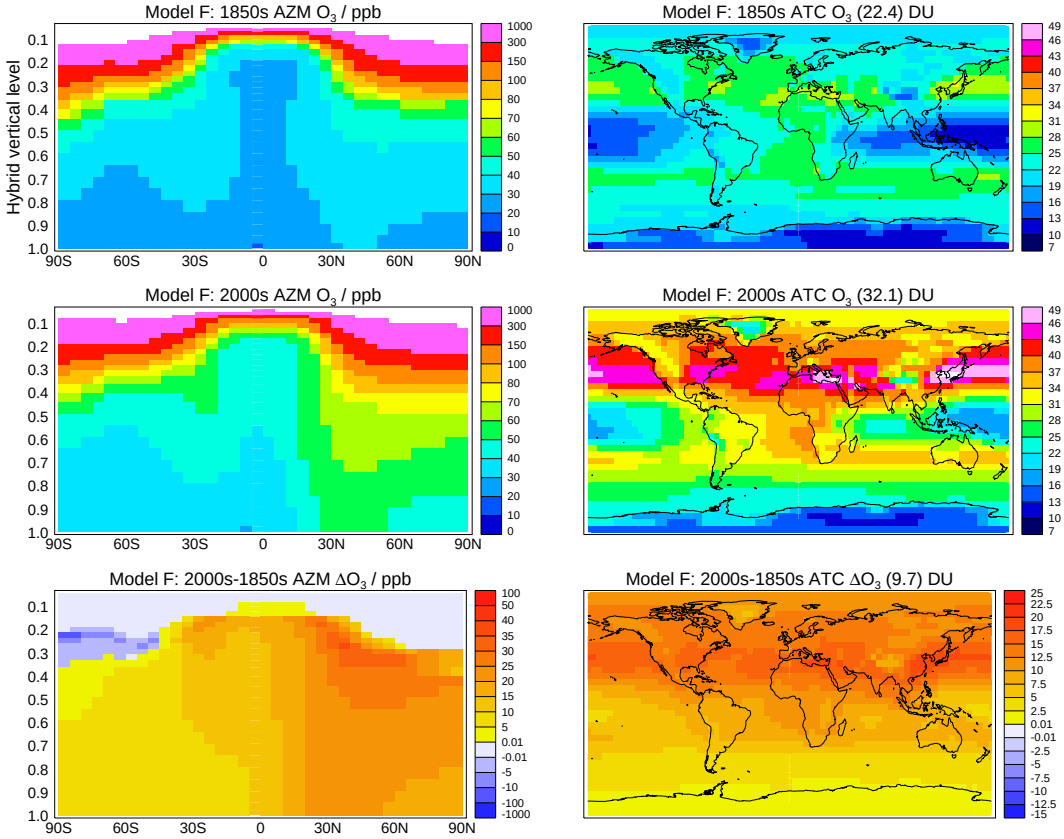

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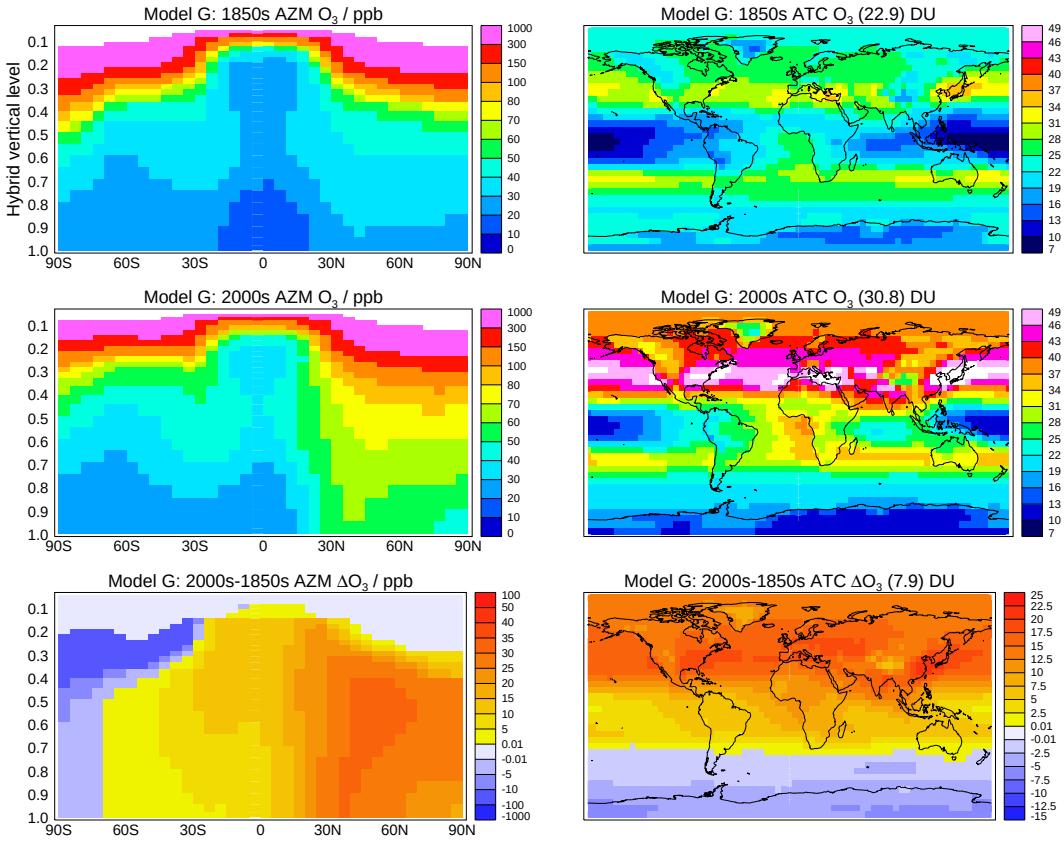

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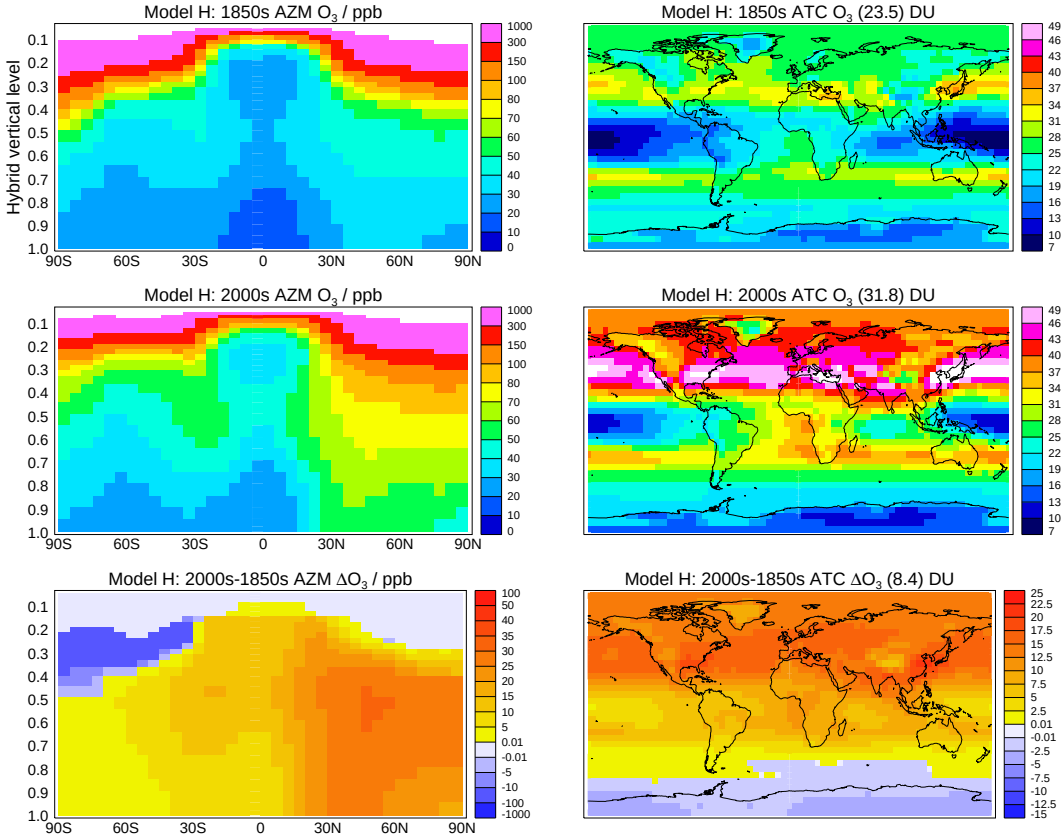

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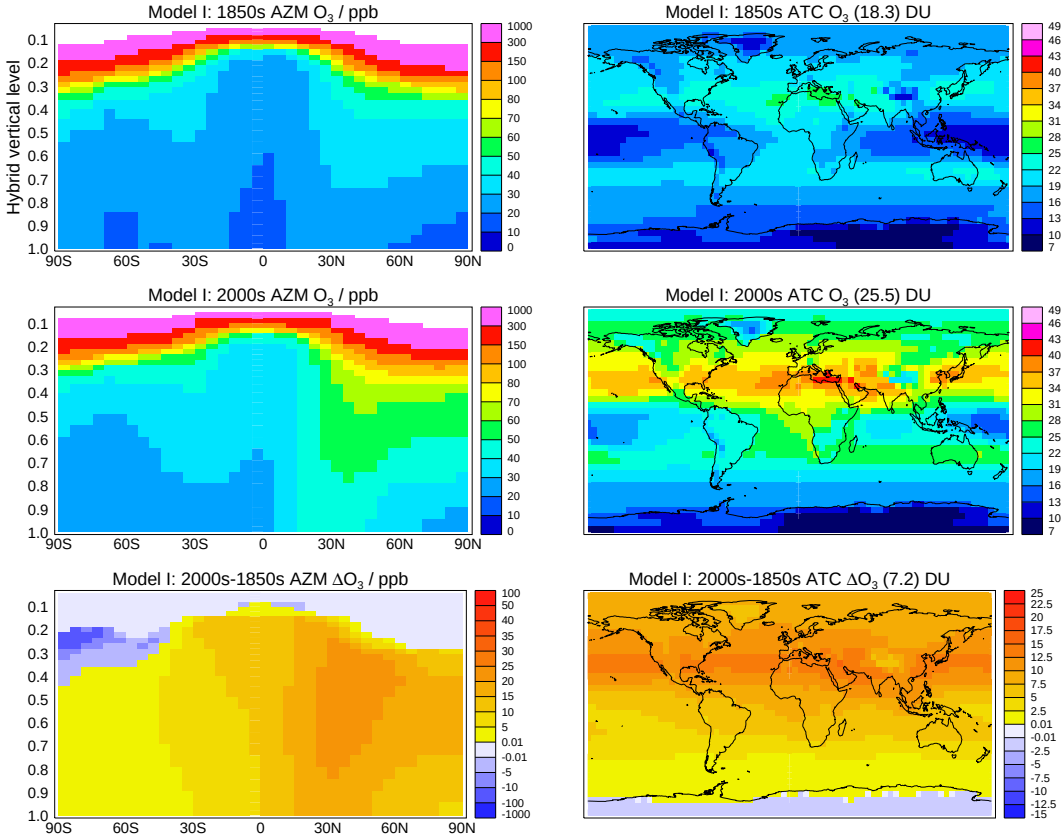

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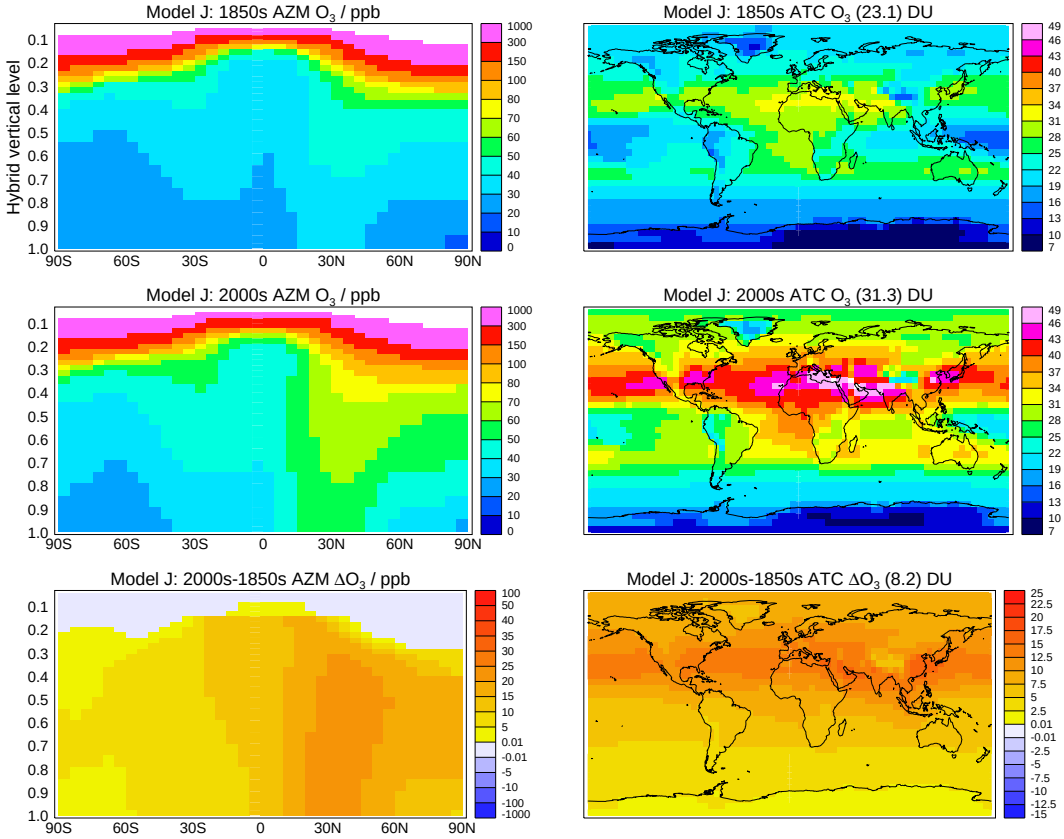

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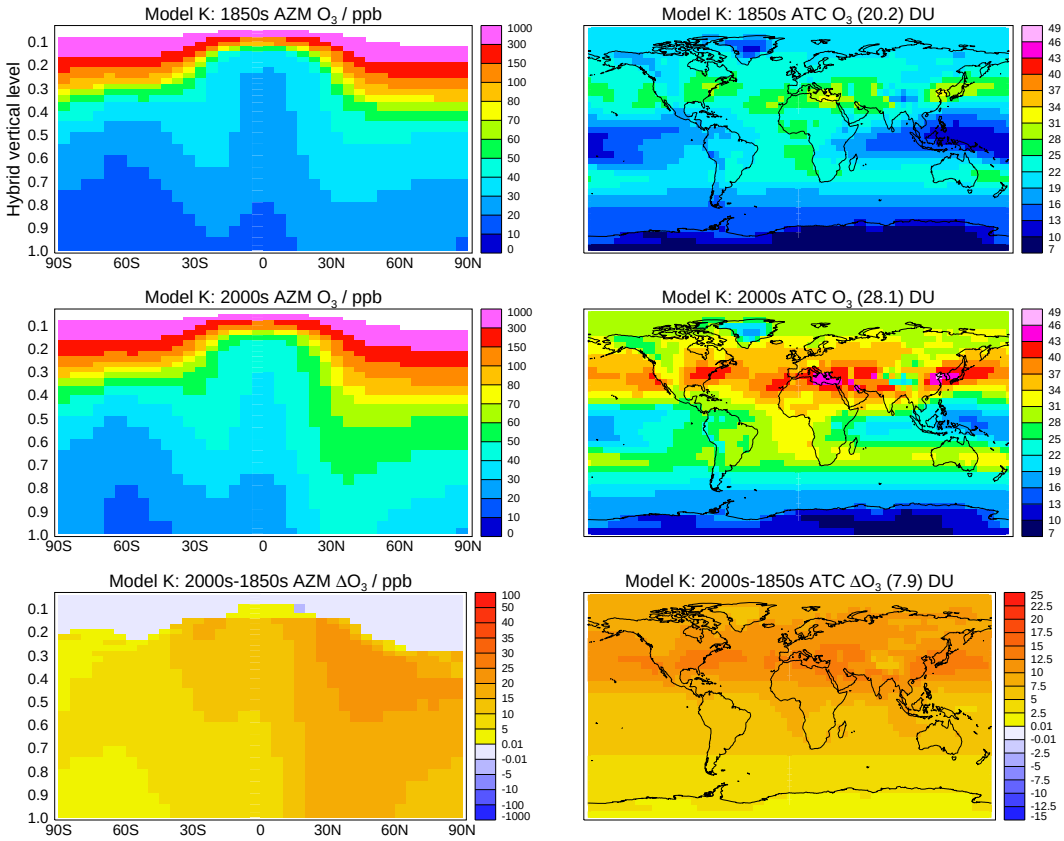

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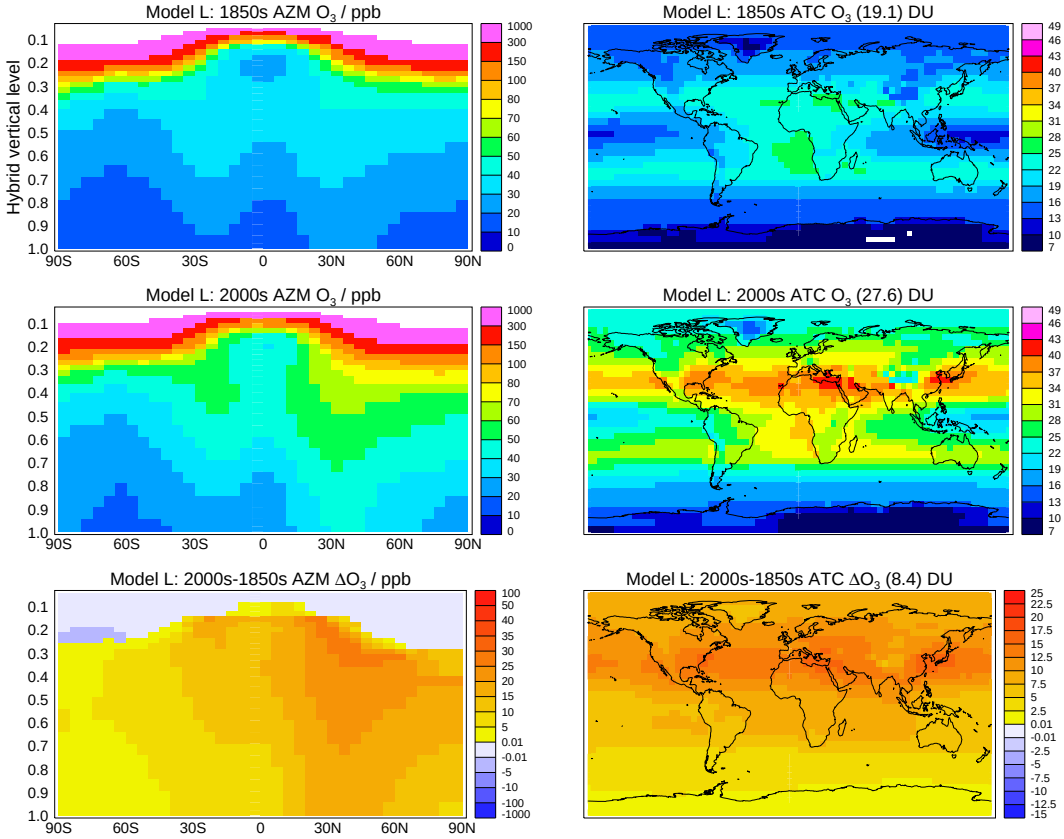

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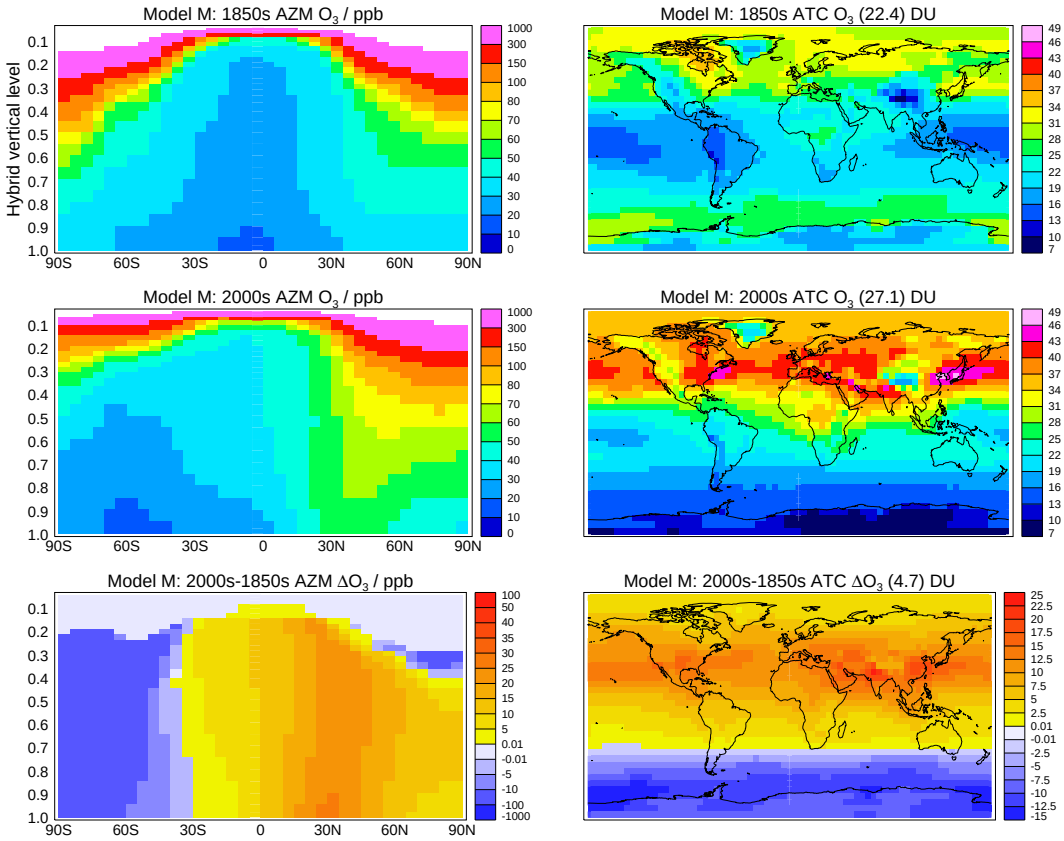

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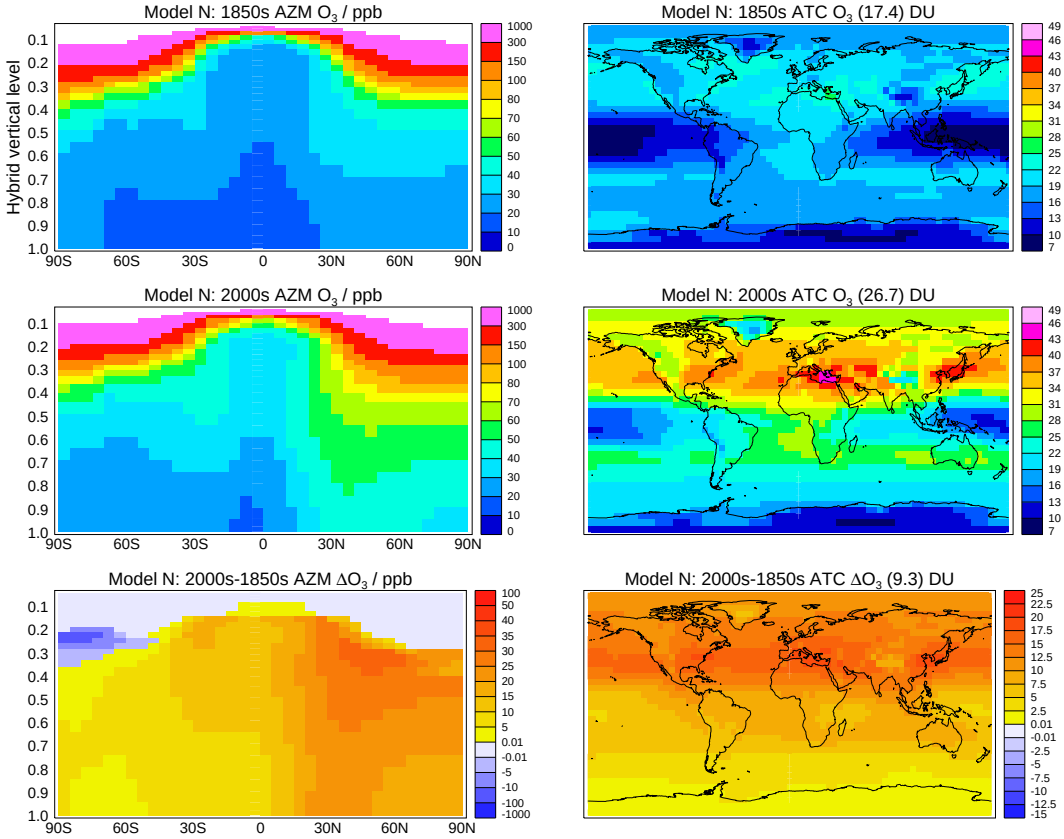

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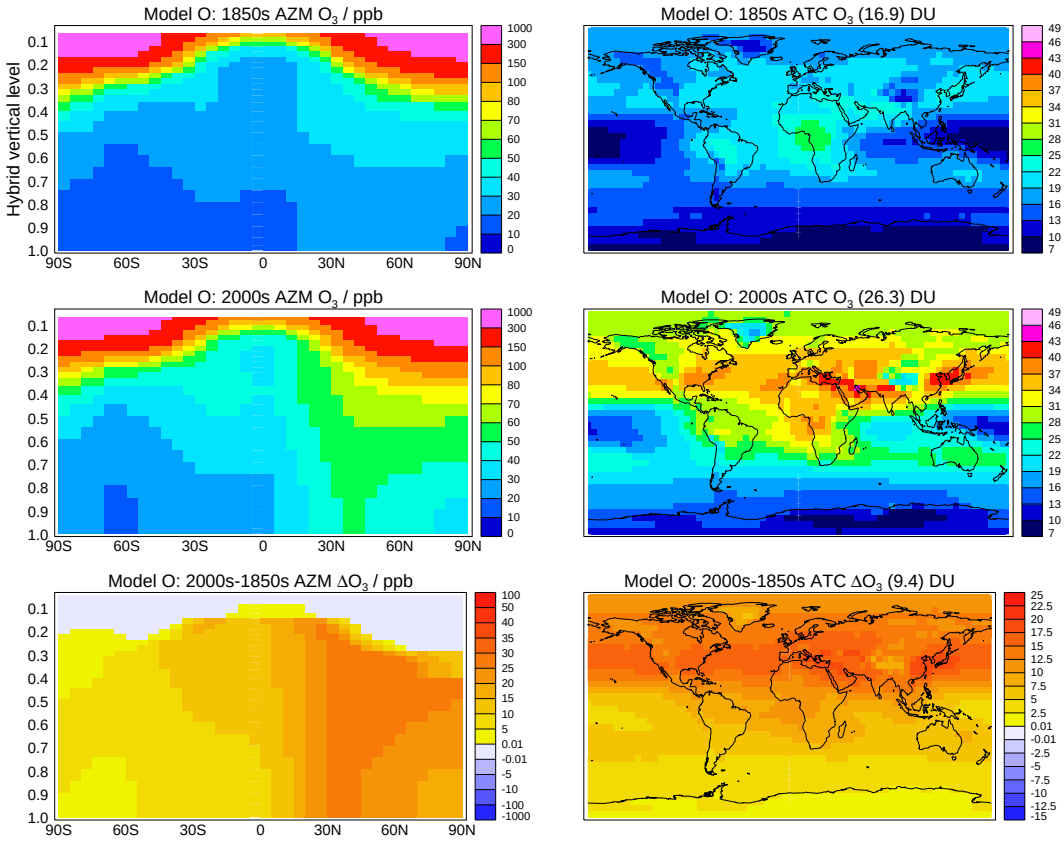

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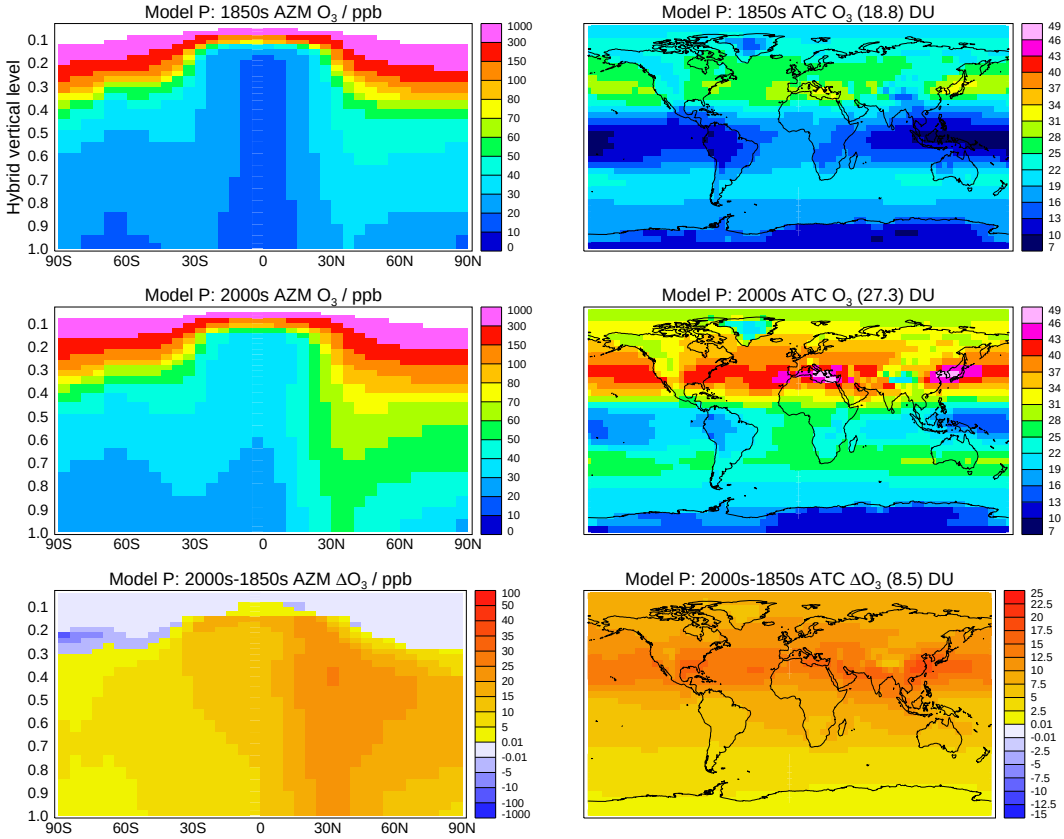

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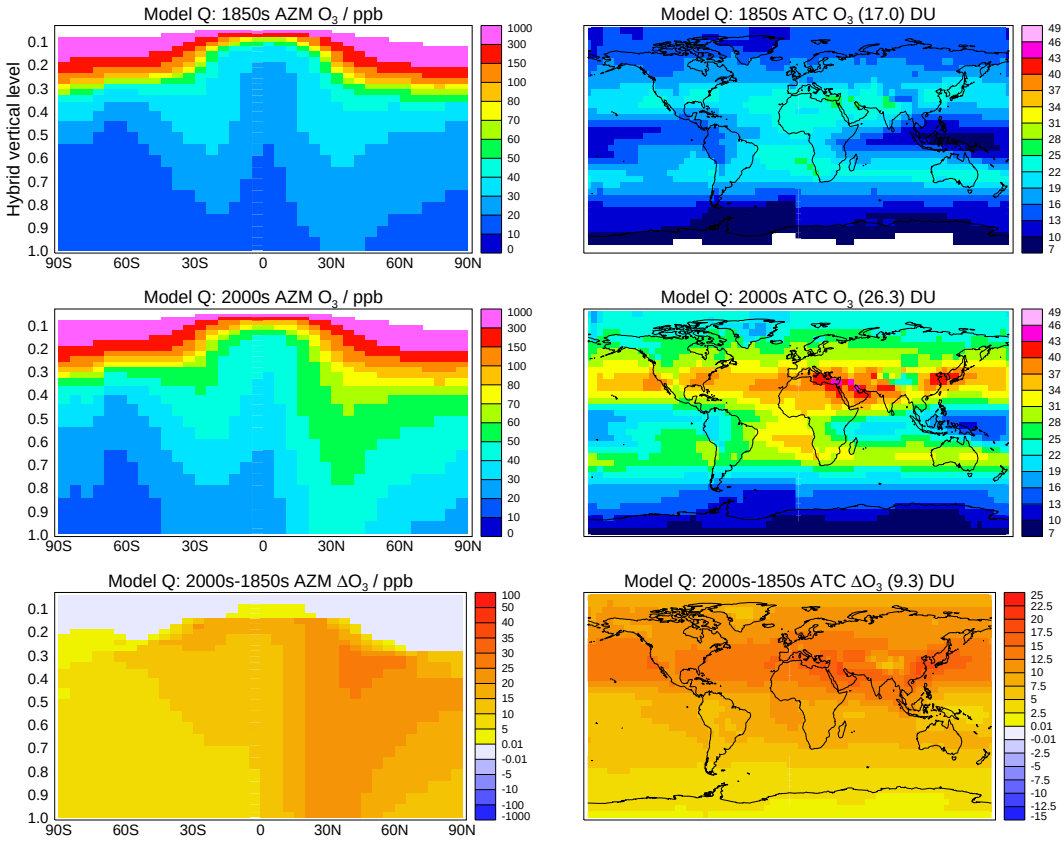

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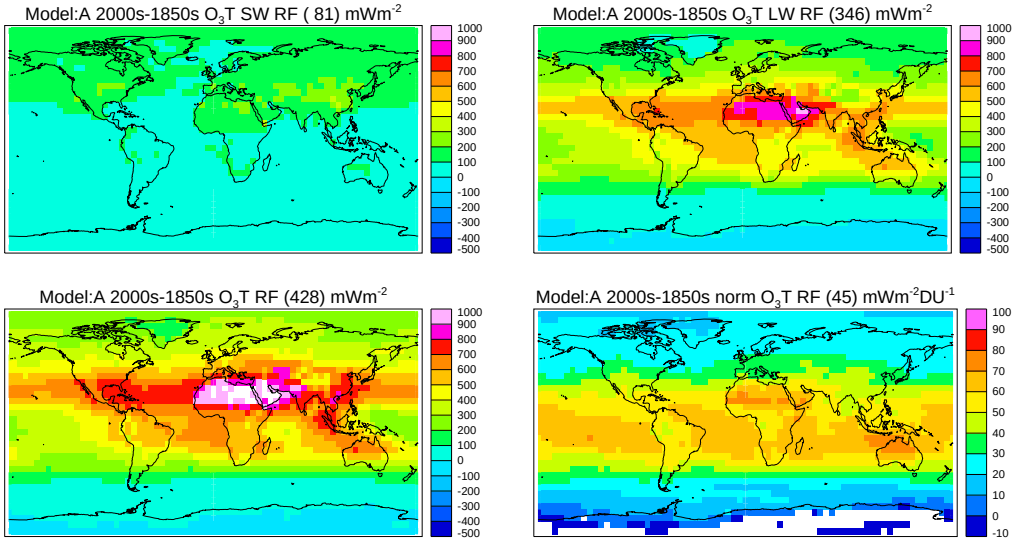

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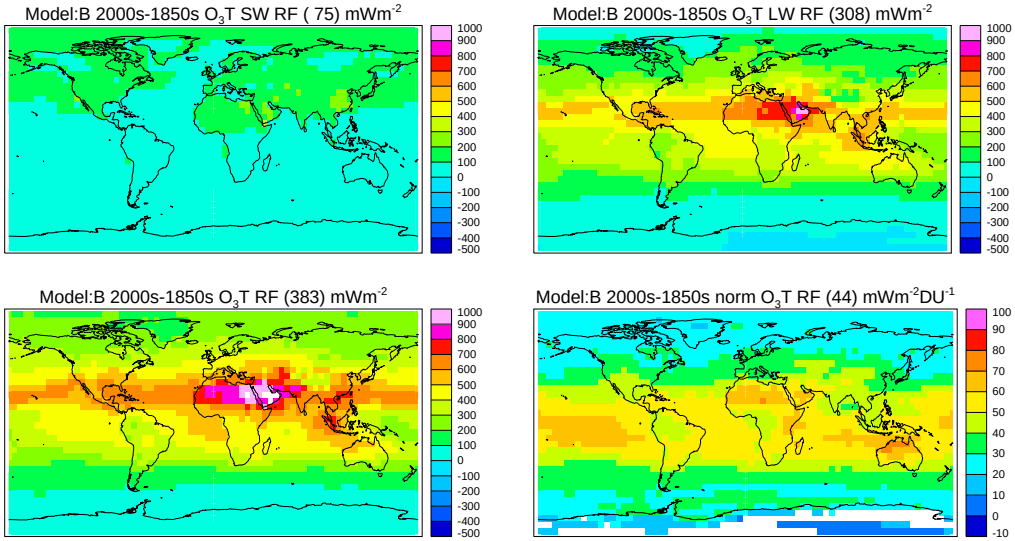

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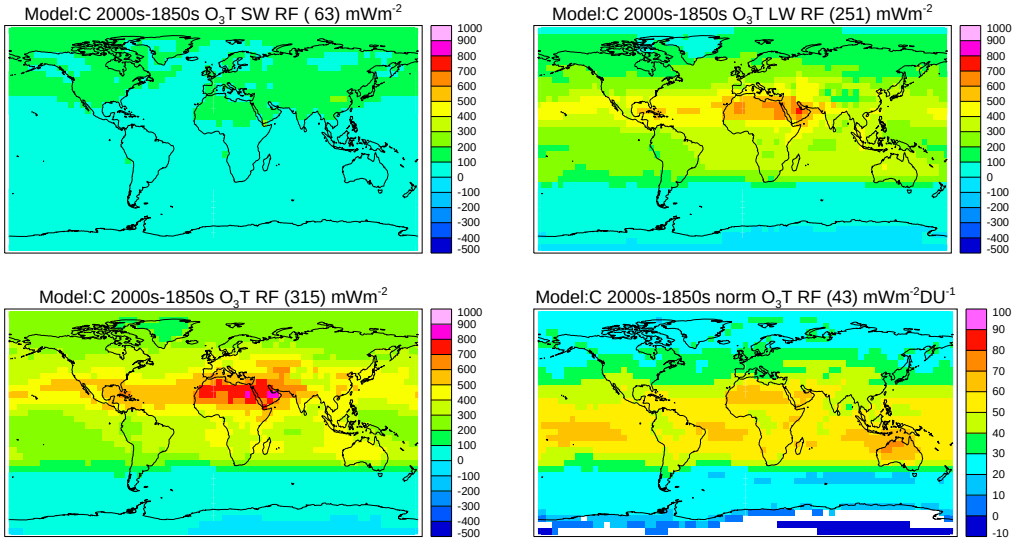

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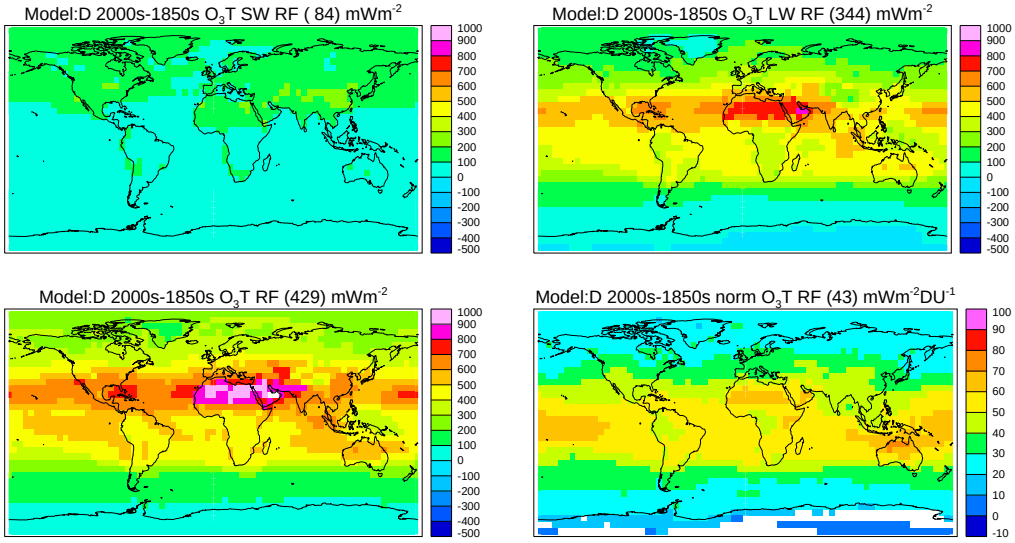

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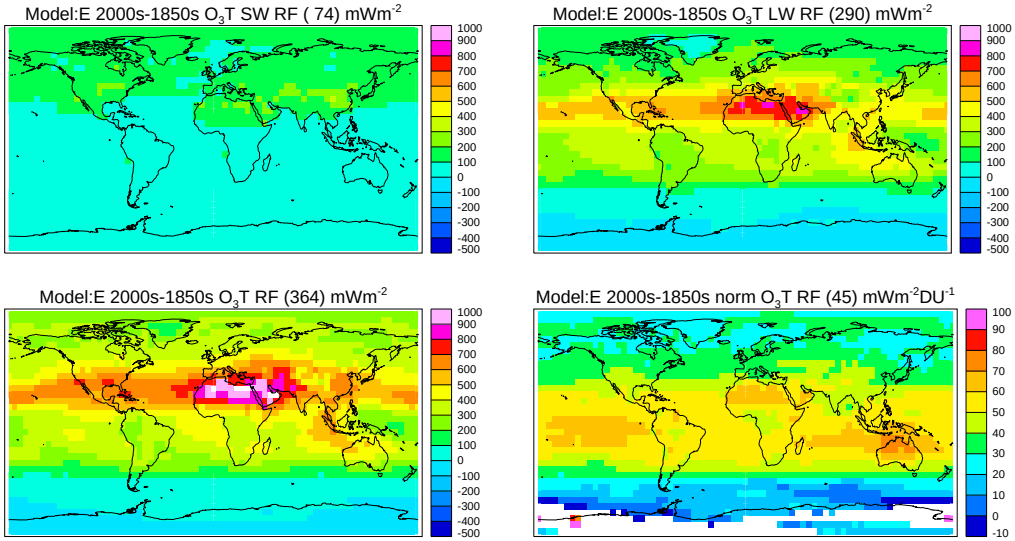

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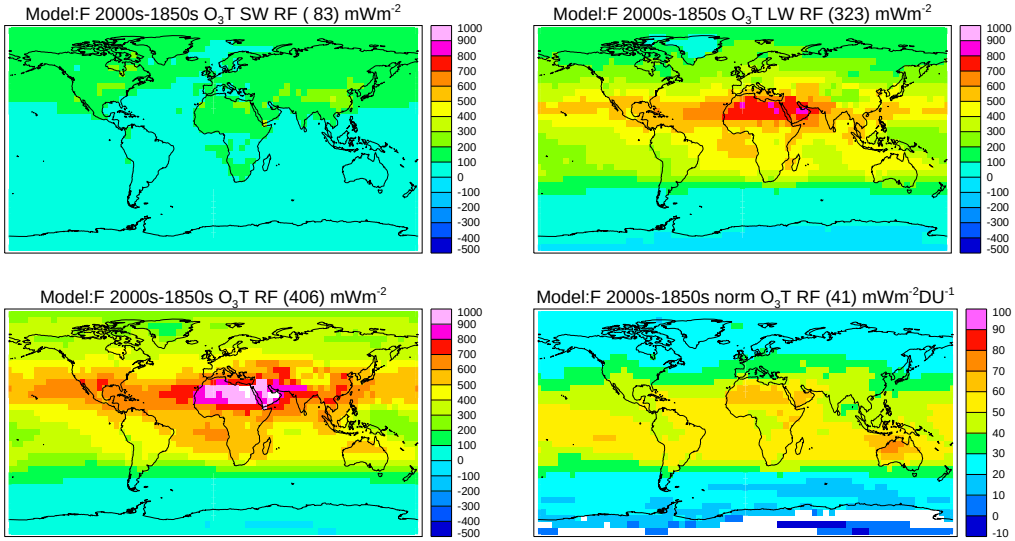

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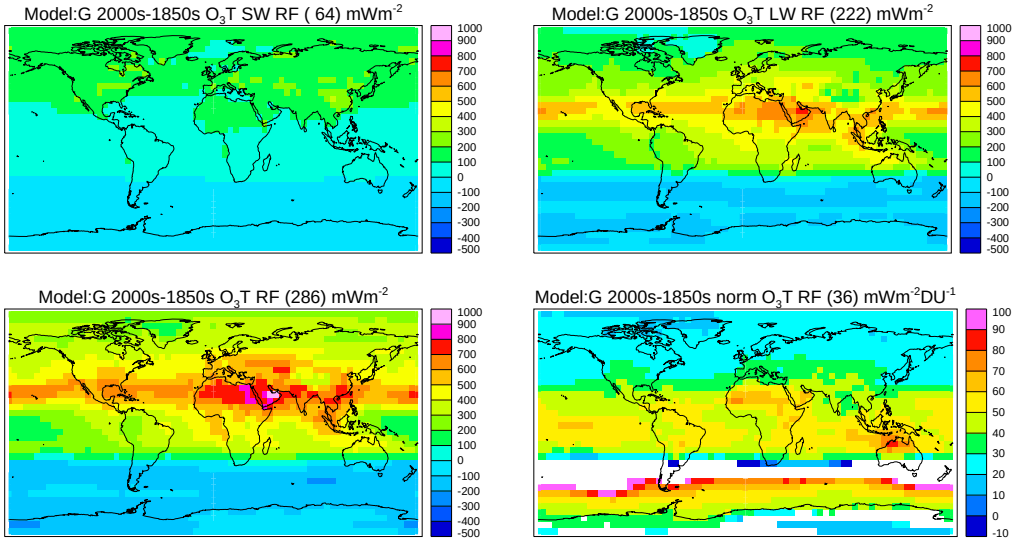

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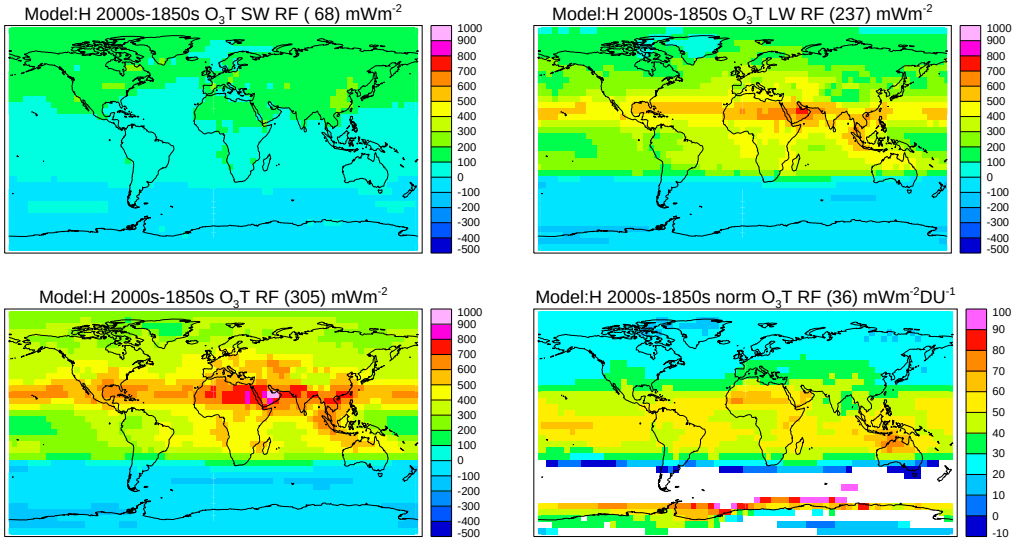

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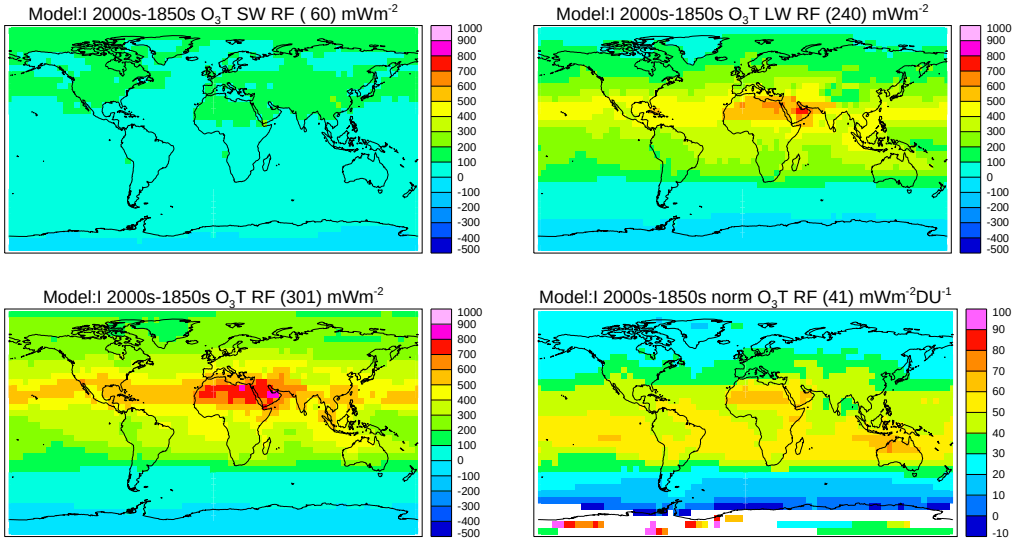

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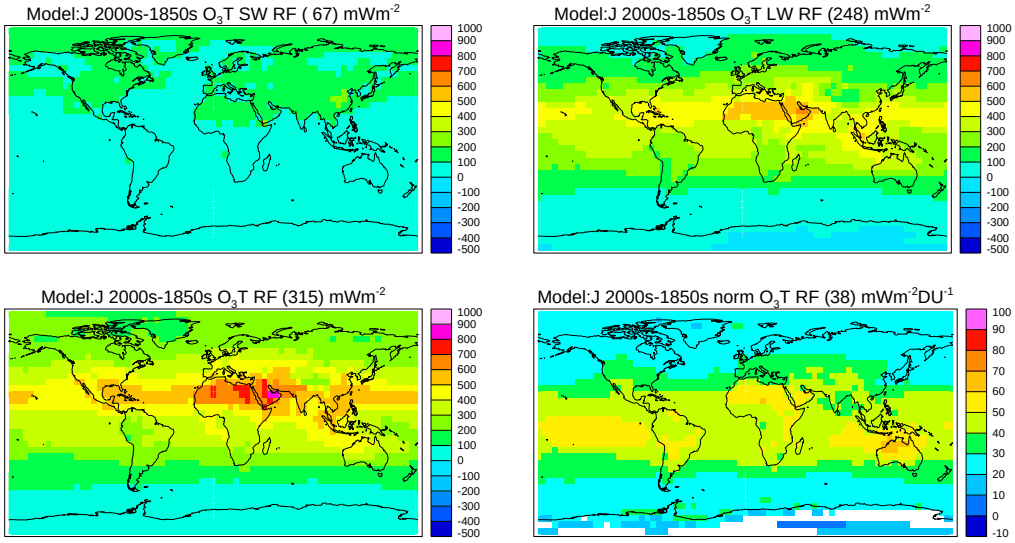

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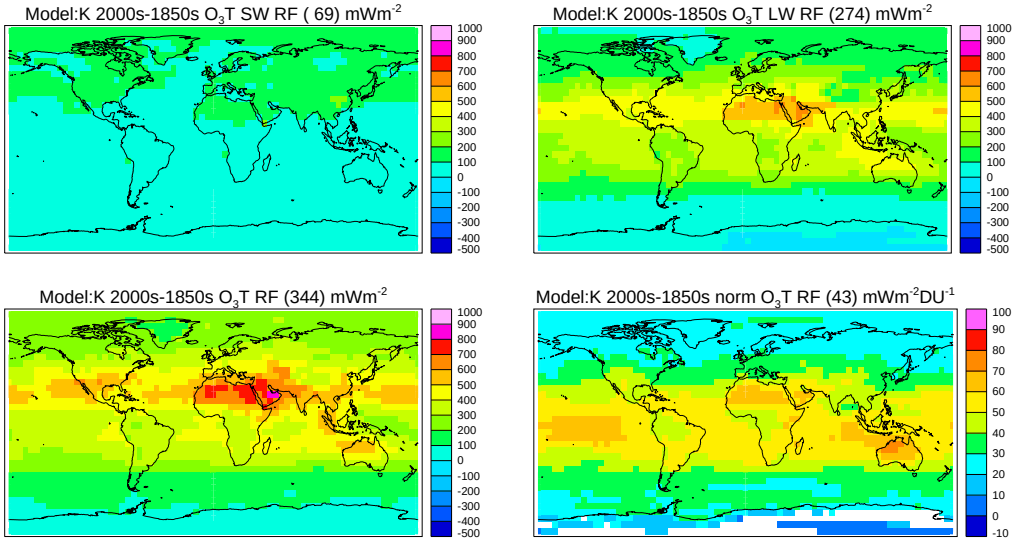

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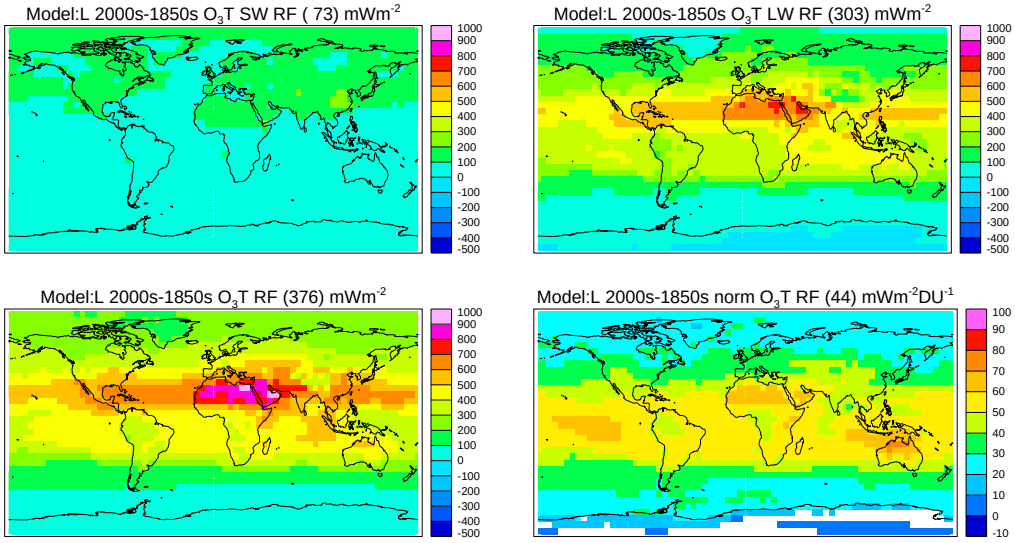

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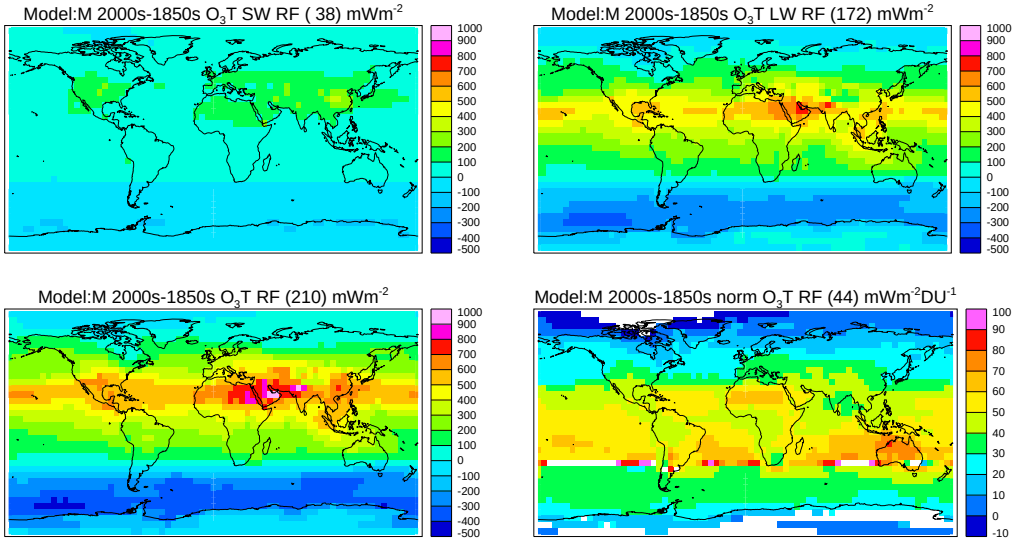

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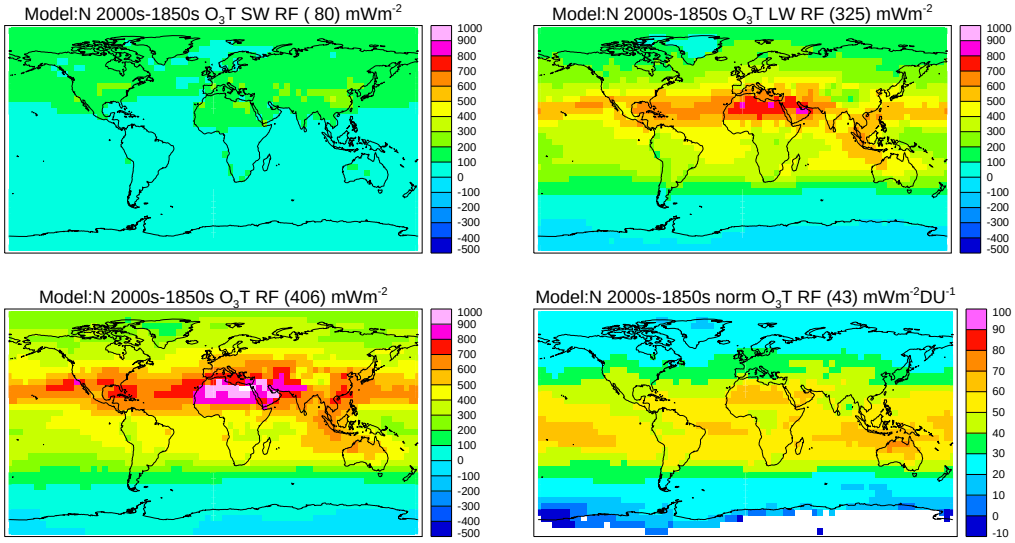

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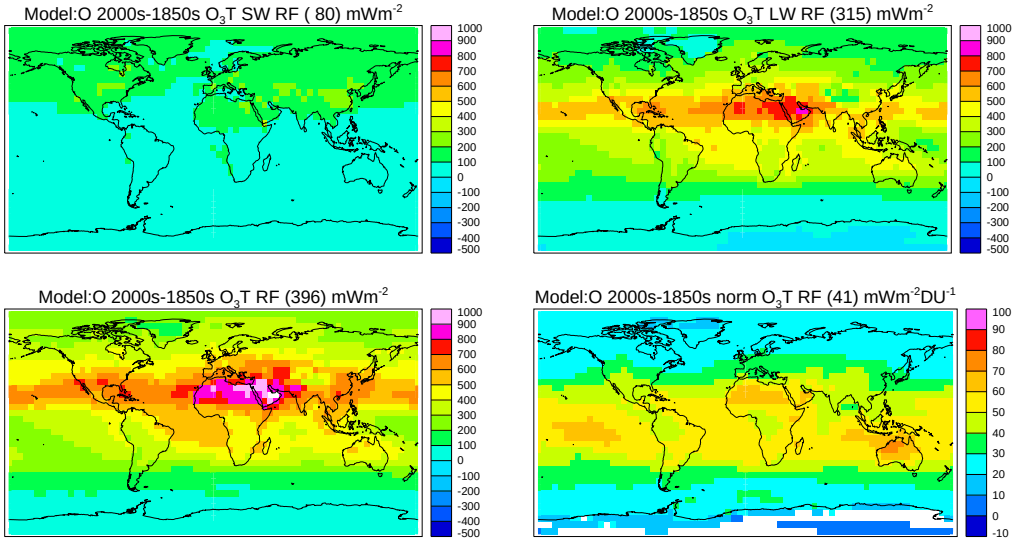

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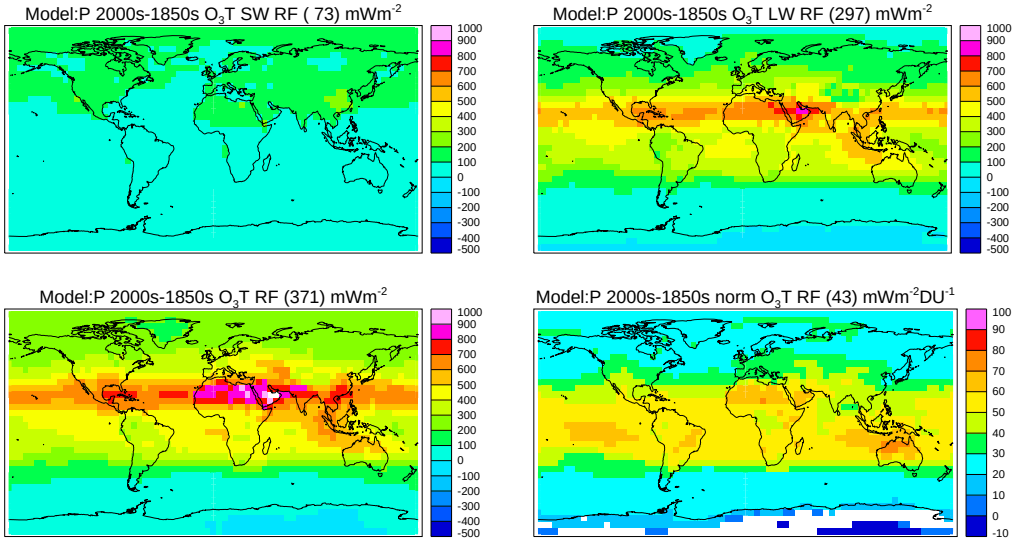

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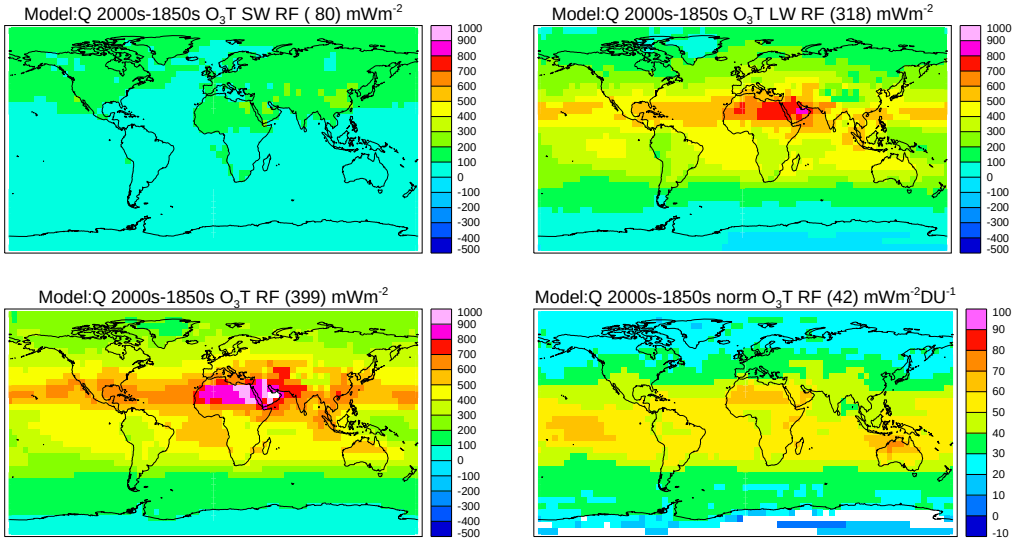

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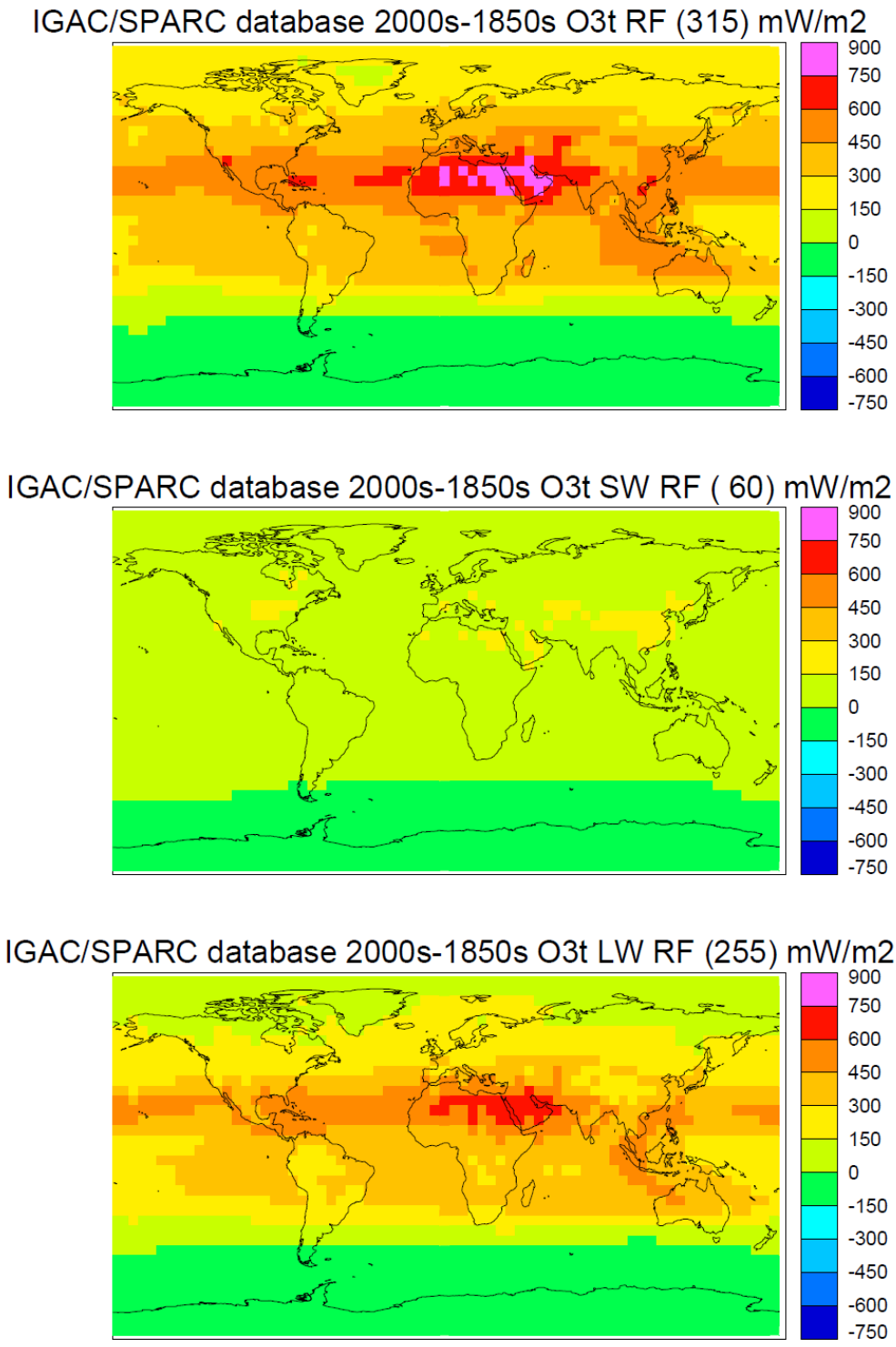


Figure S3: Annual mean tropospheric O_3 total, SW, and LW RFs for the IGAC/SPARC ozone dataset, as used in Cionni et al (2011). Compare to multi-model mean in Figure 3; also compare to Cionni et al.'s Figure 15a, which shows the total RF calculated with an earlier version of the E-S radiation code, and finds a total RF 27% lower.

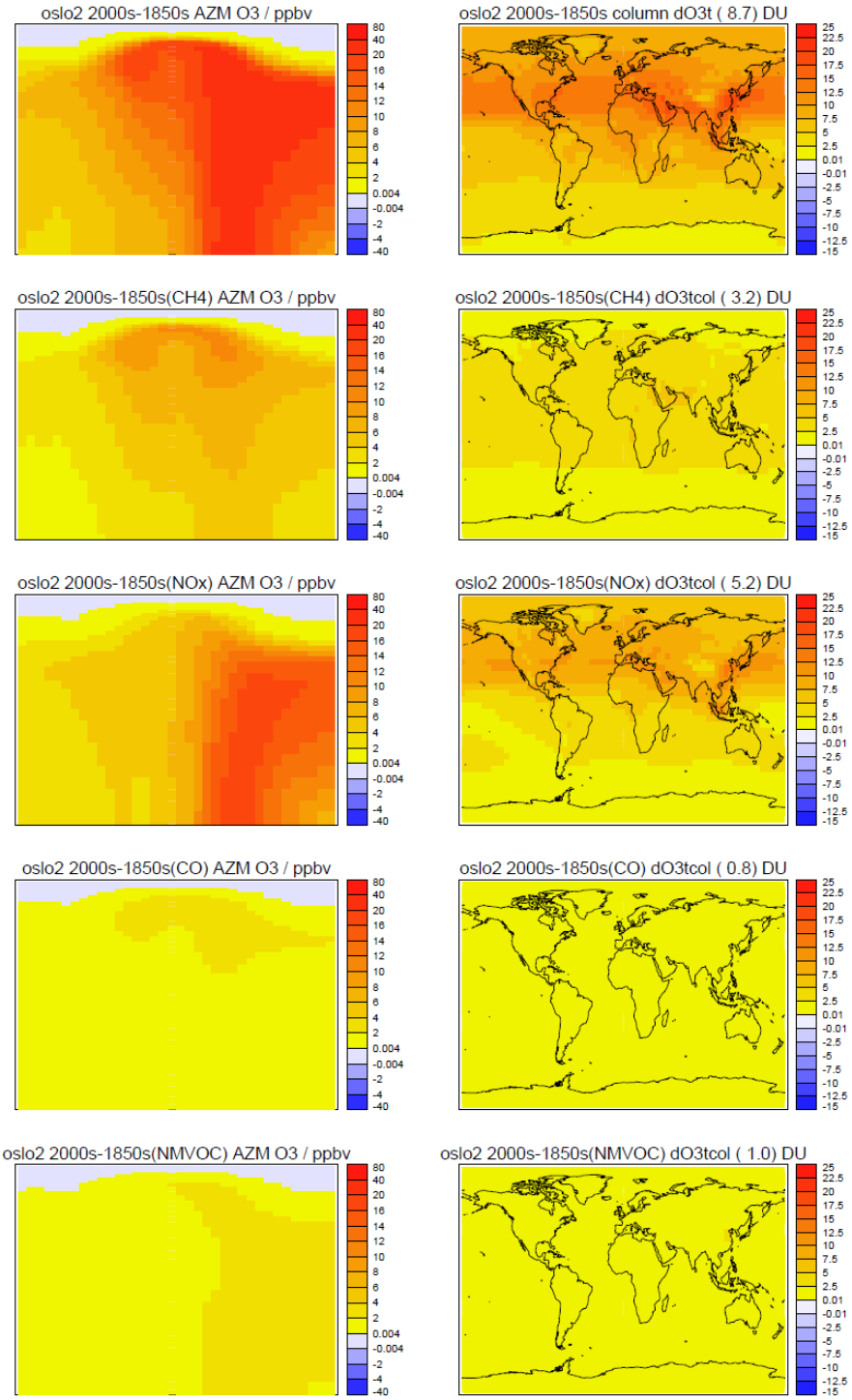


Figure S4: An example of one model's results (model B) for the attribution experiments. Left hand side shows contributions to changes in zonal annual mean ozone, right hand side shows contributions to change in annual mean tropospheric ozone column. Referring to the experiment numbers in Table 6, rows from the top show: experiment #1-#0 (all components); #1-#2 (CH_4); #1-#3 (NOx); #1-#4 (CO); #1-#5 (NMVOC).

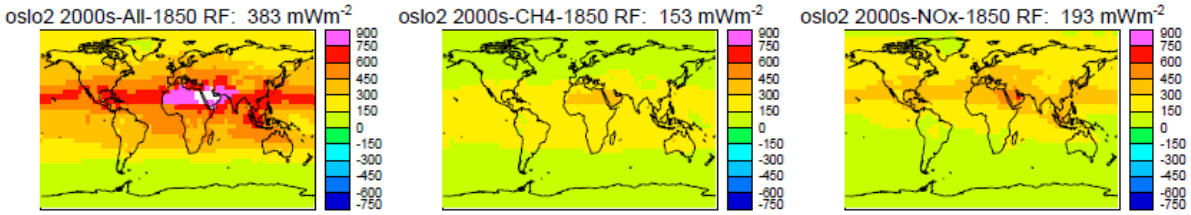


Figure S5: Radiative forcings for model B, from the attribution experiments. Left-hand plot shows total 2000s-1850s (#1-#0); middle shows the CH4 component (#1-#2); and the right-hand plot shows the NOx component (#1-#3). The CO and NMVOC components are significantly less (38 and 37 mWm^{-2}, respectively) and are not shown.

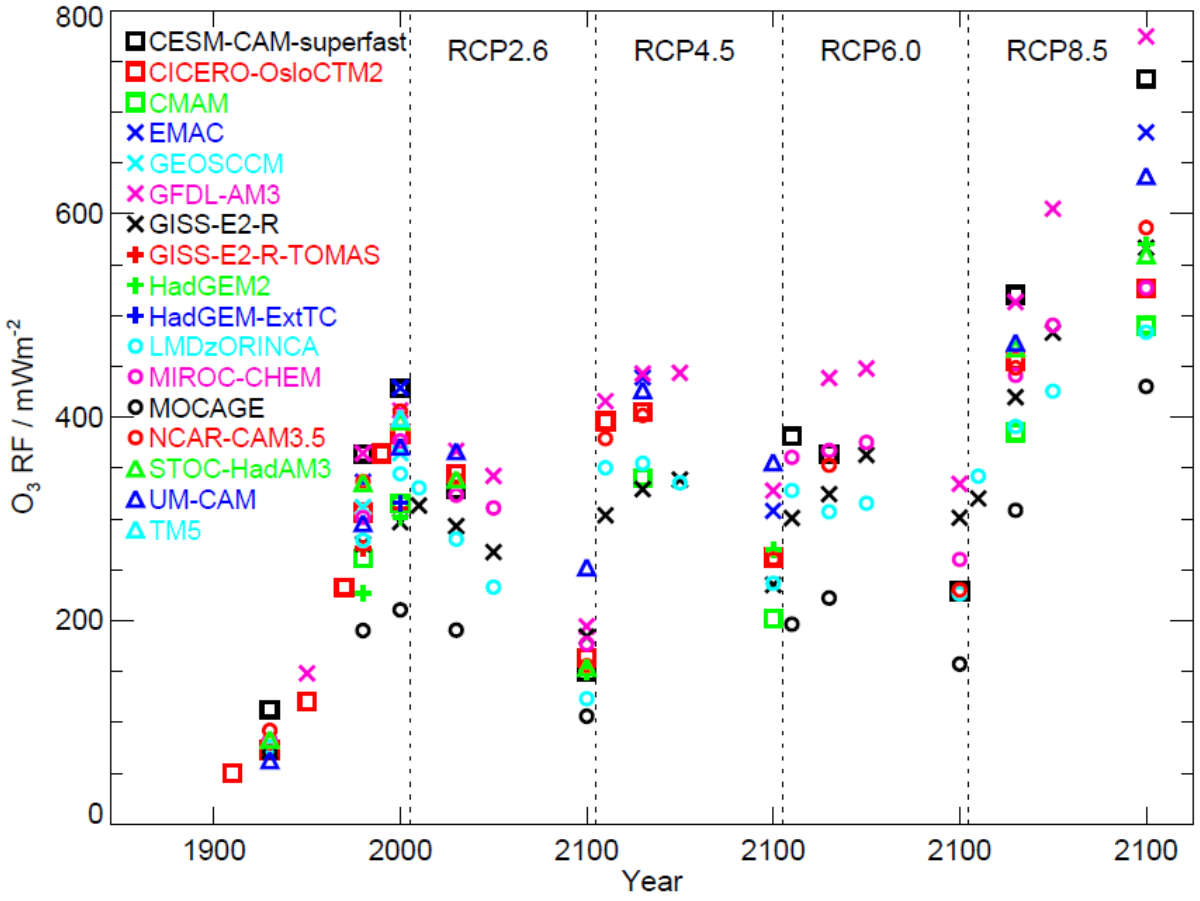


Figure S6: Tropospheric ozone radiative forcing (mW m^{-2}) relative to 1850s, for all available models and timeslice experiments (1910s, 1930s, 1950s, 1970s, 1980s, 1990s, 2000s; then for the four future RCP scenarios 2010s, 2030s, 2050s and 2100s). All models ran the 2000s and 1850s; for other timeslices, varying subsets of models are available. This plot for MASKZMT.

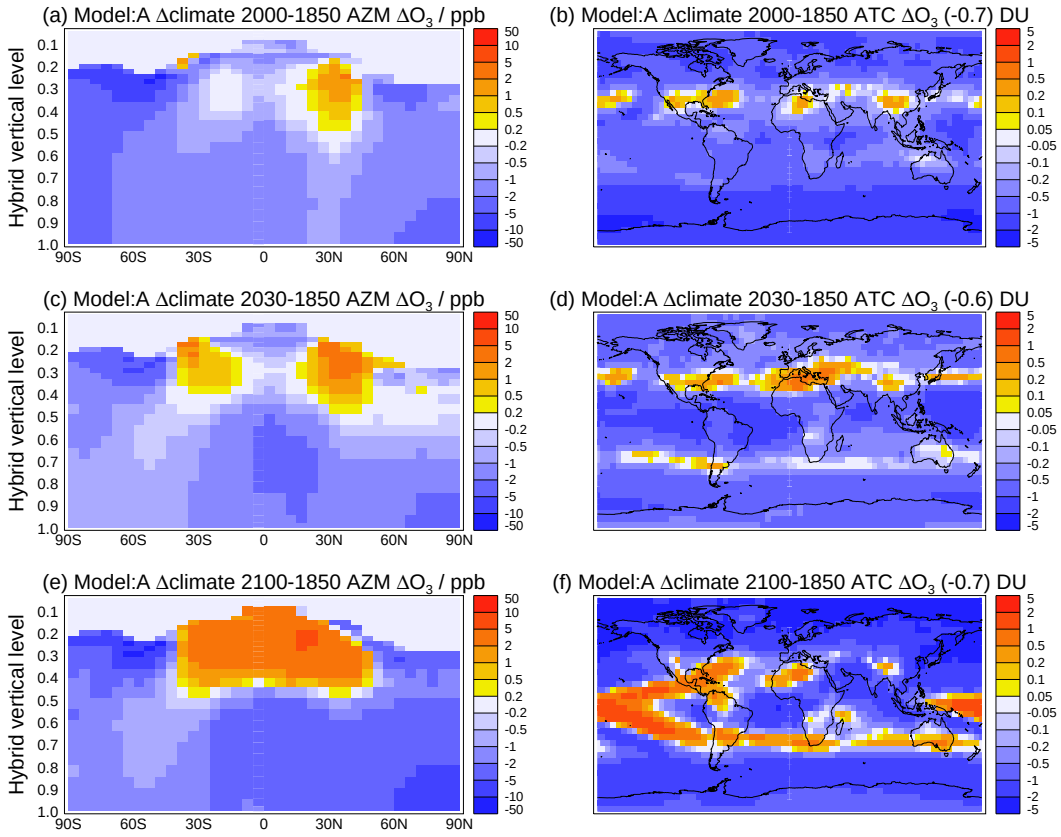
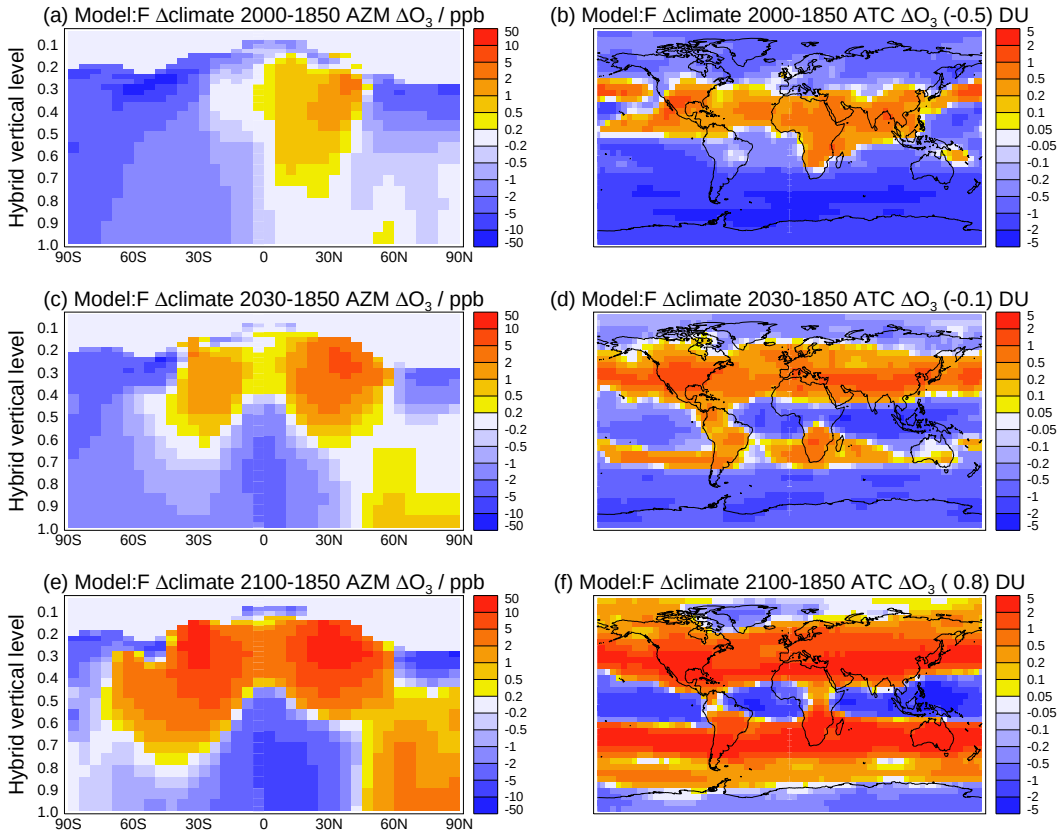


Figure S7: Same as Figure 5 from the main text, but for all individual models.



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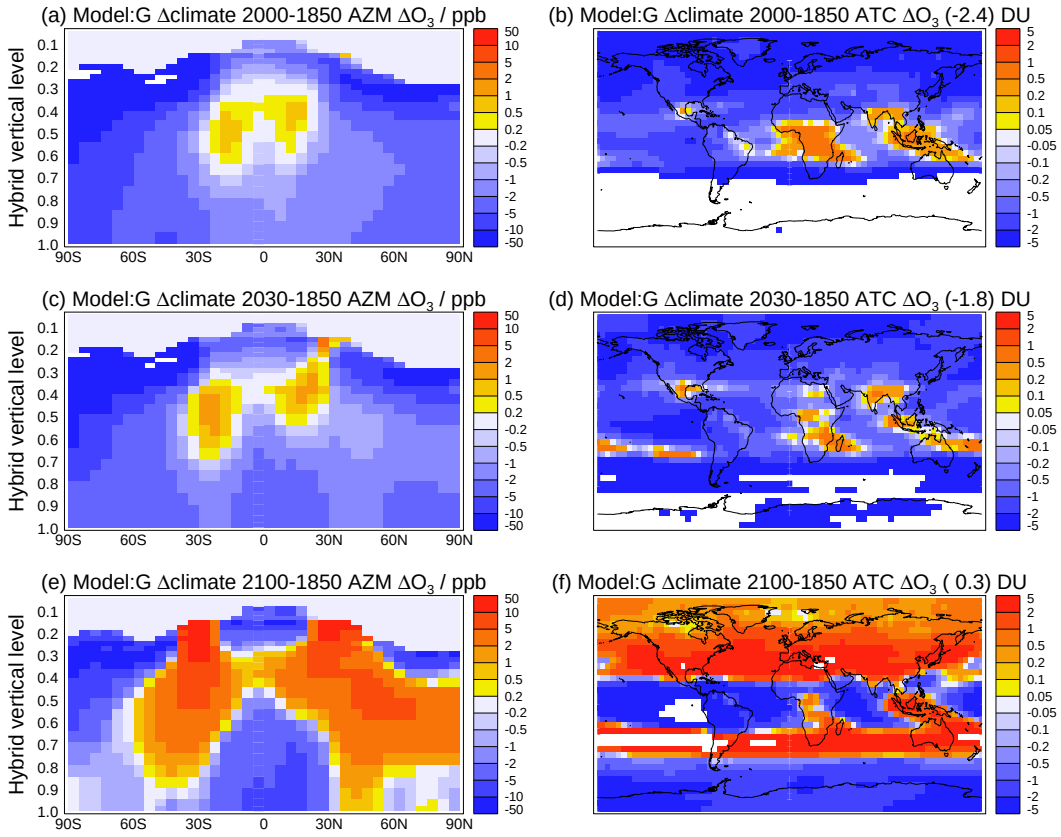

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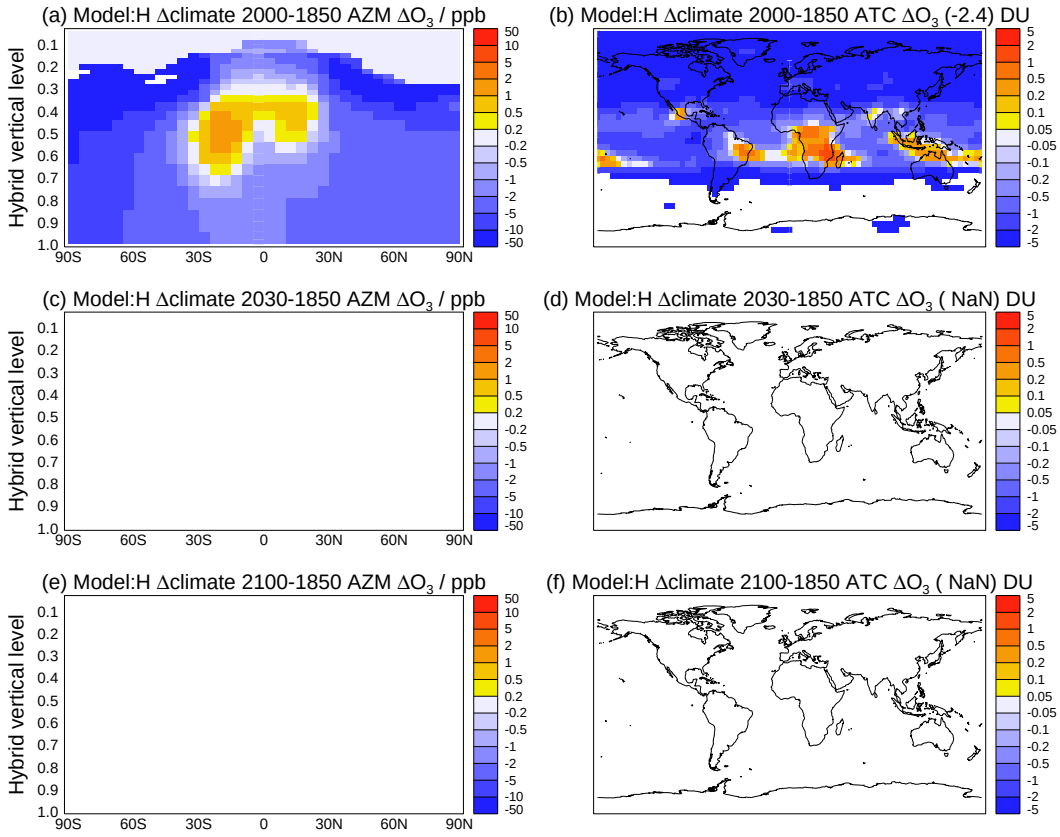

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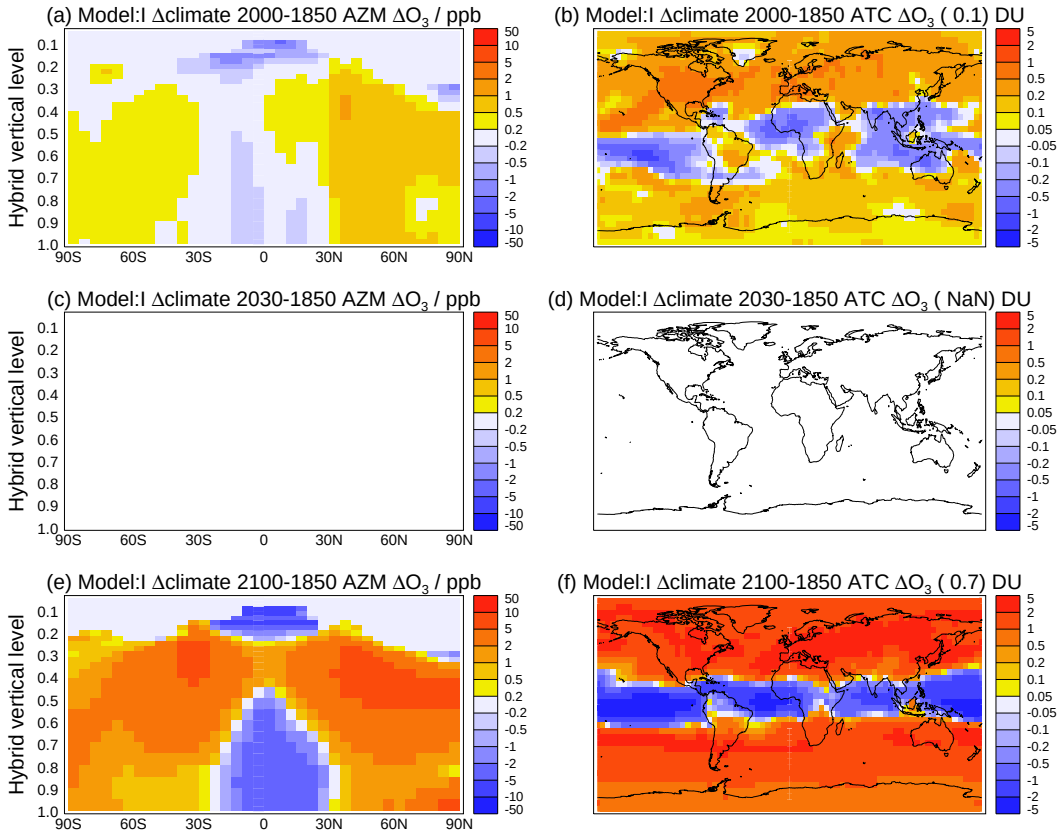

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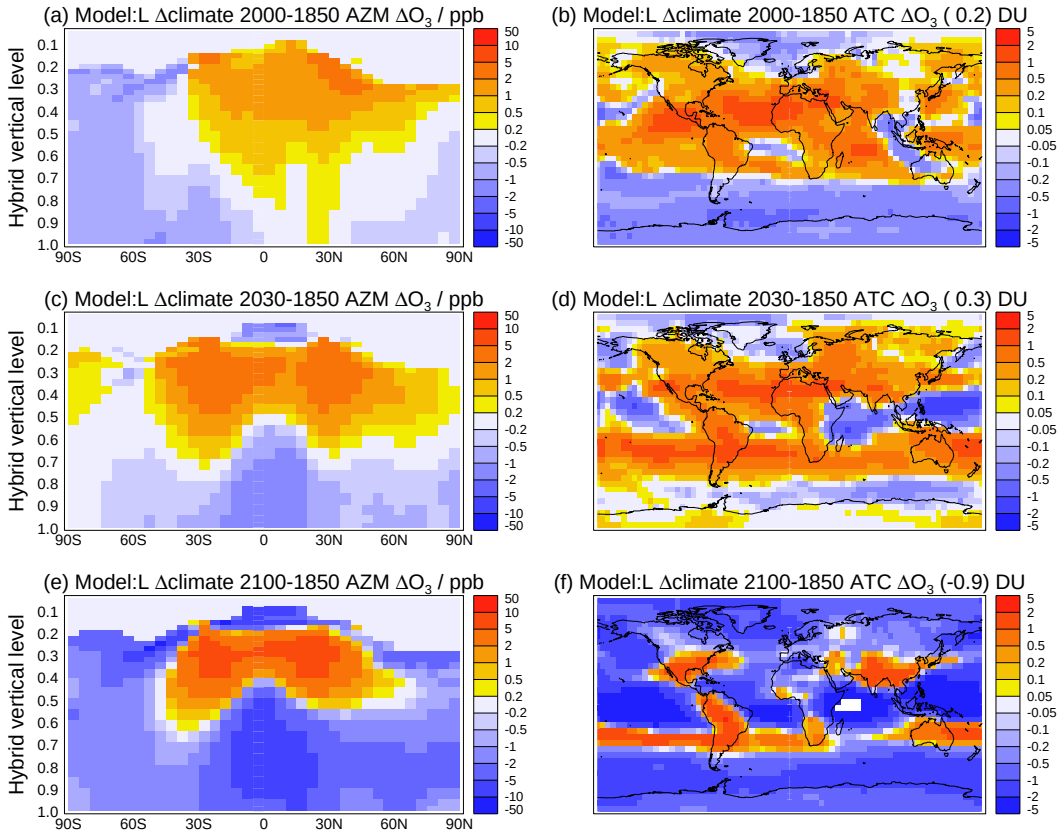

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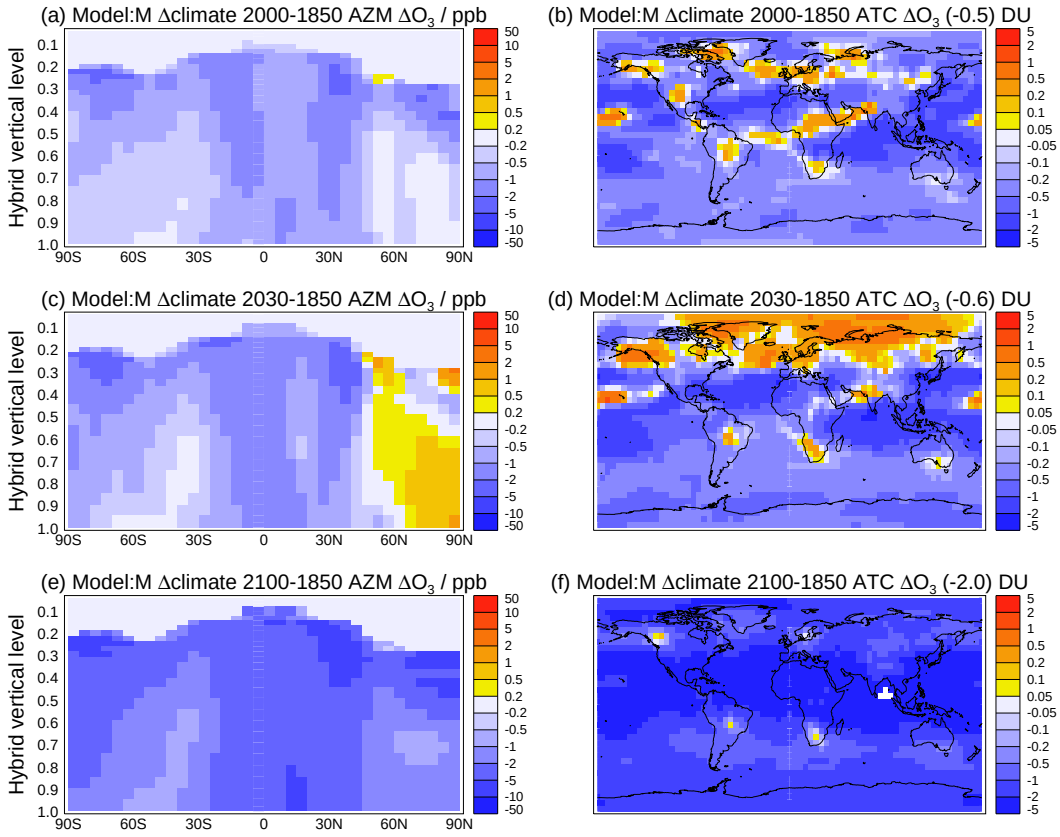

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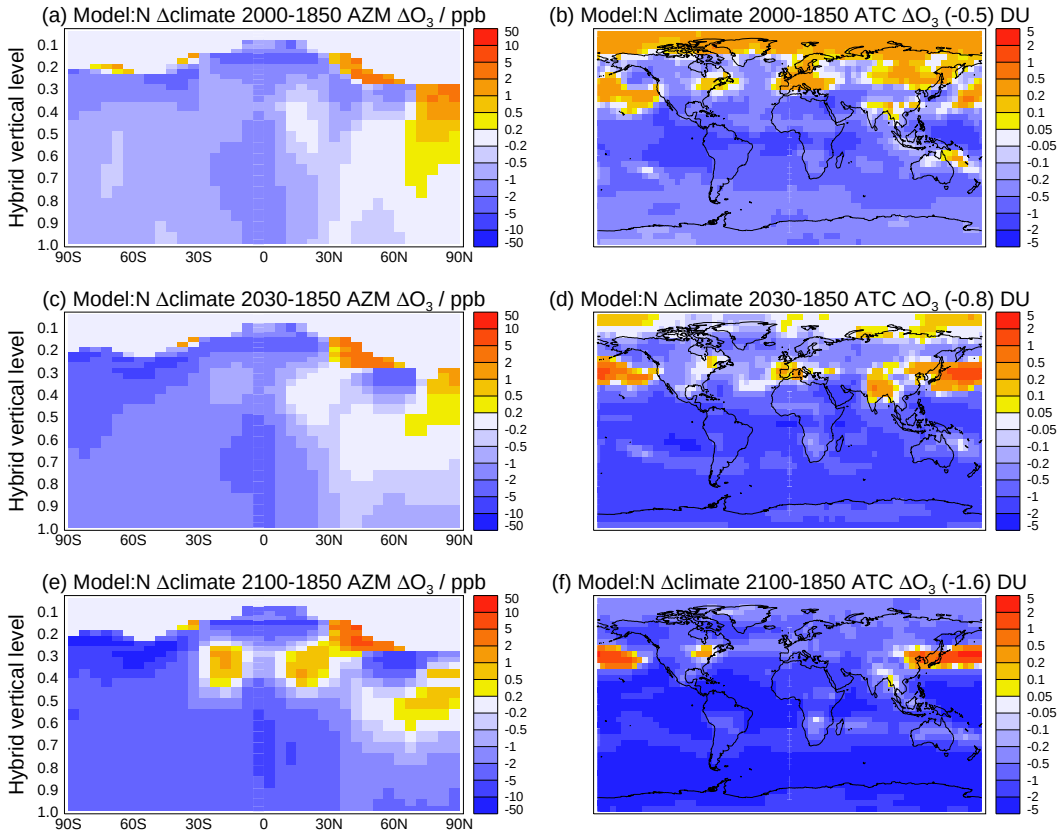

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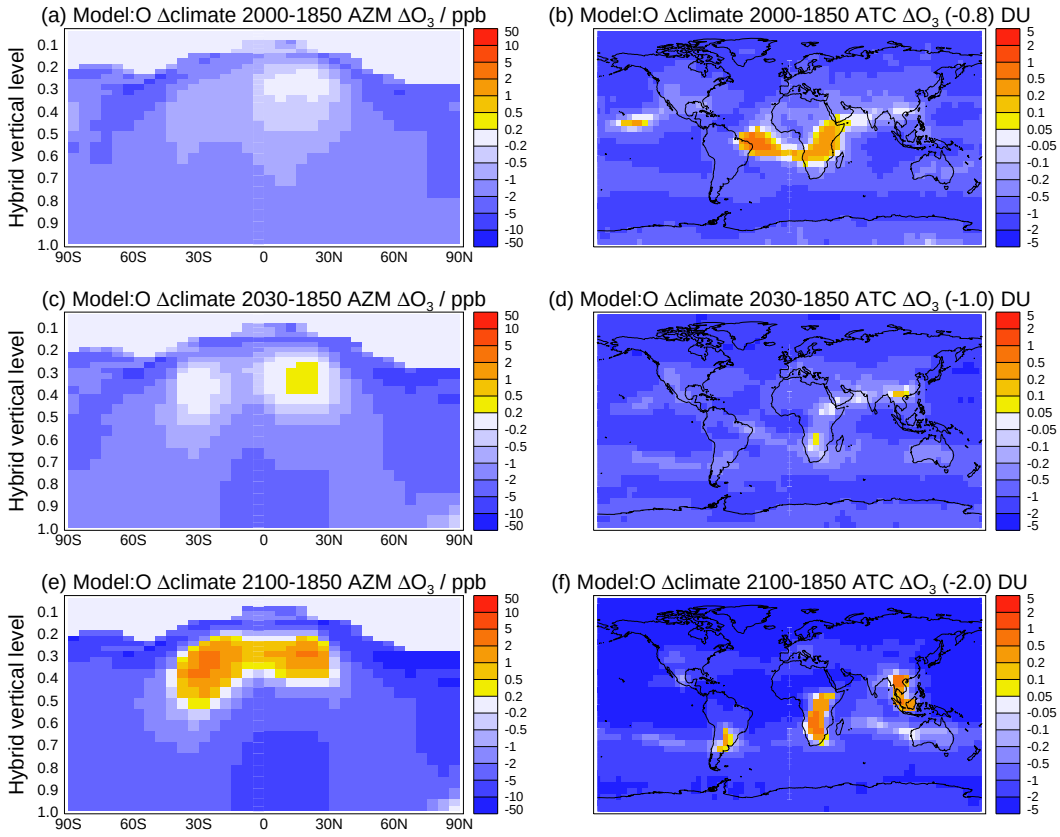

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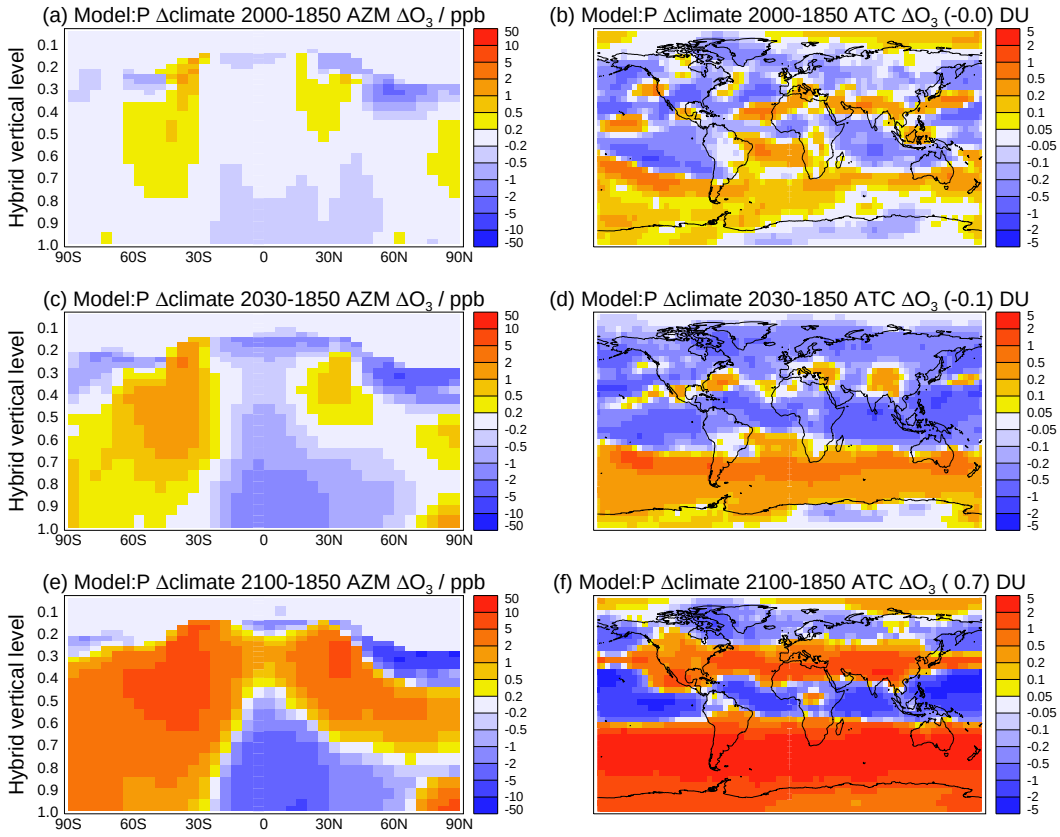

Figure S7 continued.

Radiative forcing of increased CO2 due to emissions of CH4, CO and NMVOCs from fossil sources

Emissions of CH4, CO and NMVOCs from fossil sources leads to increased levels of CO2 in the atmosphere. Assessing the emission-based impact on radiative forcing these CO2 contributions need be attributed to the emissions of the source gases. Here we have used the anthropogenic emissions from Lamarque et al. (2010) since 1850 for CO and NMVOCs, while emissions methane were taken from the emission data recommended for CMIP5 use (http://www.iiasa.ac.at/web-apps/tnt/RcpDb/dsd?Action=htmlpage&page=download).
Emissions from the sector Power plants, Energy Conversion, Extraction and distribution is regarded as fossil, while the other sectors are assumed to be non-fossil.

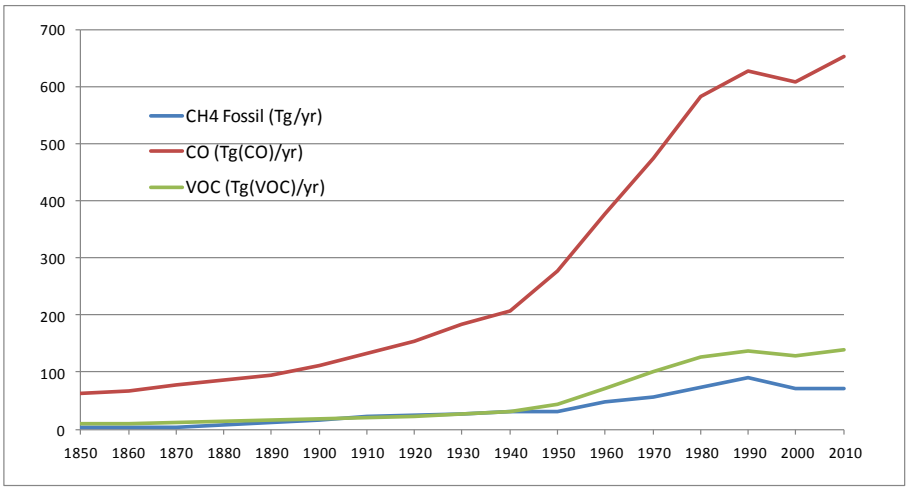

Fig. 1 Historic emissions of CH4, CO and NMVOCs (all in Tg/yr) from fossil/anthropogenic sources.

The oxidation of CH4, CO and NMVOCs in the atmosphere leads to an atmospheric source of CO2. For methane the lifetime in the atmosphere is long enough to allow for a non-neglible fraction of the historical emissions to be left in the atmosphere. Using a lifetime for methane of 9.6 years (IPCC, 2001) we find that 12% of the methane has not yet been oxidized to CO2.

From the emissions and the atmospheric lifetime of CH4 we calculate the corresponding atmospheric source of CO2 as a function of time. For the NMVOCs we have assumed an average carbon content of 80% by mass. We then calculate the resulting development in the CO2 concentrations using the impulse response function for CO2 (IRF[1]) given in IPCC (2007). The change in the mixing ratio of CO2 ($X_{CO2}(t)$) is given by

$$X_{CO2}(t) = \int_0^t Em_{CO2}(t') \cdot IRF(t - t')dt'$$

The contribution to atmospheric CO2 is given in figure 2a.

The radiative forcing due to the CO2 from the fossil/anthropogenic emissions of methane, CO and NMVOCs are calculated by the simple parameterization given in IPCC (2001)

$$RF = 5.35 \cdot ln\left[\frac{X}{X_0}\right]$$

The calculations are done with X_0 = 278 ppm. The radiative forcings are shown in figure 2b. In 2010 the RFs are 18, 87 and 33 mWm^{-2} for emissions of methane, CO and NMVOC respectively.

[1] $IRF(t) = a_0 + \sum_{i=1}^{3} a_i \cdot e^{-t/\tau_i}$

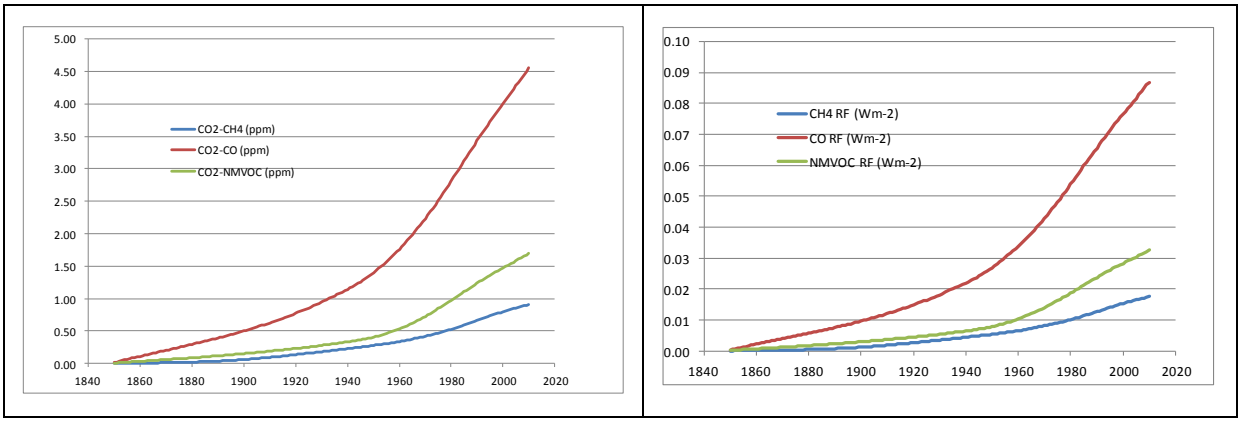

Figure 2. Contribution from fossil/anthropogenic emissions of CH4, CO and NMVOCs to atmospheric CO2 and radiative forcing.