Supplementary Material:

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

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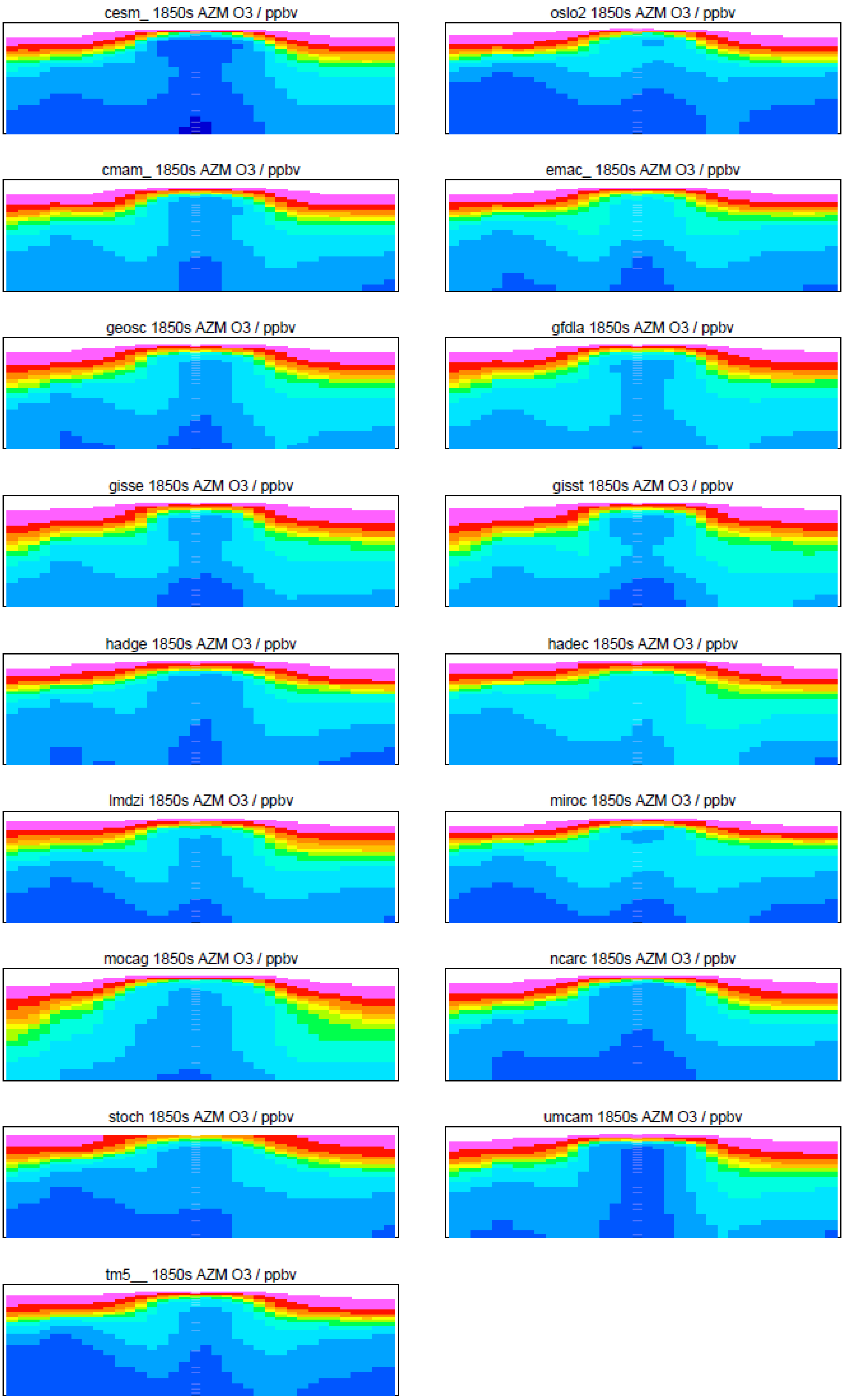

Figure S1: Annual zonal mean ozone (ppbv) for the 1850s and 2000s. For scale see Figure 1.

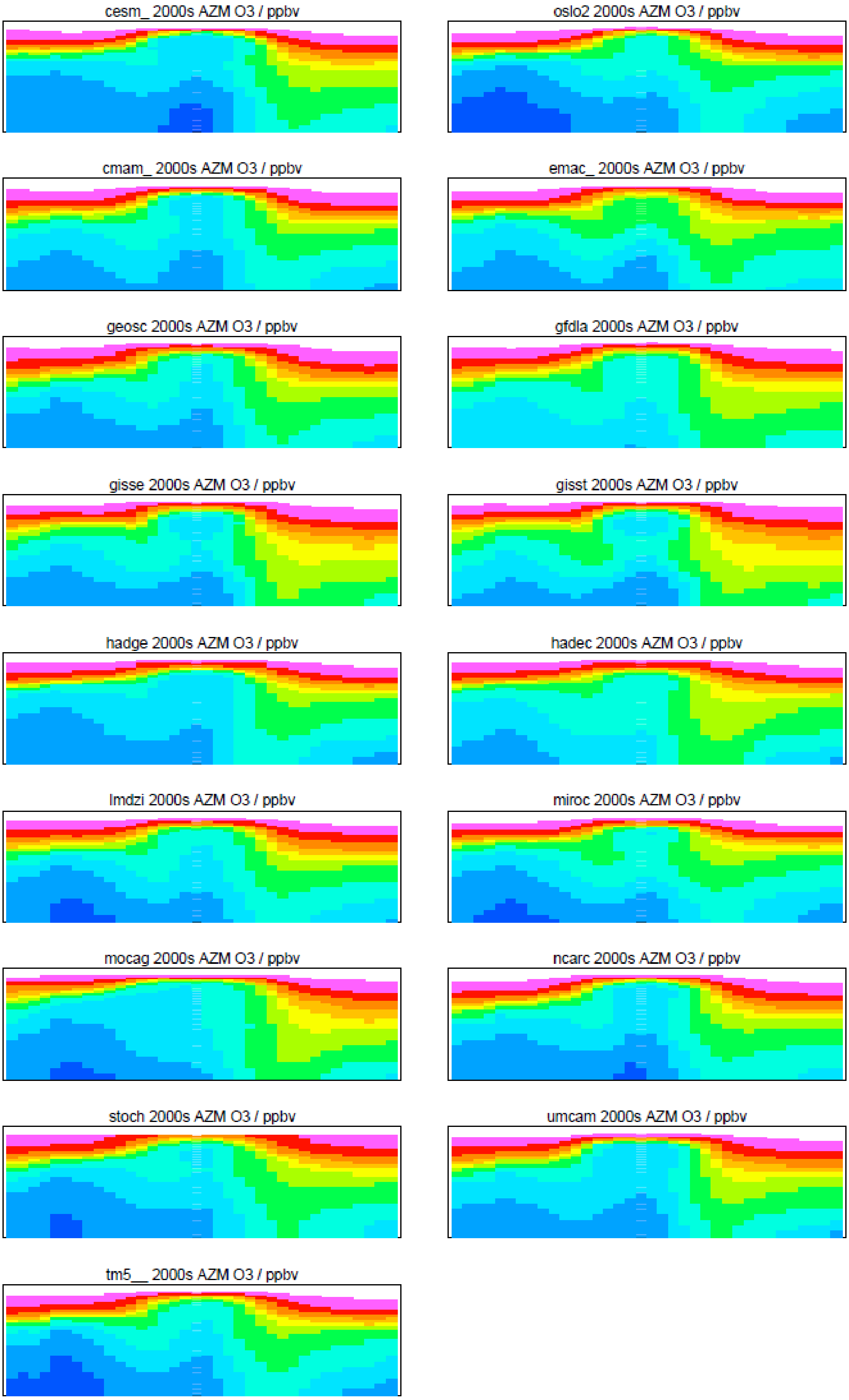

Figure S1 (continued)

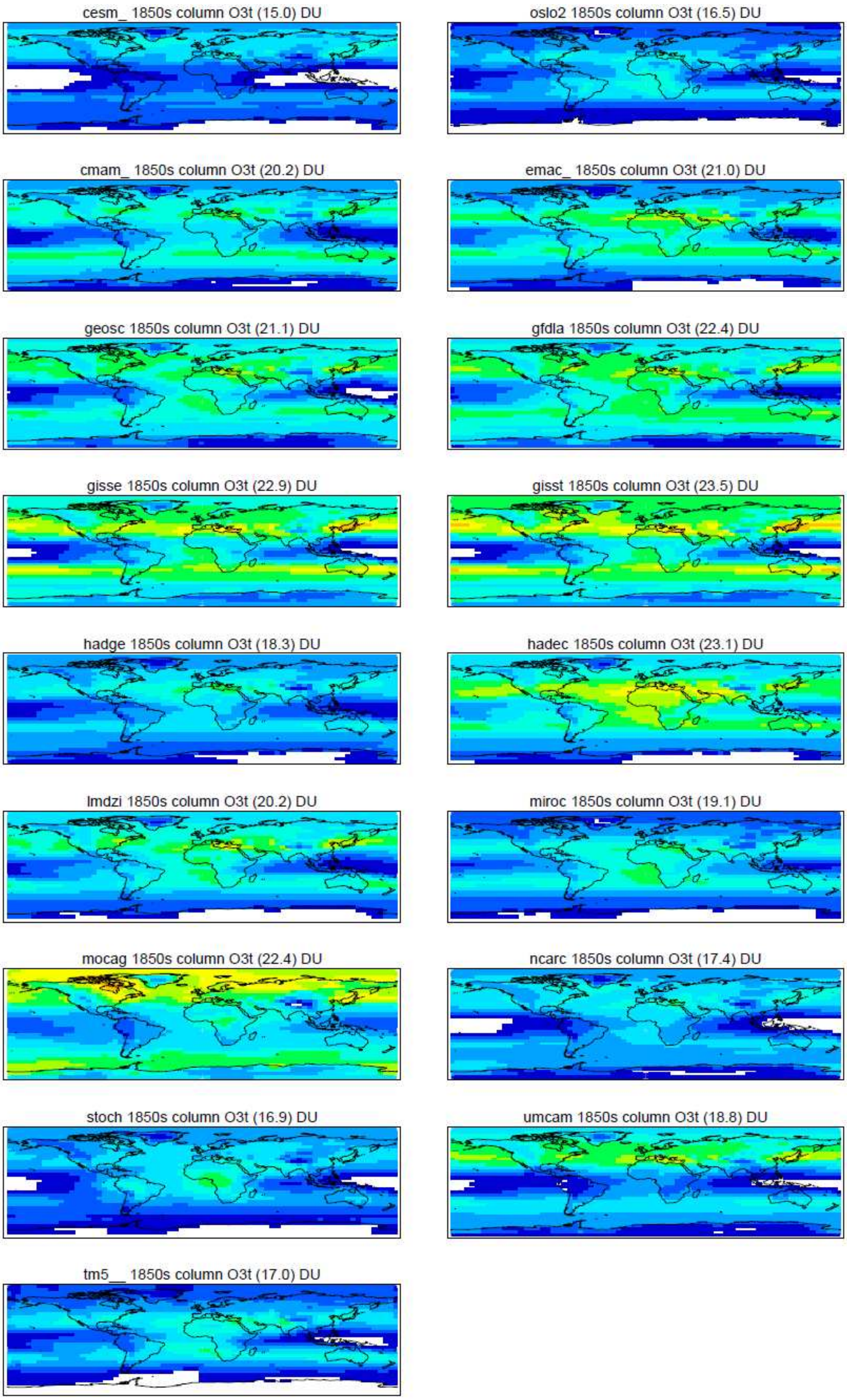

Figure S2: Annual mean tropospheric column ozone (DU), for the 1850s and 2000s, using the MASKZMT. For scale see Figure 2.

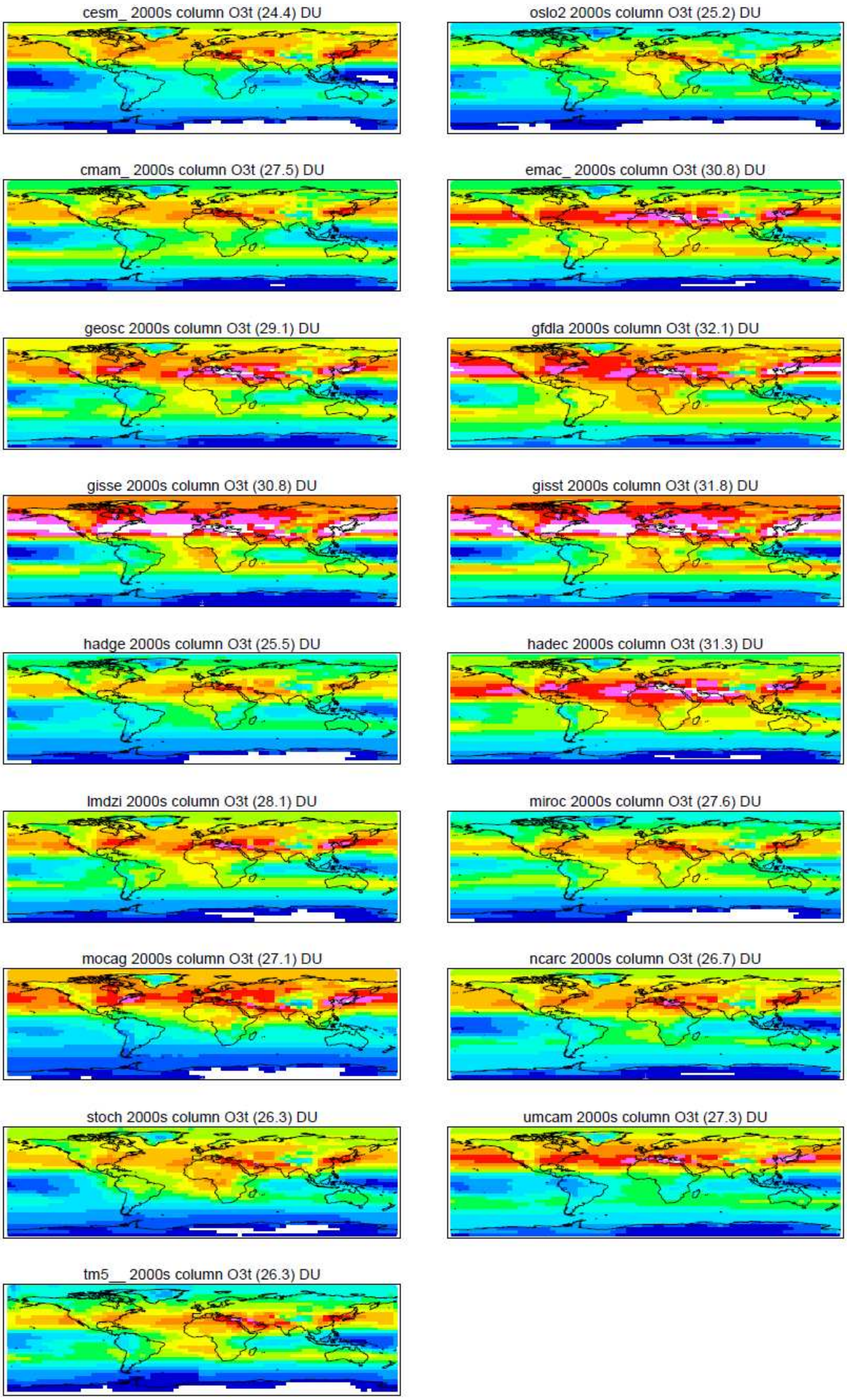

Figure S2 (continued)

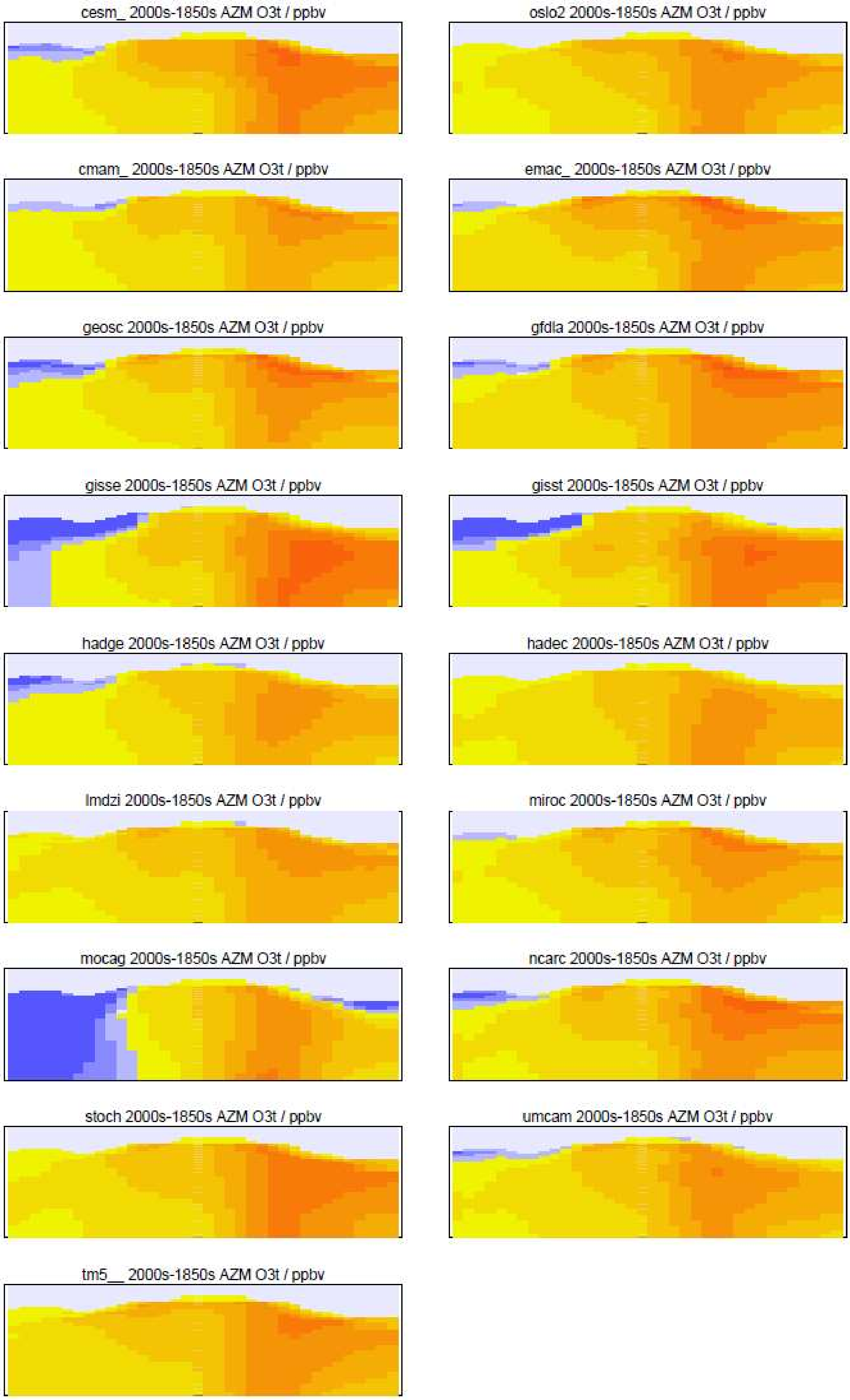

Figure S3: Changes (2000s-1850s) in annual zonal mean ozone (ppbv) and tropospheric column ozone (both masked using MASKZMT). See Figure 3 for scales.

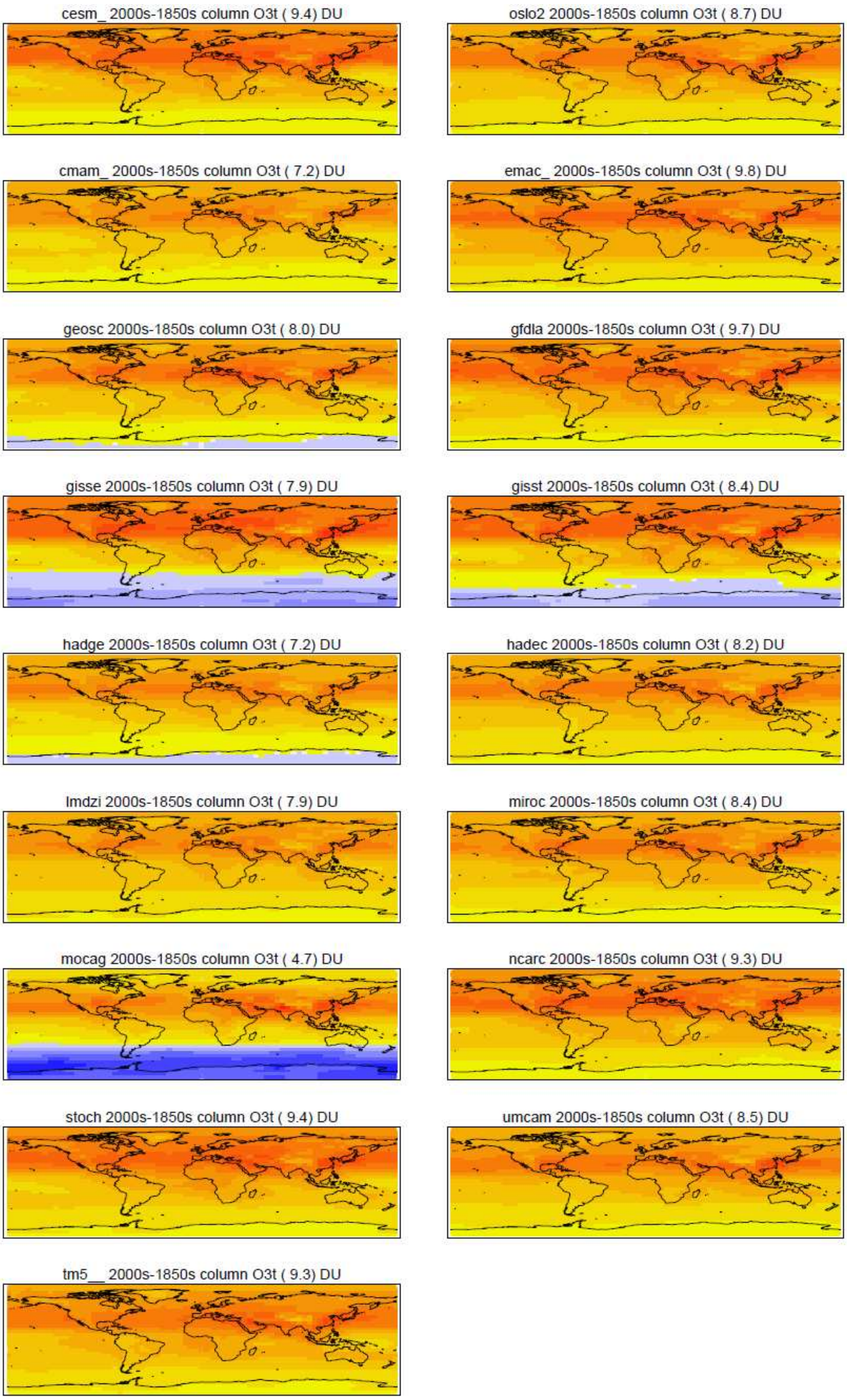

Figure S3 (continued)

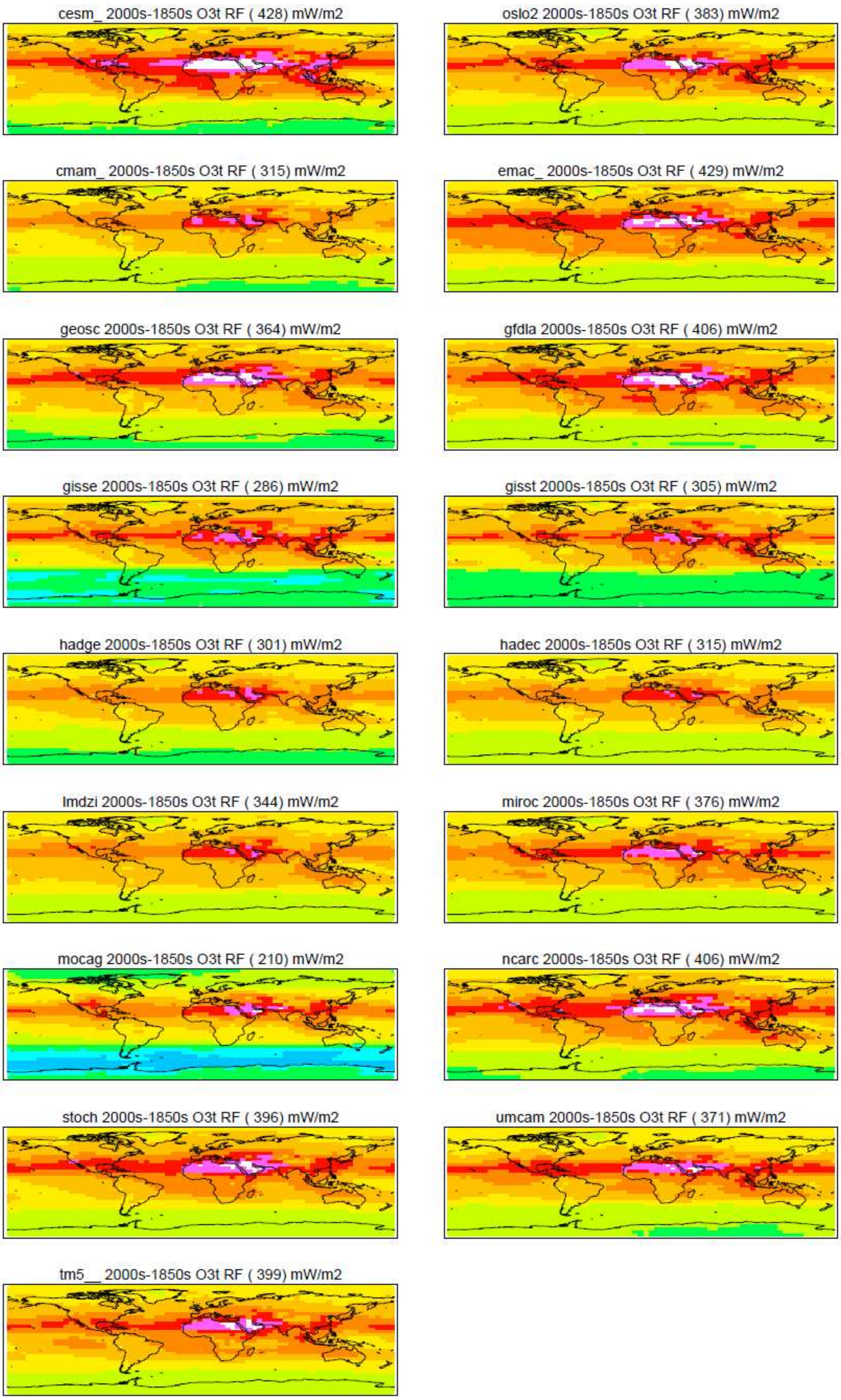

Figure S4a: Total ozone RFs for all models, masked using MASKZMT.

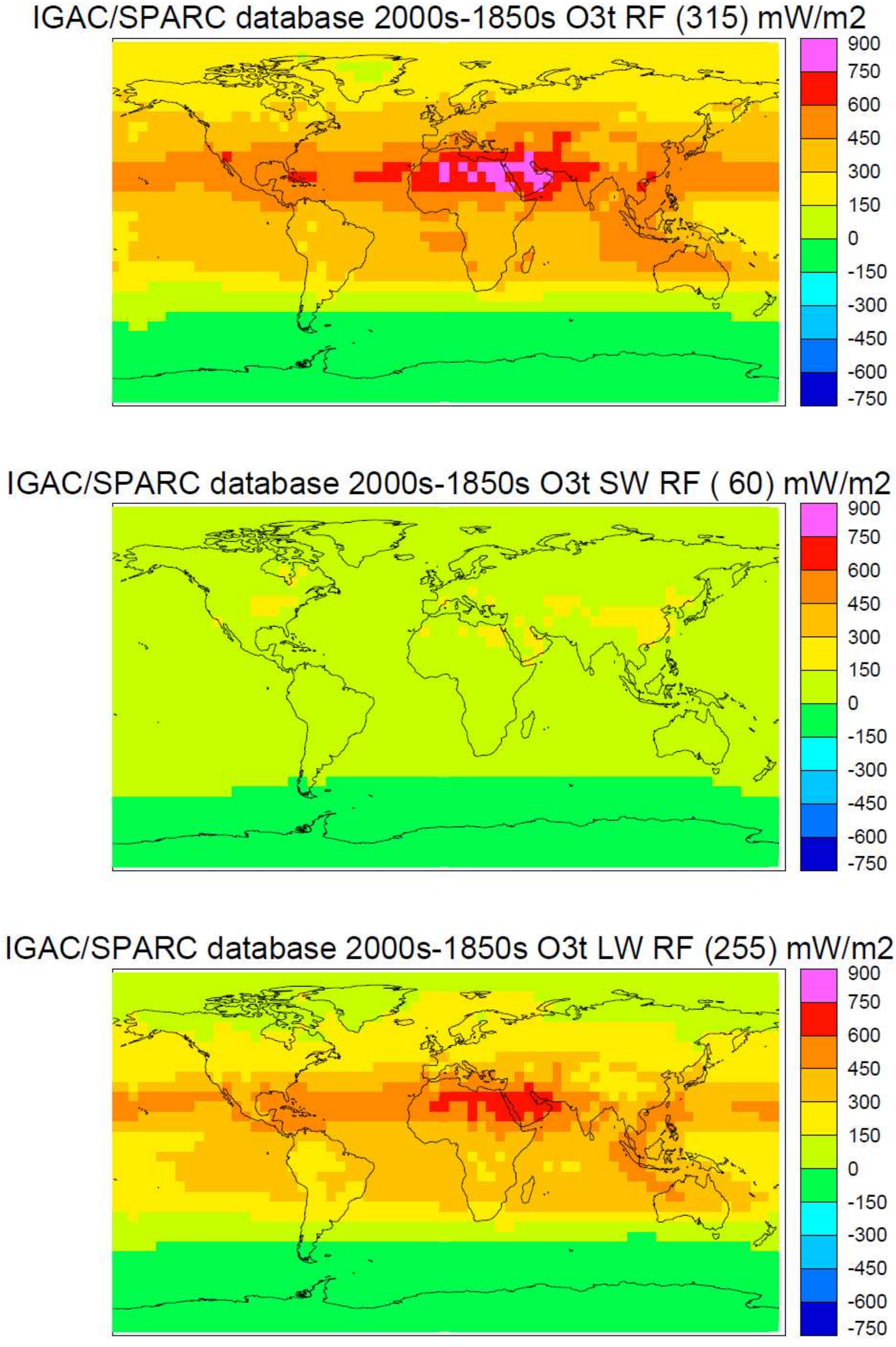

Figure S4b: Annual mean tropospheric ozone total, SW, and LW RFs for the IGAC/SPARC ozone dataset, as used in Cionni et al (2011). Compare to the multi-model mean in Figure 4 and also compare to Figure 15a of Cionni et al. (2011), which shows the total RF calculated with an earlier version of the E-S radiation code, and finds a total RF 27% lower.

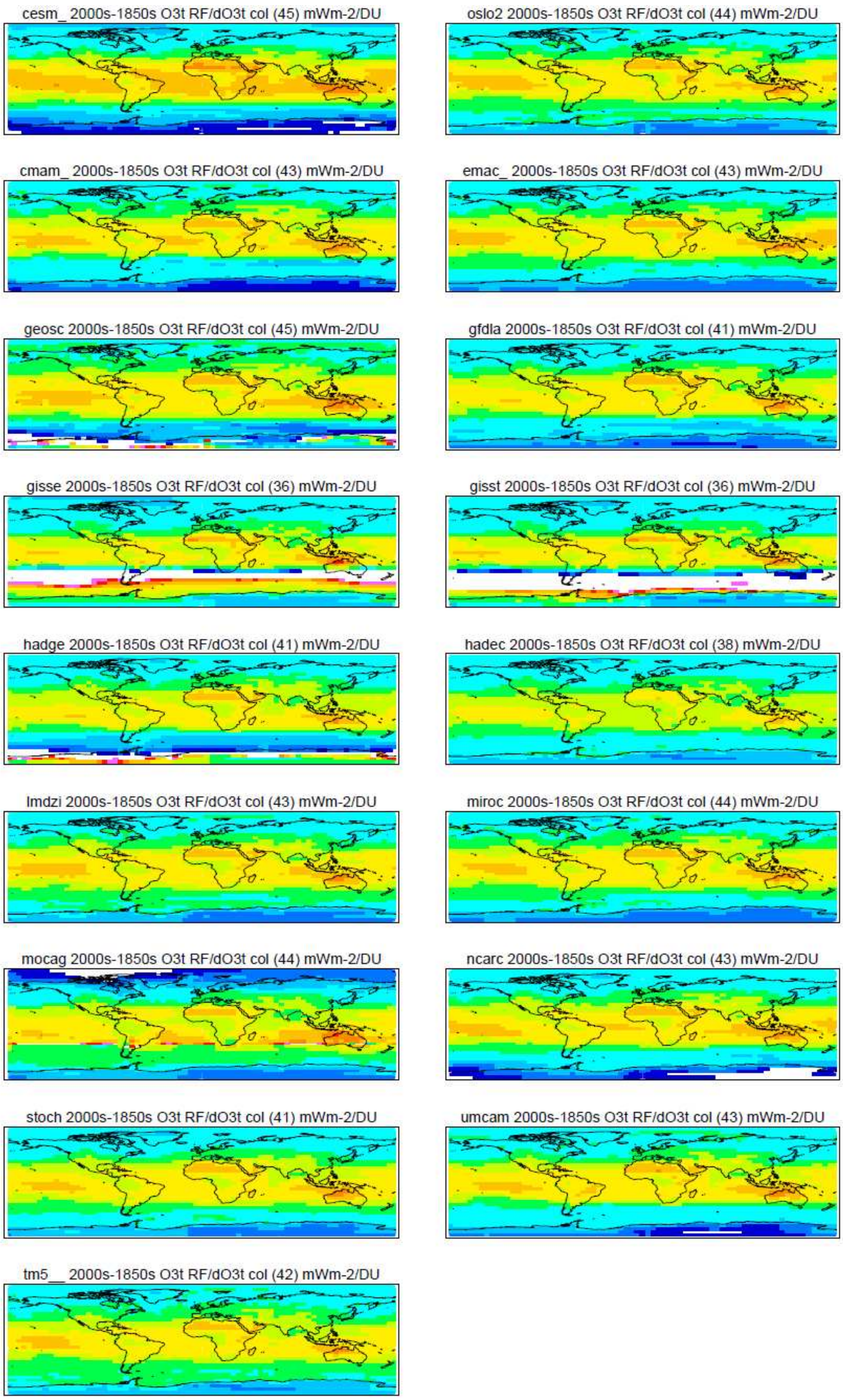

Figure S5: Normalised RFs for MASKZMT. See Figure 5 for scale.

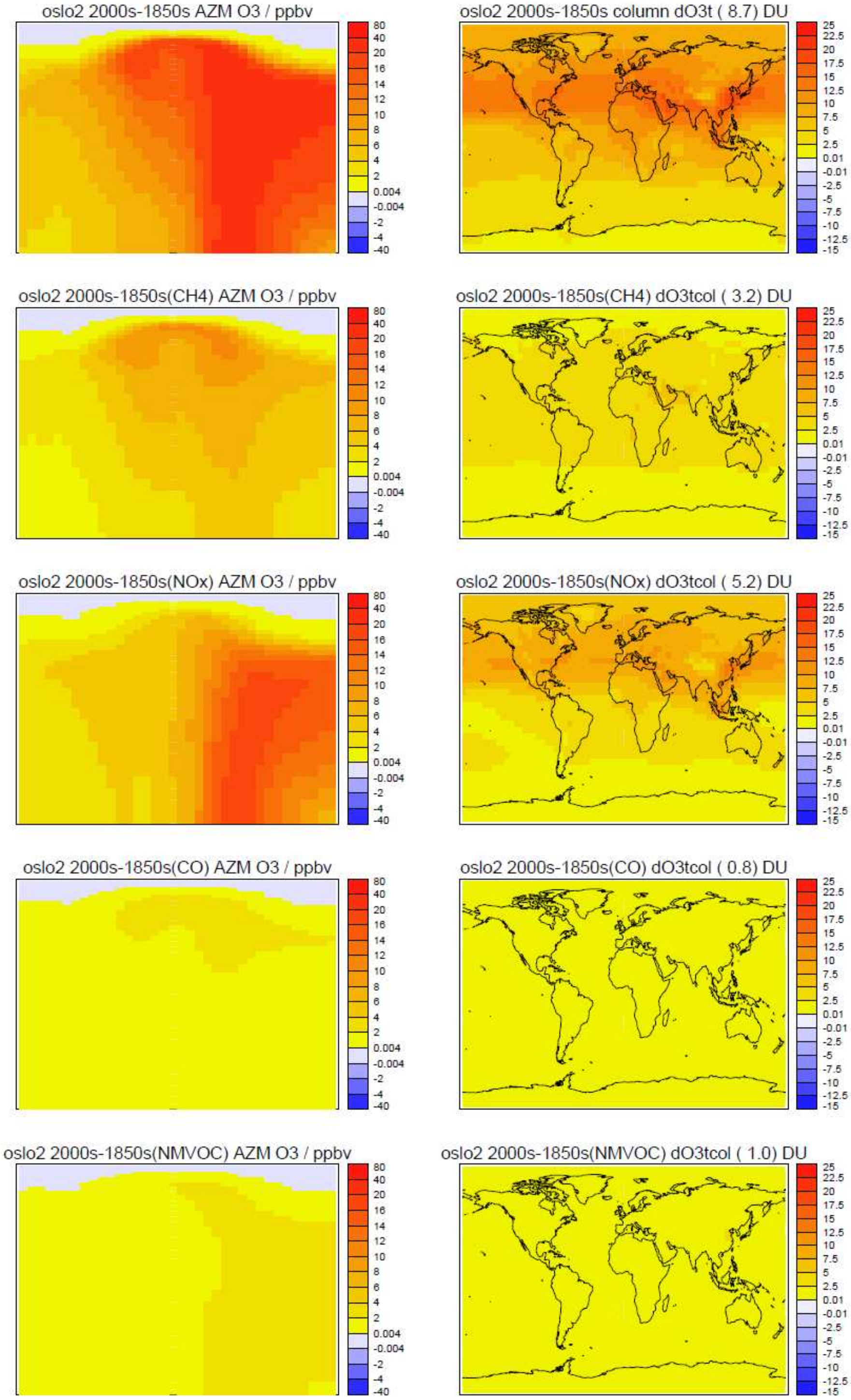

Figure S6: An example of one model's results (model B) for the attribution experiments. Left hand side shows contributions to changes in zonal annual mean ozone, right hand side shows contributions to change in annual mean tropospheric ozone column. Referring to the experiment numbers in Table 3, rows from the top show: experiment #1-#0 (all components); #1-#2 (CH_4); #1-#3 (NOx); #1-#4 (CO); #1-#5 (NMVOC).

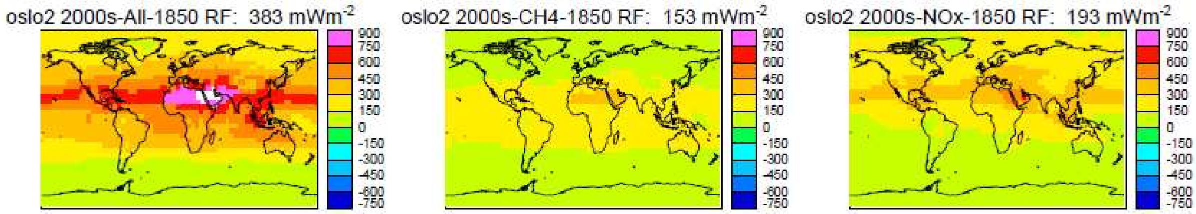

Figure S7: Radiative forcings for model B, from the attribution experiments. Left-hand plot shows total 2000s-1850s (#1-#0); middle shows the CH4 component (#1-#2); and the right-hand plot shows the NOx component (#1-#3). The CO and NMVOC components are significantly less (38 and 37 mWm^{-2}, respectively) and are not shown.

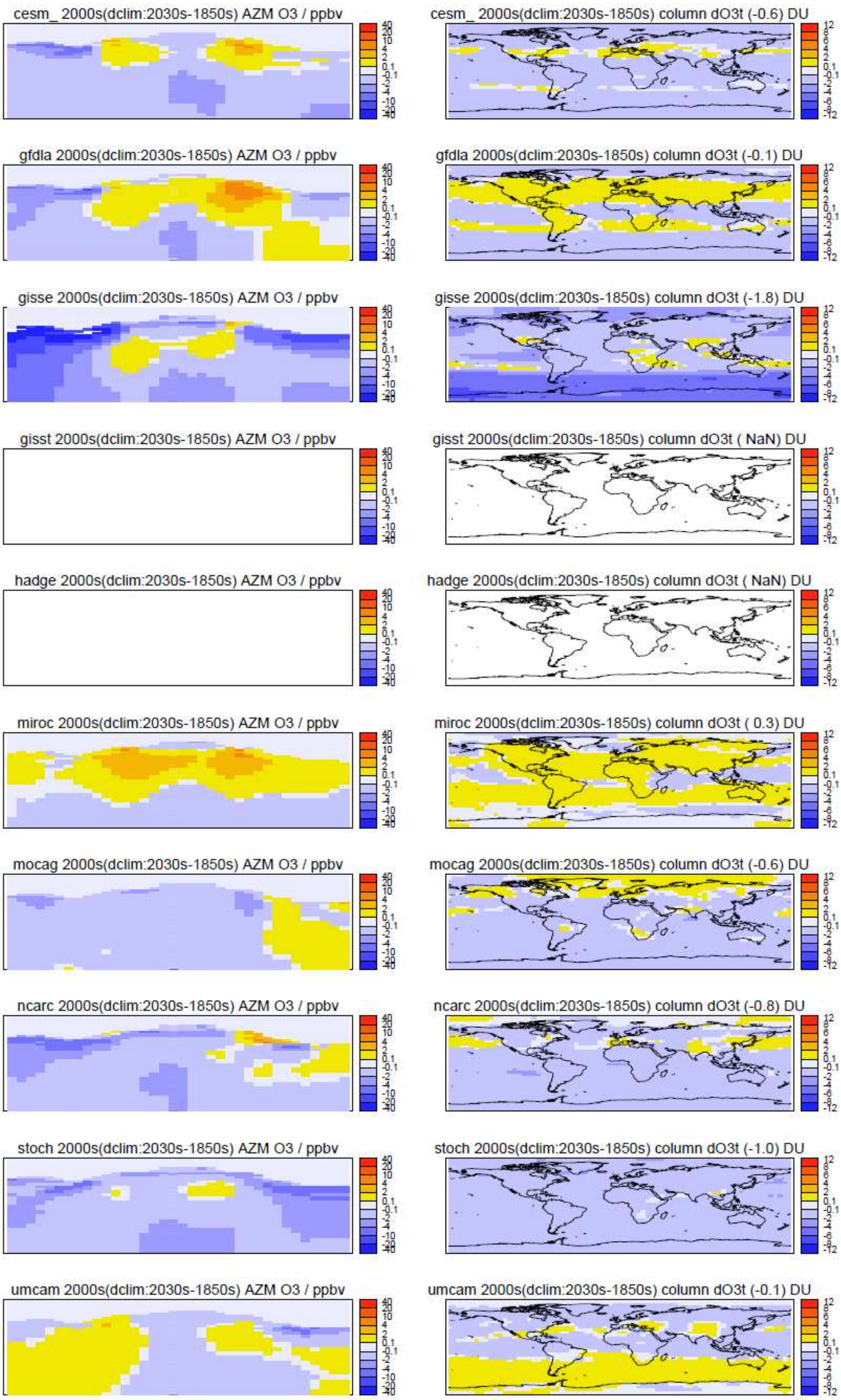

Figure S8: As Figure 6, but showing the impact of climate change (scenario RCP8.5) on ozone from 1850s to the (a) 2030s; (b) 2100s.

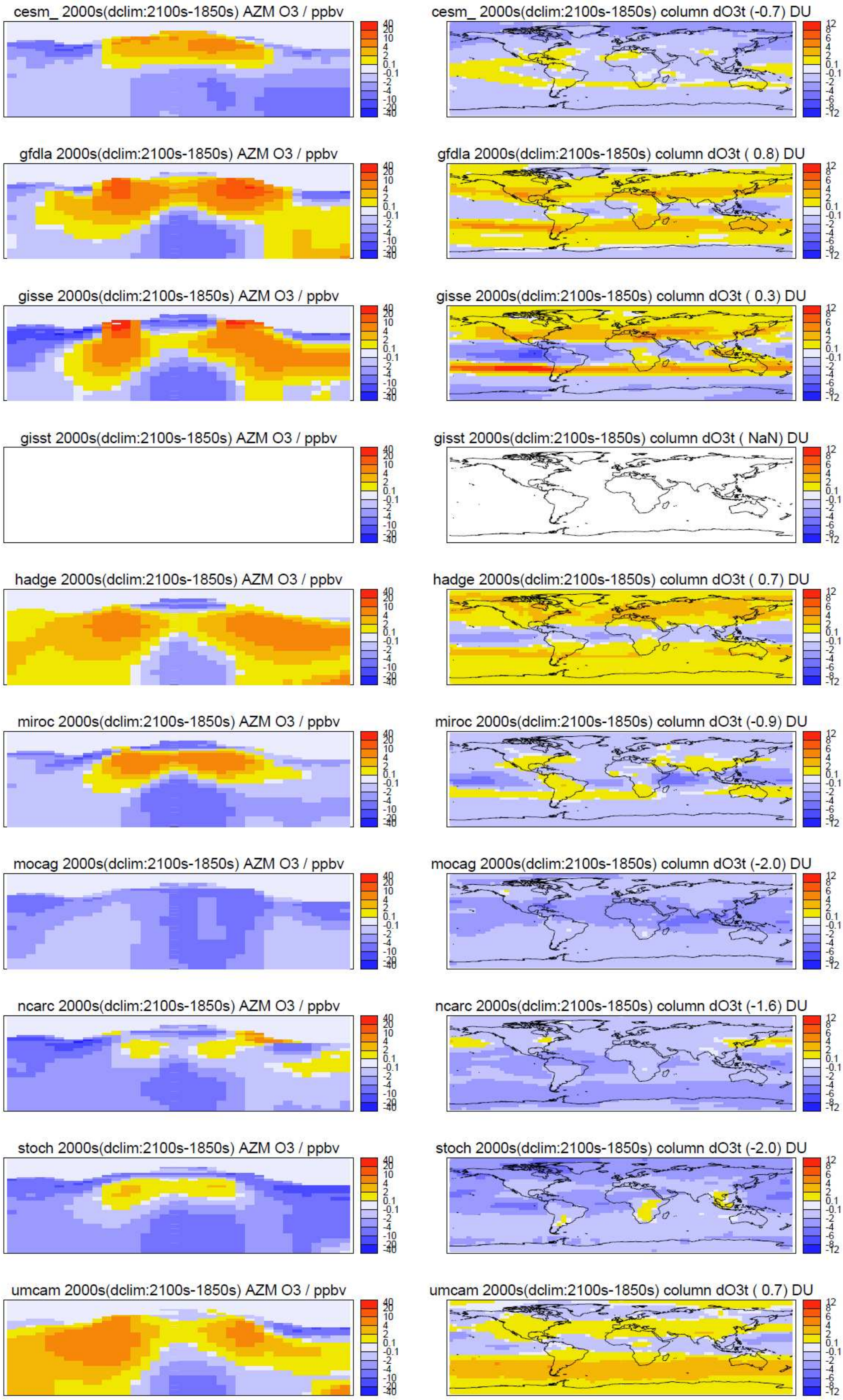

Figure S8 continued

1 Radiative forcing of increased CO_2 due to emissions of CH_4, CO and NMVOCs from fossil sources

Emissions of CH_4, CO and NMVOCs from fossil sources leads to increased levels of CO_2 in the atmosphere. Assessing the emission-based impact on radiative forcing these CO_2 contributions need be attributed to the emissions of the source gases. Here we have used the anthropogenic emissions from Lamarque et al. (2010), covering 1850 onwards for CO and NMVOCs, while methane emissions 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 power plants, energy conversion, extraction and distribution sectors are regarded as fossil, while other sectors are assumed to be non-fossil.

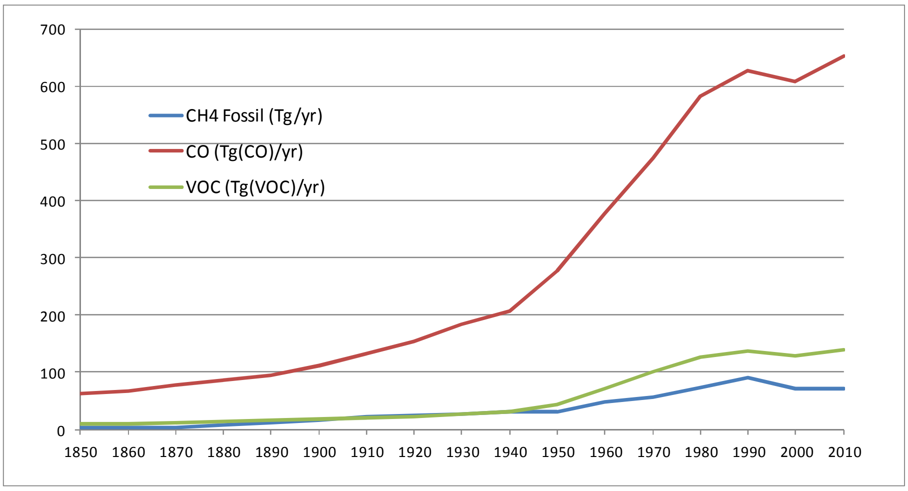

Figure S9: Historic emissions of CH_4, CO and NMVOCs (all in Tg/yr) from fossil/anthropogenic sources.

The oxidation of CH_4, CO and NMVOCs in the atmosphere leads to an atmospheric source of CO_2. For methane the lifetime in the atmosphere is long enough to allow for a non-negligible 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 oxidised to CO_2.

From the emissions and the atmospheric lifetime of CH_4 we calculate the corresponding atmospheric source of CO_2 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 CO_2 concentrations using the impulse response function for CO_2 (IRF[1]) given in IPCC (2007). The change in the mixing ratio of CO_2 ($X_{CO2}(t)$) is given by

$$X_{co2}(t) = \int_0^t Em_{co2}(t') \cdot IRF(t - t')dt'$$

The contribution to atmospheric CO_2 is given in figure 2a.

The radiative forcing due to the CO_2 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 mW m^{-2} for emissions of methane, CO and NMVOC respectively.

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

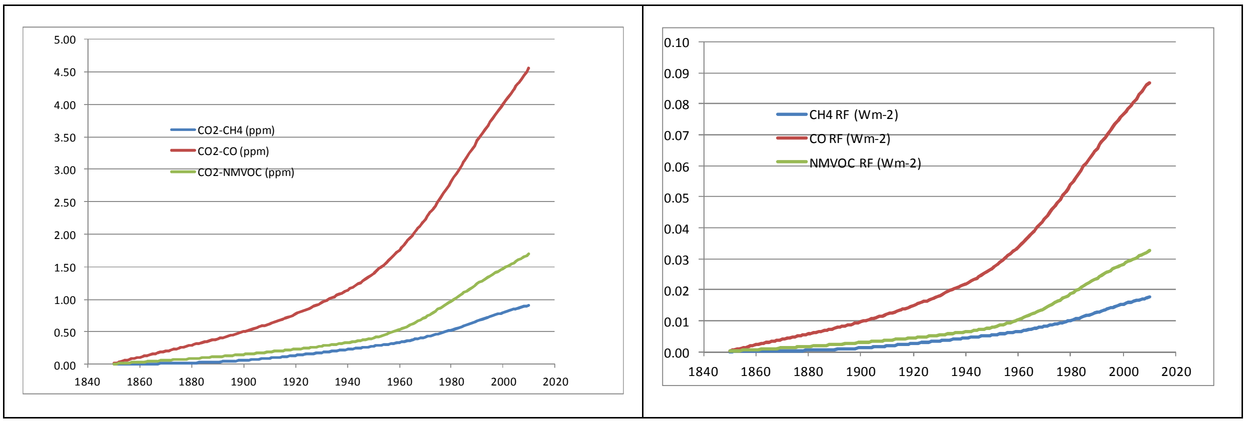

Figure S10: Contribution from fossil/anthropogenic emissions of CH_4, CO and NMVOCs to atmospheric CO_2 and radiative forcing.