1. Emissions

In this section, we show the various emissions used in the simulation scenarios (Table 1 and Table 2).

1.1 Methane

As shown below, CTL total emissions (annually-repeating natural sources (i.e., wetlands and biomass burning) and annually-varying anthropogenic sources) are higher in the northern hemisphere by about 20% while EXTRA emissions (all emissions vary) are higher by about 20% in the tropics (Patra et al., 2011).

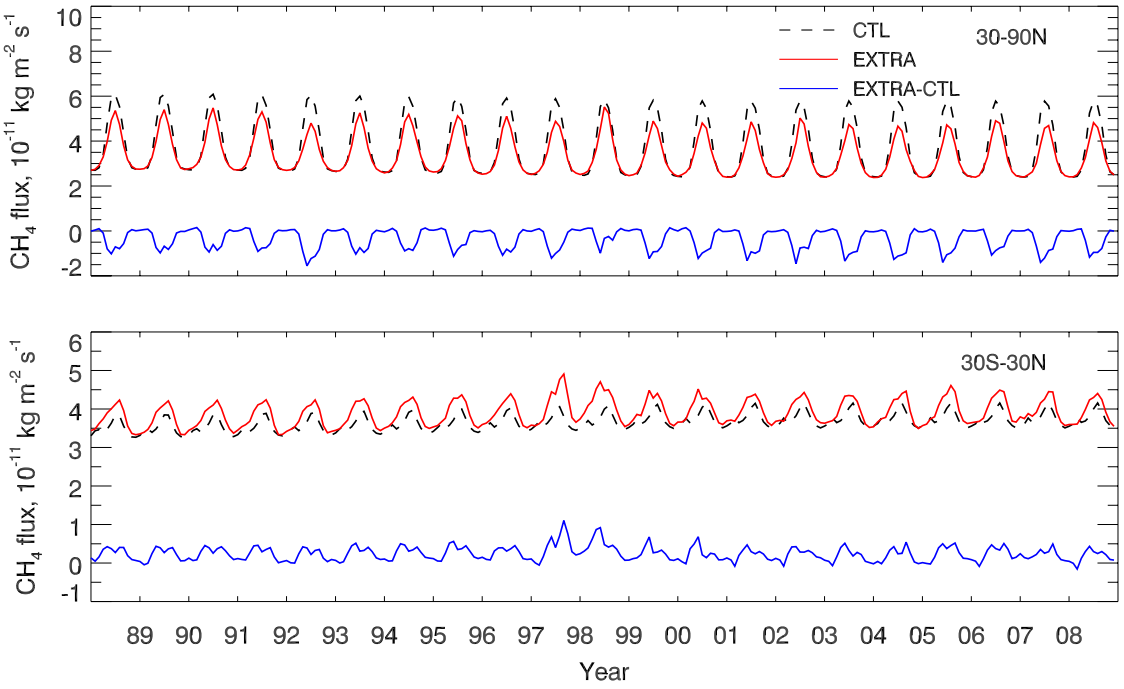

Figure S 1: Monthly methane CTL (dashed) and EXTRA (red) emissions ($x10^{-11}$ $kg/m^2/s$) used in the *Base* and $E_{CH4}Vary$ scenarios, respectively. The difference between them is shown in blue (EXTRA-CTL).

1.2 CO

Here, we show the biomass burning (BB) and fossil fuel (FF) CO emissions used in the *Base* and *AllVary* scenarios.

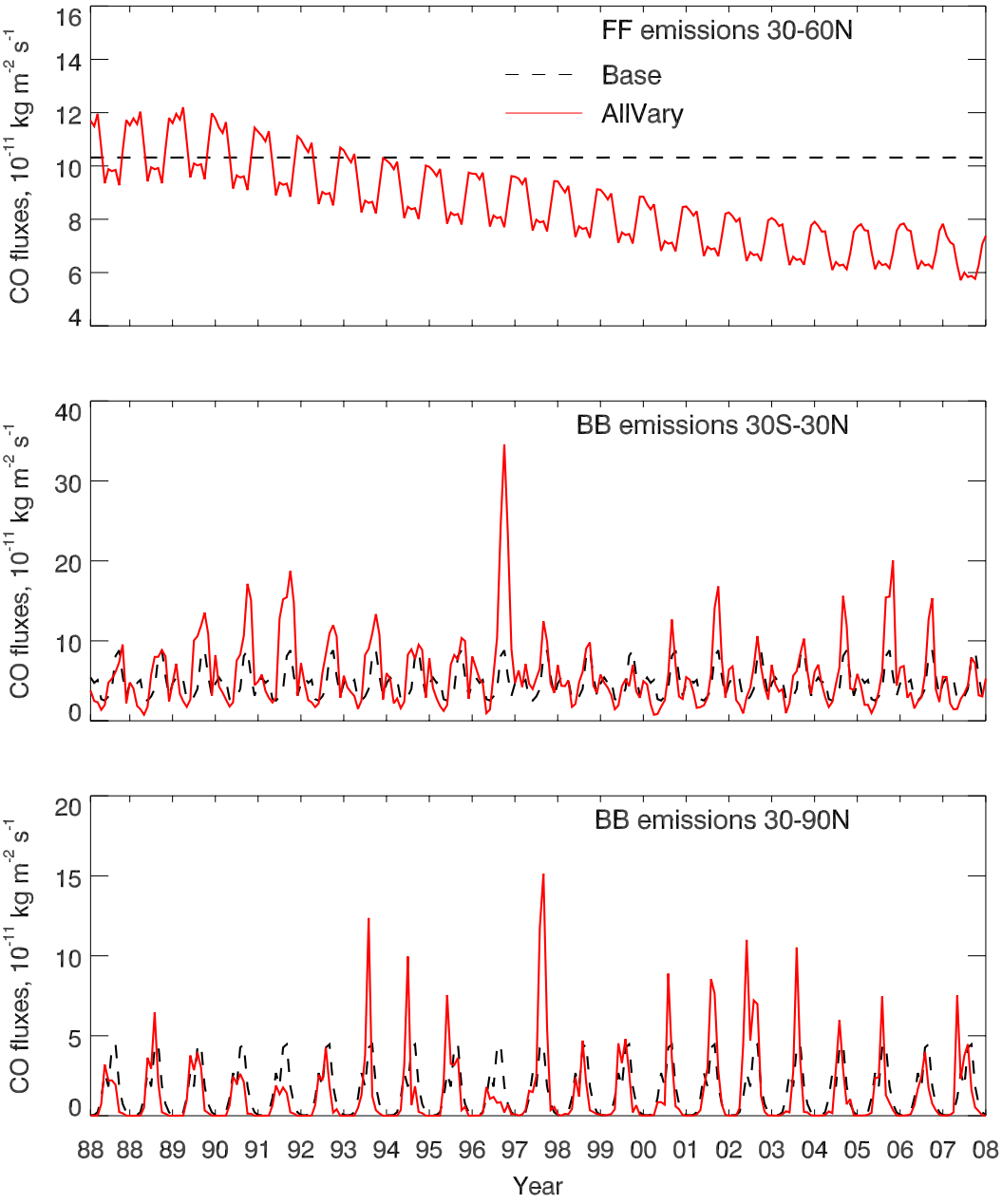

Figure S 2: Monthly CO emissions (x10^{-11} kg/m^2/s) used in the *Base* and *AllVary* scenarios.

The figure below shows the sensitivity of the global burdens of methane, CO, and OH to emissions. For instance, the simulated larger burdens of CO levels in the *$BBE_{CO}Vary$* scenario lead to decreased OH levels and thus higher methane burdens compared to the *$E_{CH4}Vary$* scenario.

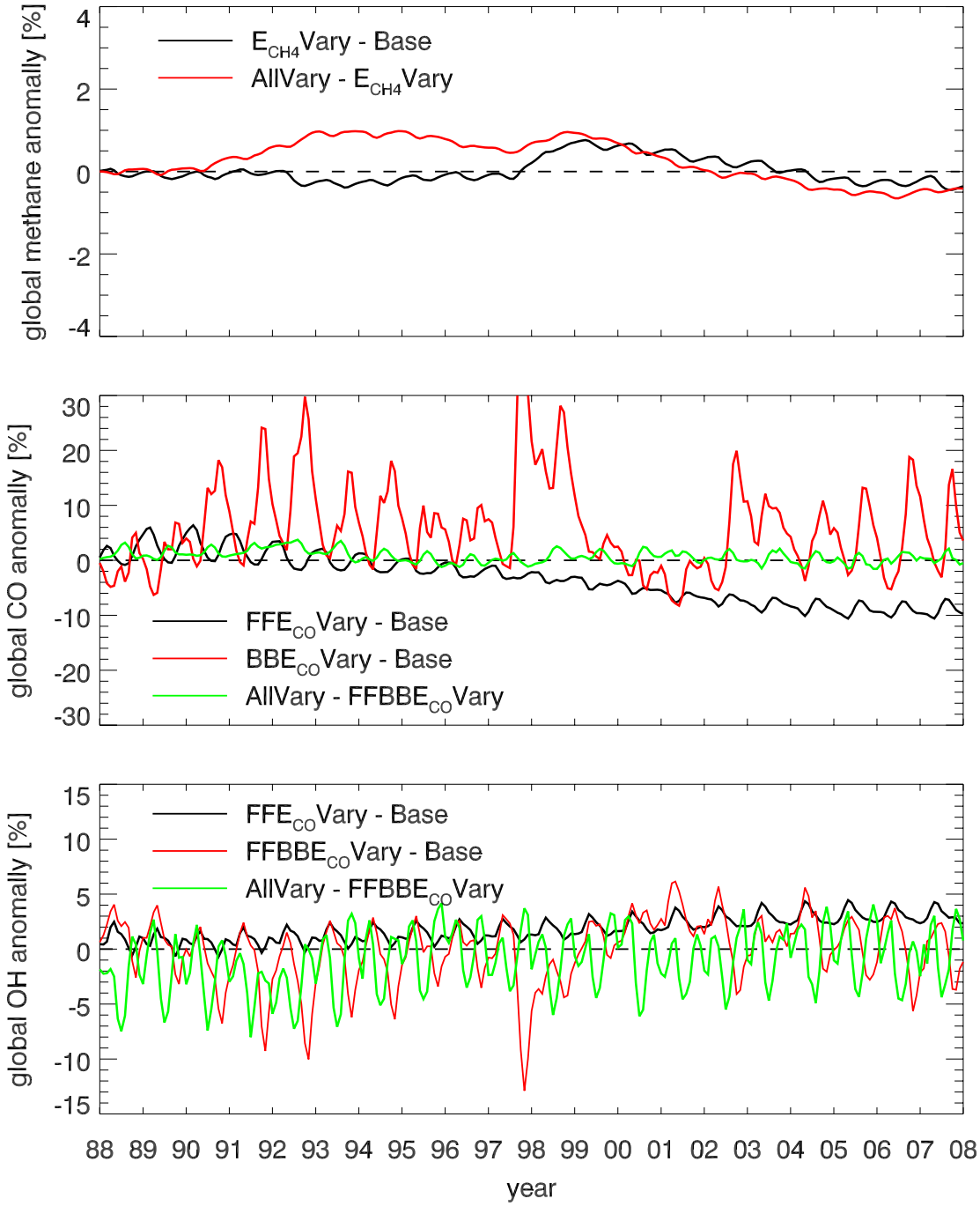

Figure S 3: Relative difference (%) of globally mass-weighted tropospheric methane, CO, and OH (from up to bottom) between the different scenarios.

2. Comparison to measurements

2.1 Methane

Global Methane Growth Rate

We reproduce Figure 4a in the manuscript but show the difference between the *Base* and $OH_{input}Vary$ (Figure S 4) and $FFBBE_{CO}Vary$ (Figure S 5) scenarios. These figures incorporate the results concluded in Sect. 4.3 demonstrating the non-linear feedbacks on methane's growth rate. It further demonstrates that non-linear feedbacks on growth rates in 1994-1997 are mainly due to interannual variability in OH constraints (Figure S 4) while the other non-linear feedbacks are related to interannual variability in CO emissions (Figure S 5).

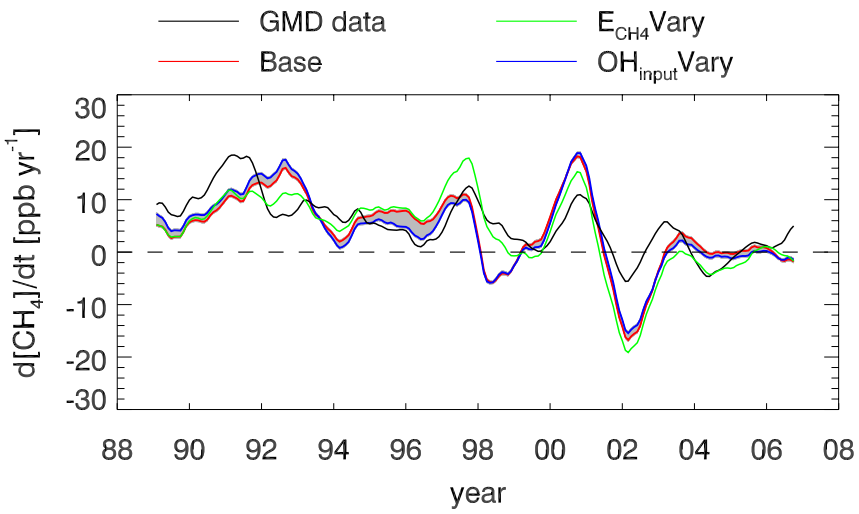

Figure S 4: 12-month running mean atmospheric growth rate of methane (ppbv yr^{-1}) for the average of 92 GMD stations and from model output averaged for those station locations for several scenarios. The shaded area is the difference between the $OH_{input}Vary$ and *Base* scenarios.

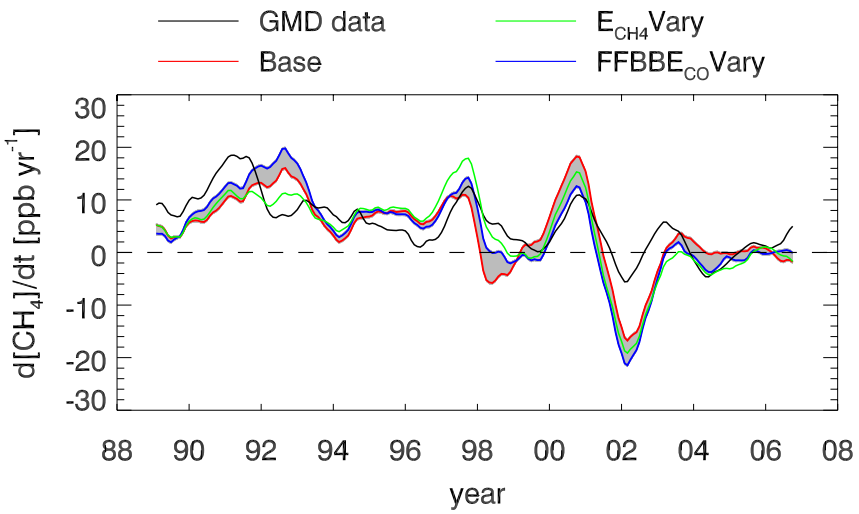

Figure S 5: Same as Figure S 4 but the shaded area is the difference between the *FFBBE$_{CO}$Vary* and *Base* scenarios.

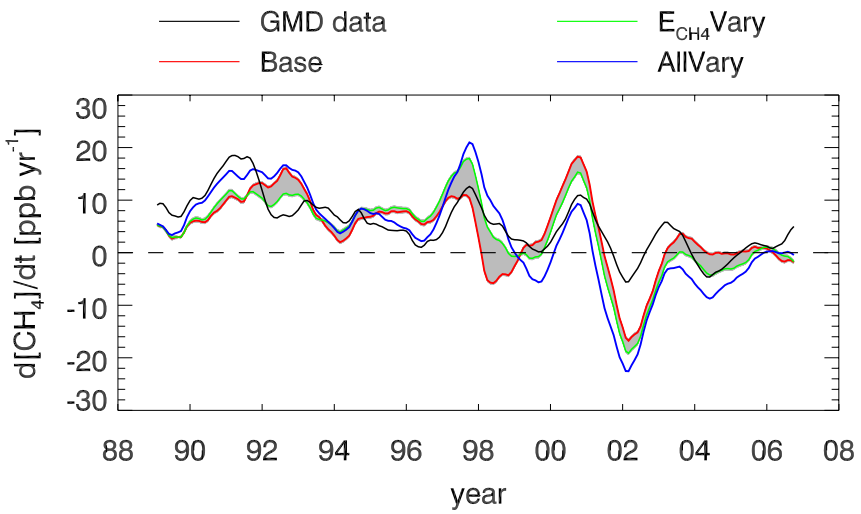

Figure S 6: Same as Figure S 4 but the shaded area is the difference between the *E_{CH4}Vary* and *Base* scenarios.

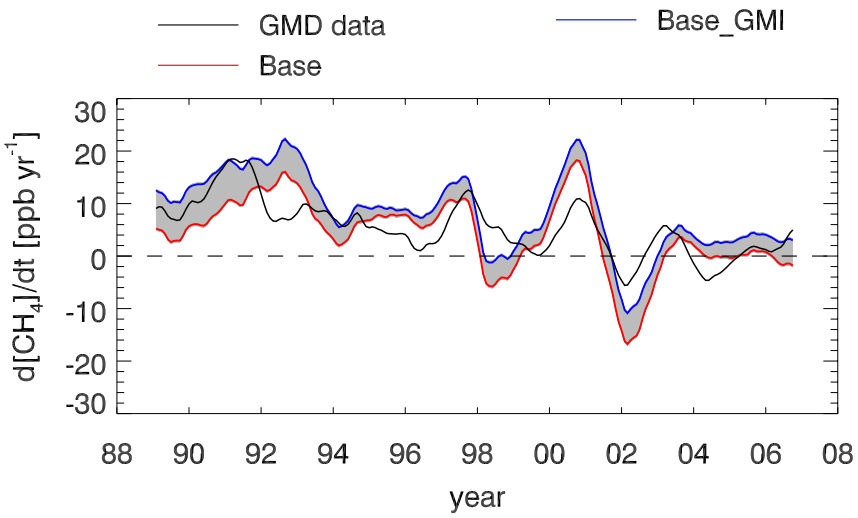

Figure S 7: Same as Figure S 4 but the shaded area is the difference between the *Base_GMI* and *Base* scenarios. The *Base_GMI* scenario is similar to the *Base* scenario, except that OH concentrations are from a full chemistry simulation of the NASA Global Modeling Initiative (GMI) model.

GMD Measurements

Here, we show the comparison of simulated methane by different scenarios (that are not shown in the manuscript) as compared to GMD measurements.

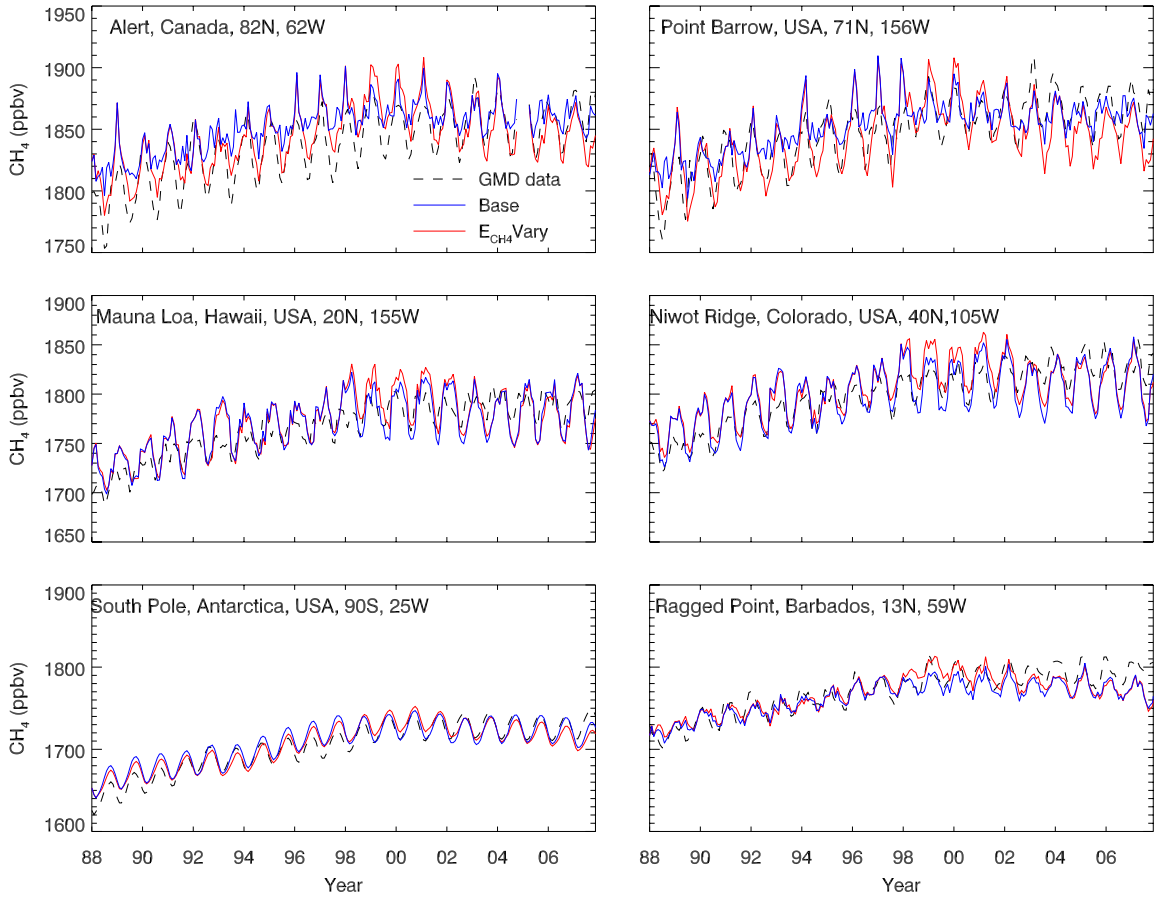

Figure S 8: Monthly methane (ppbv) from the *Base* and $E_{CH4}Vary$ scenarios and observations from six GMD stations.

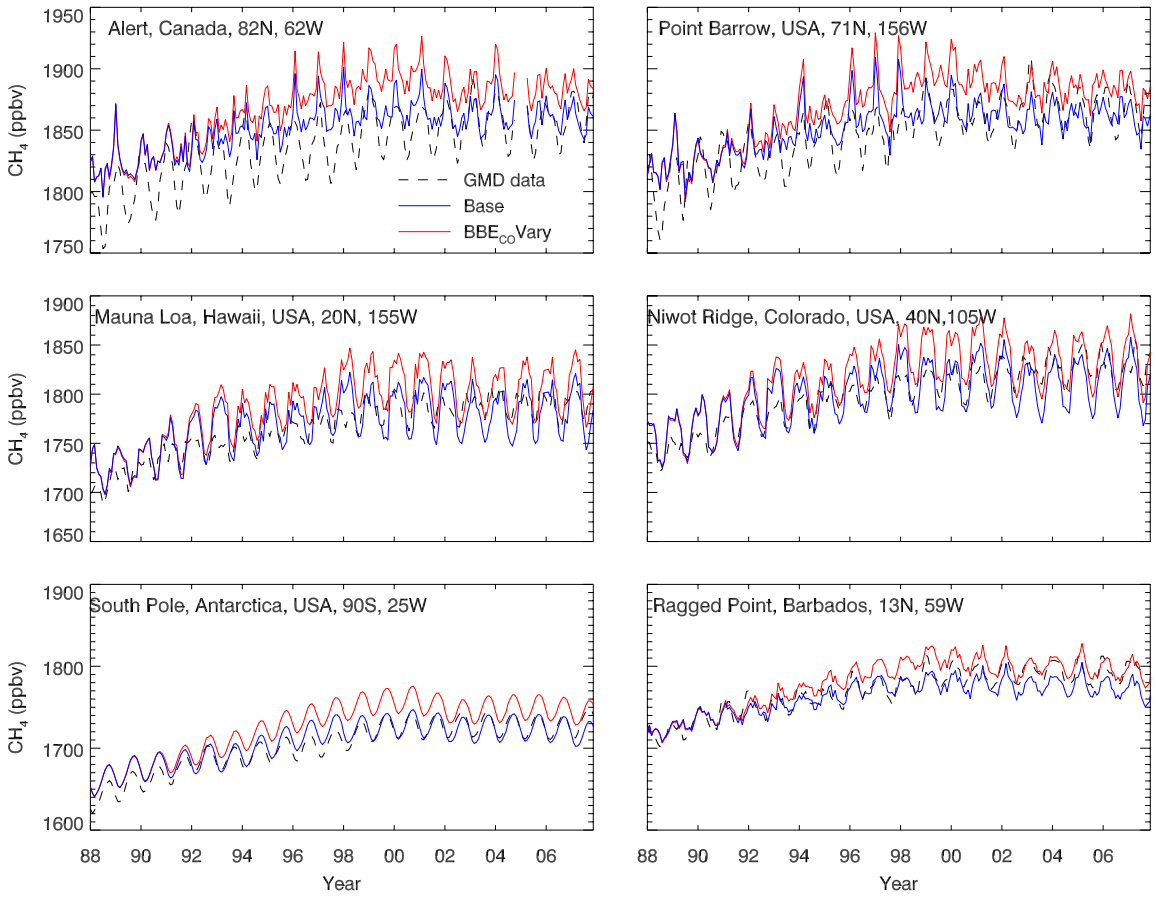

Figure S 9: Monthly methane (ppbv) from the *Base* and *BBE$_{CO}$Vary* scenarios and observations from six GMD stations.

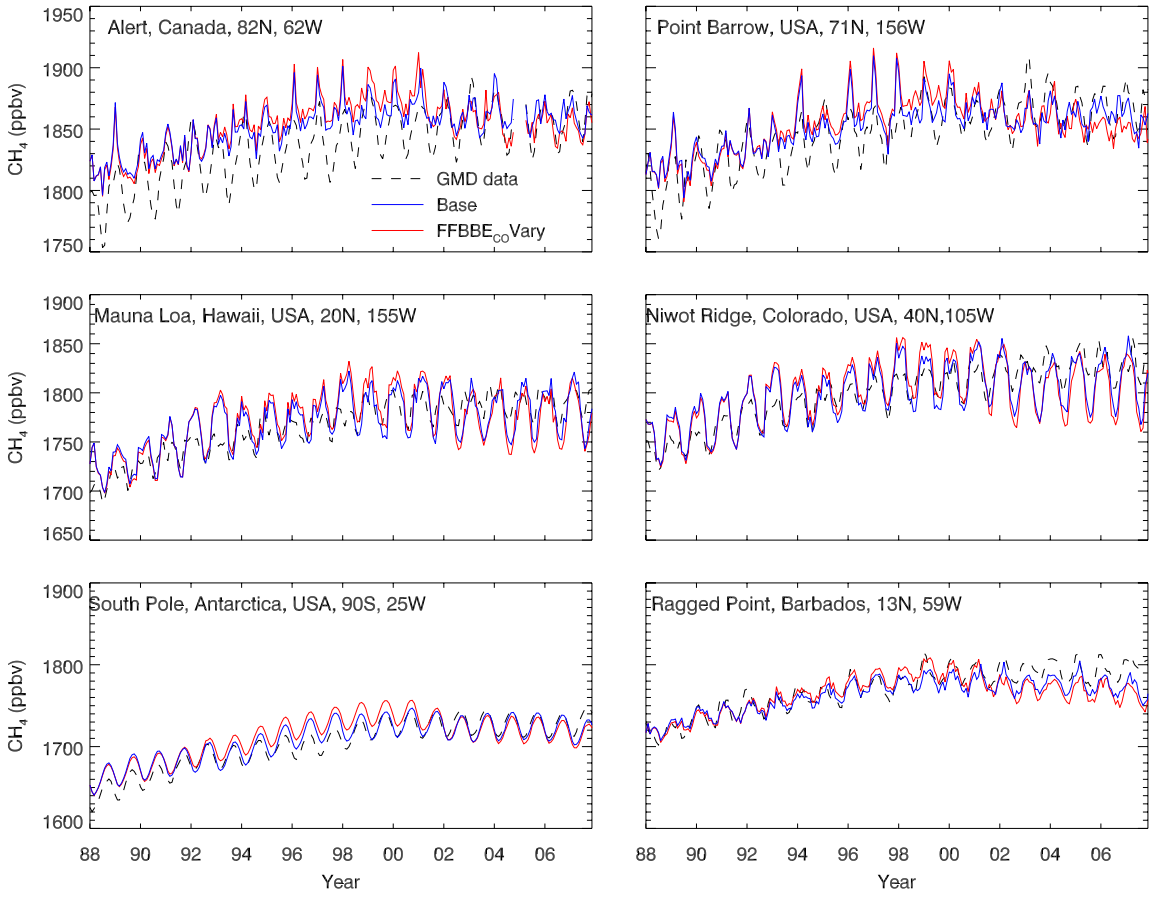

Figure S 10: Monthly methane (ppbv) from the *Base* and *FFBBE$_{CO}$Vary* scenarios and observations from six GMD stations.

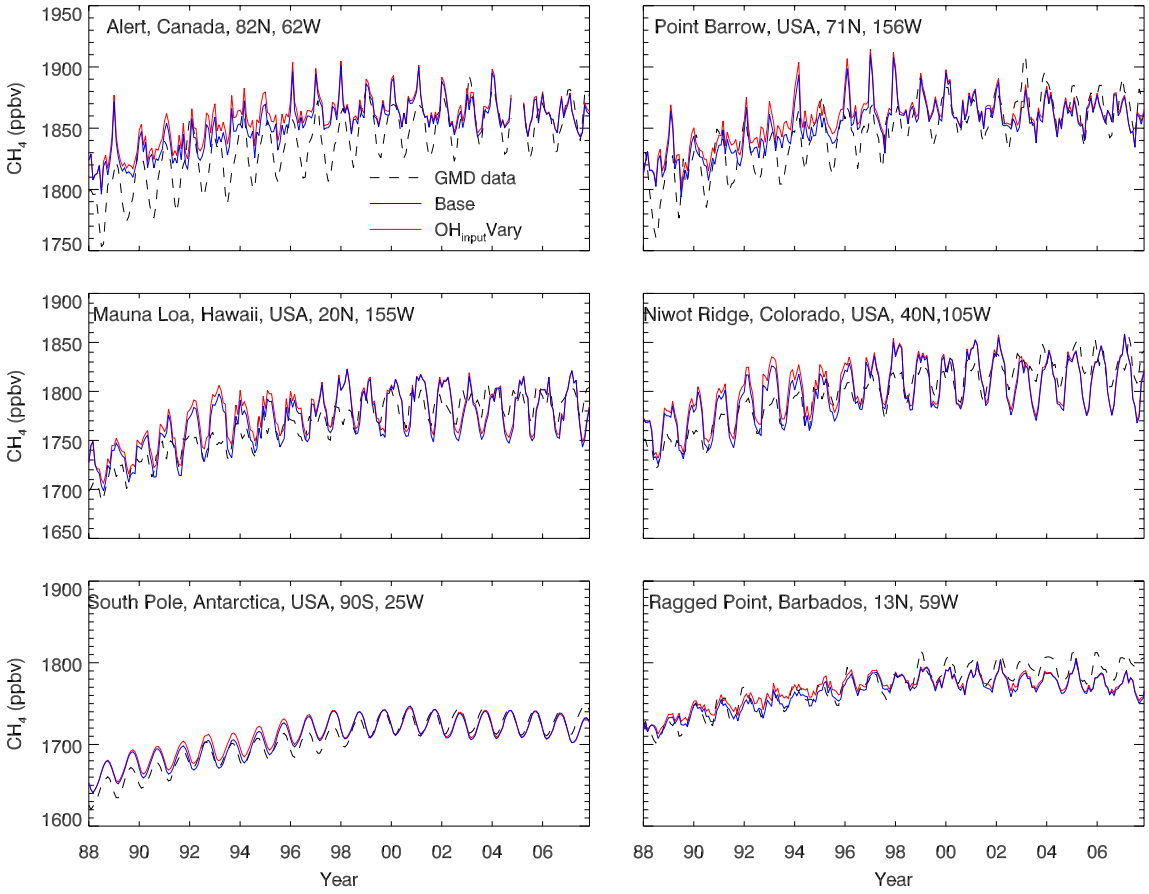

Figure S 11: Monthly methane (ppbv) from the *Base* and $OH_{input}Vary$ scenarios and observations from six GMD stations.

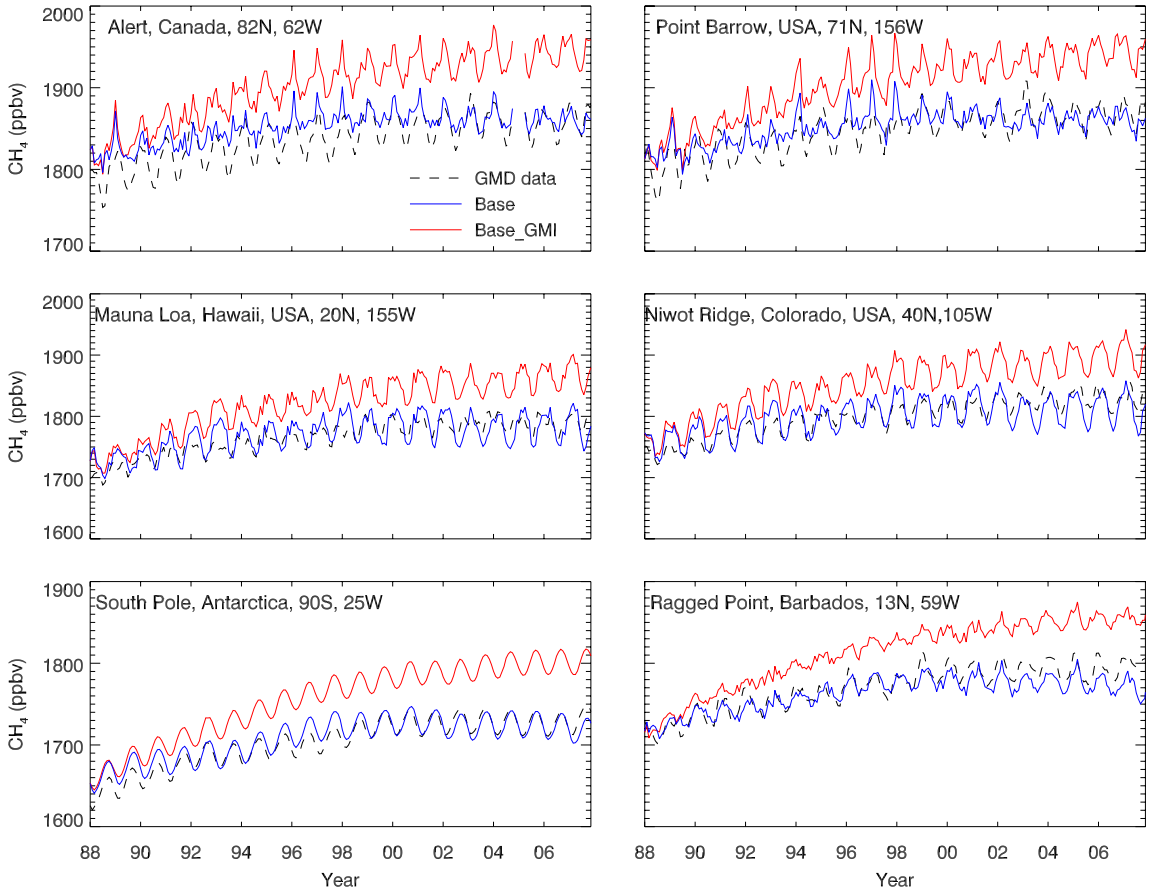

Figure S 12: Monthly methane (ppbv) from the *Base* and *Base_GMI* scenarios and observations from six GMD stations. The *Base_GMI* scenario is similar to the *Base* scenario, except that OH concentrations are from a full chemistry simulation of the GMI model.

2.2 CO

Here, we show additional figures for the comparison of simulated CO as compared to measurements.

GMD measurements

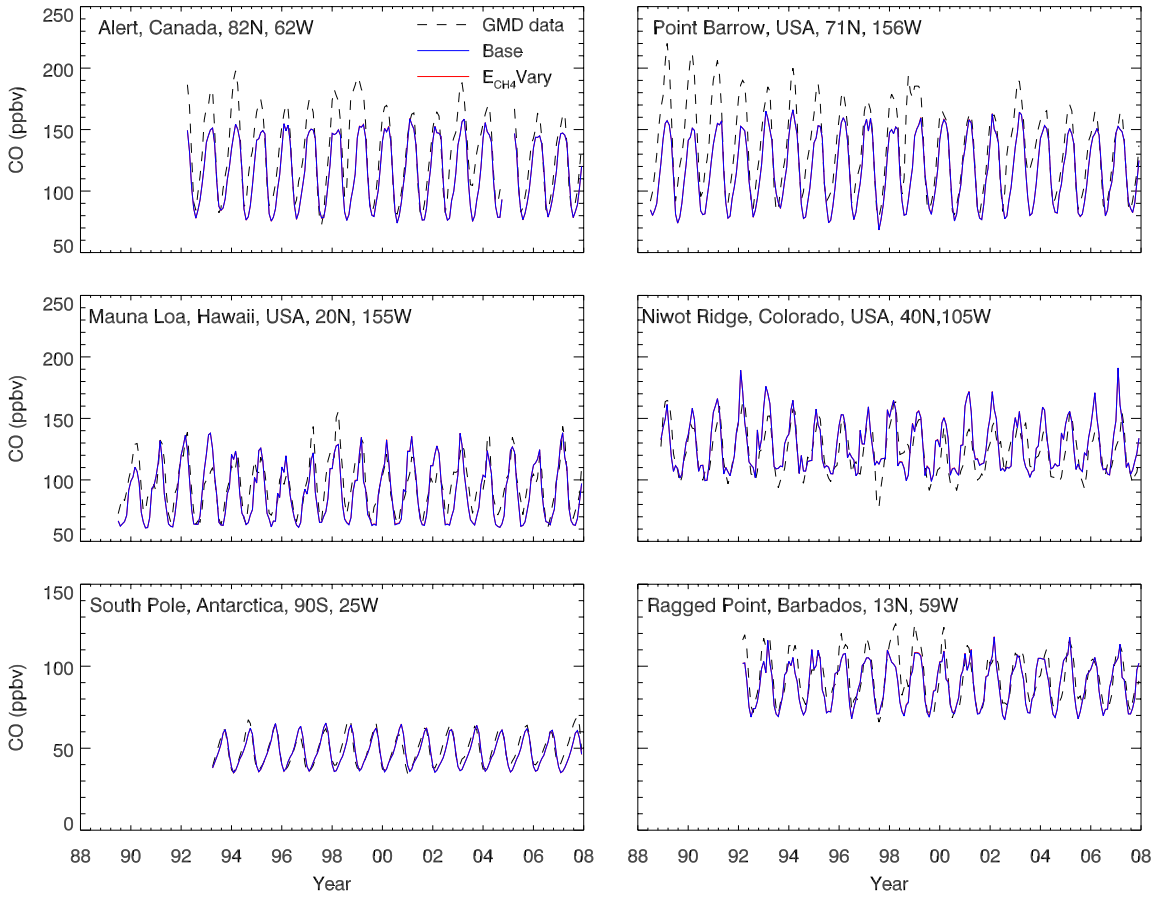

Figure S 13: Measured and simulated monthly near surface CO levels from the *Base* and E_{CH4}*Vary* scenarios.

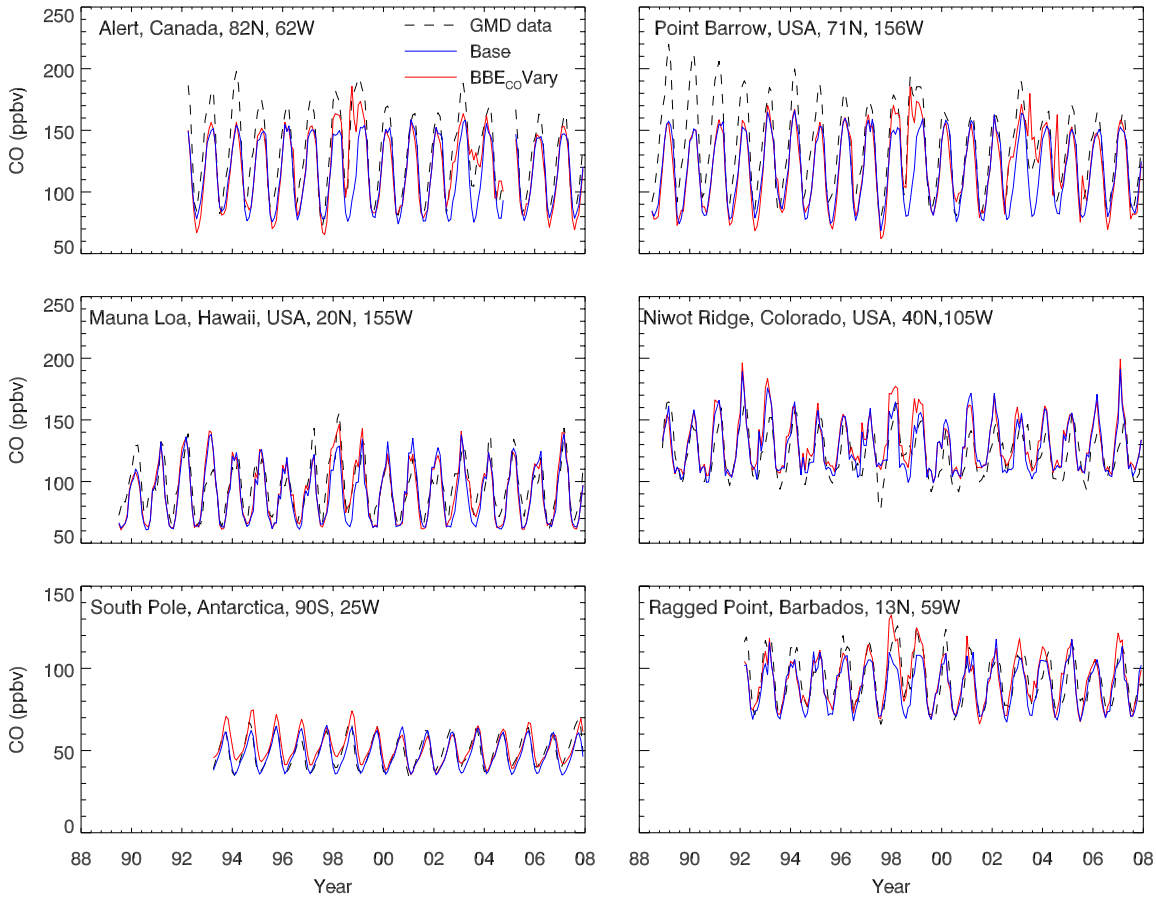

Figure S 14: Measured and simulated monthly near surface CO levels from the *Base* and *BBE$_{CO}$Vary* scenarios.

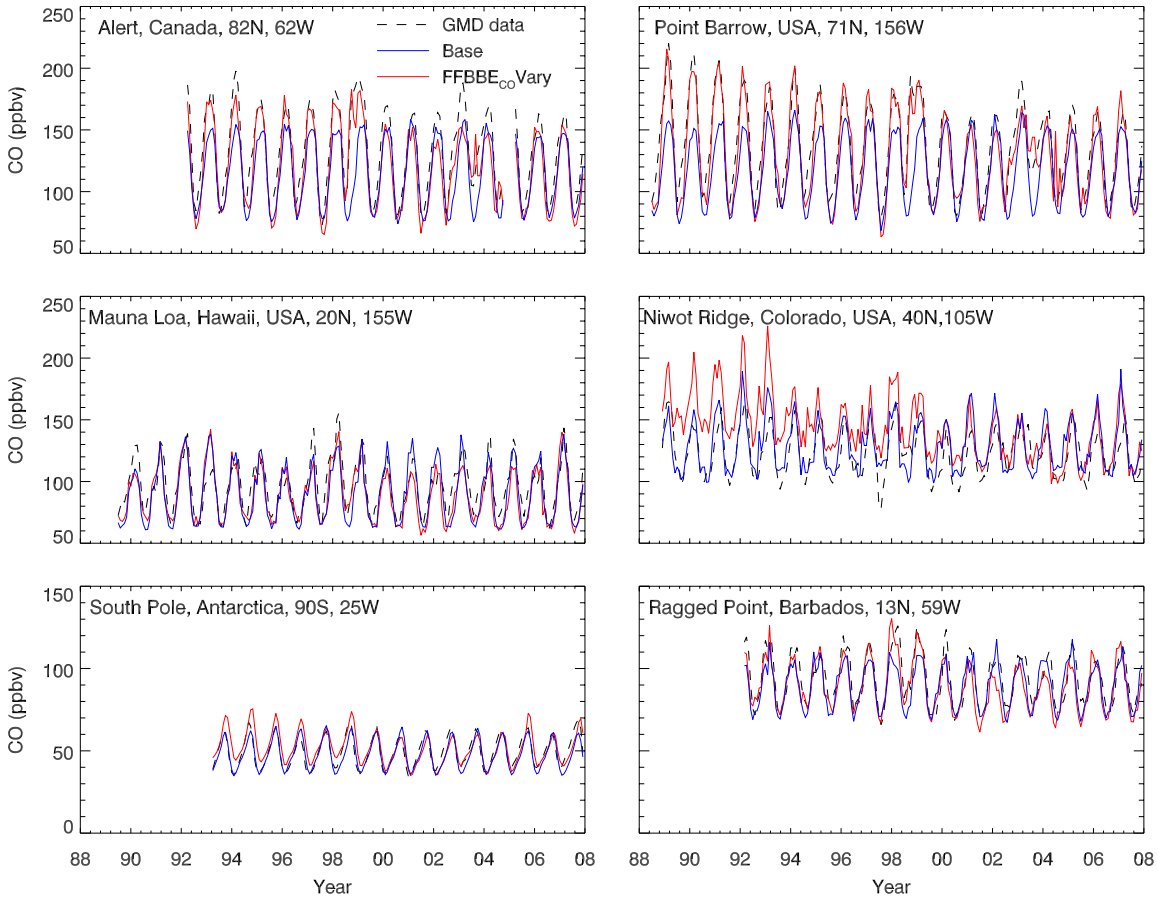

Figure S 15: Measured and simulated monthly near surface CO levels from the *Base* and *FFBBE$_{CO}$Vary* scenarios.

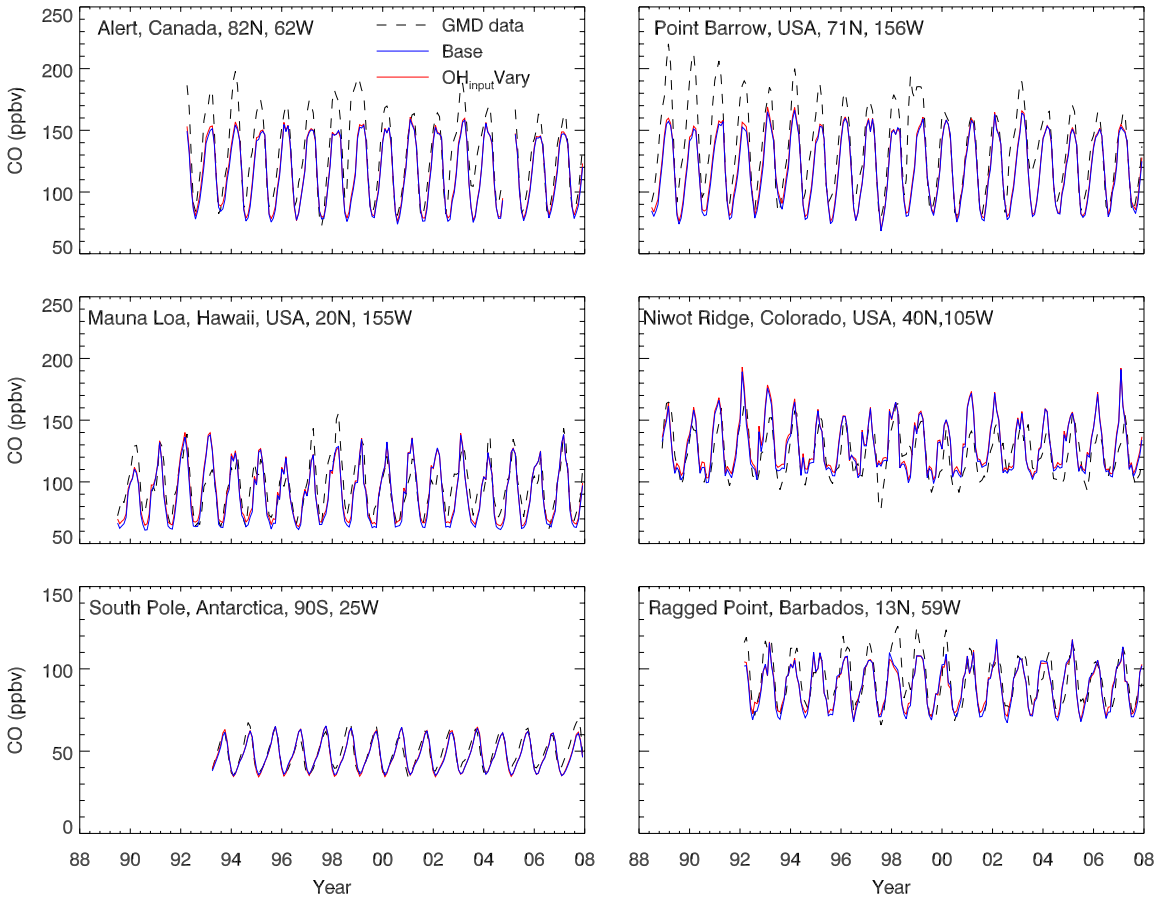

Figure S 16: Measured and simulated monthly near surface CO levels from the *Base* and $OH_{input}Vary$ scenarios.

3. Comparison of simulated OH to full chemistry simulation.

Here, we compare simulated OH from the *Base* and *AllVary* scenario to that of ACCMIP.

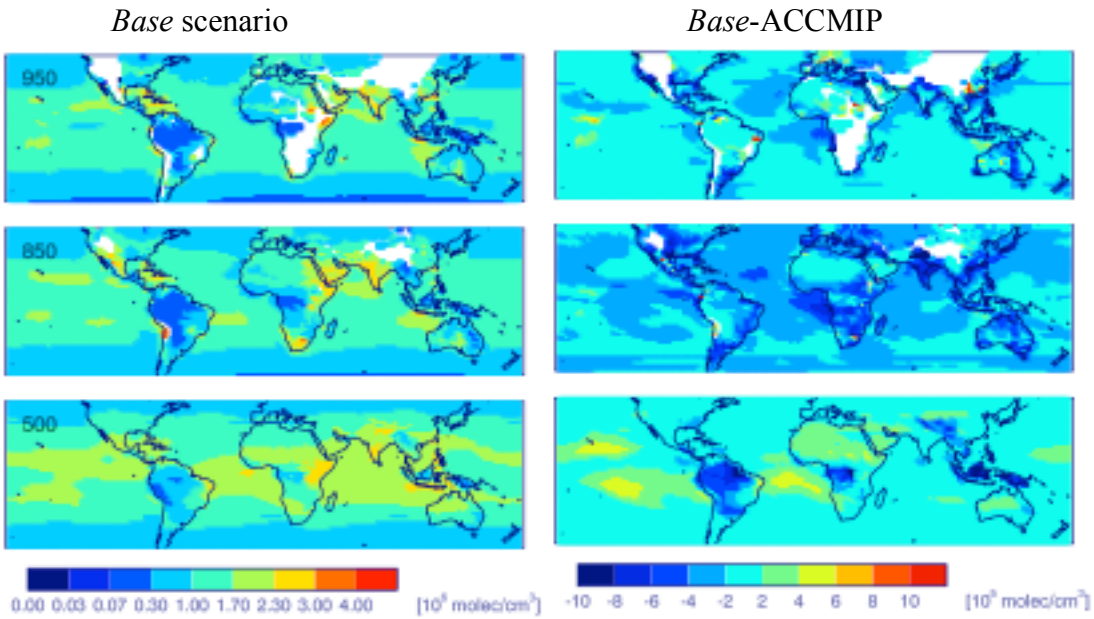

Figure S 17: Annual mean OH (left column; x10^6 molecules/cm^3) from 1999-2007 for the *Base* scenario and their corresponding difference (x10^5 molecules/cm^3) from the full chemistry ACCMIP (GEOS5CCM) simulation (*Base*-ACCMIP, right panels) at 950, 850 and 500 mb (from top to bottom). White gaps indicate no model output at that pressure level.

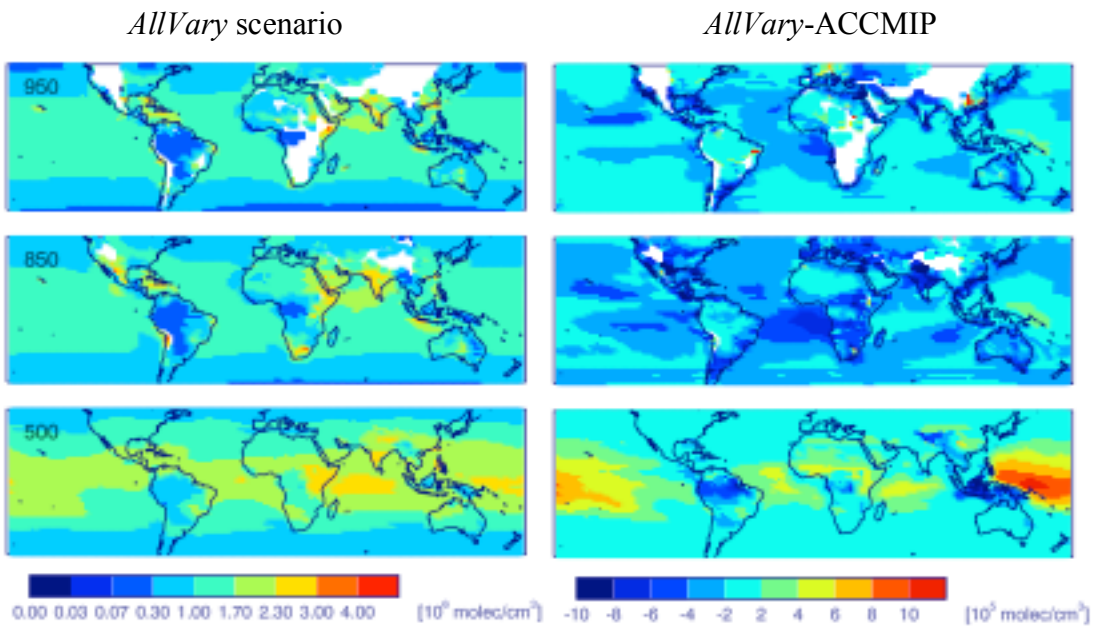

Figure S 18: Annual mean OH (left column, 10^6 molecules/cm^3) from 1999-2007 for the *AllVary* scenario and the corresponding difference (10^5 molecules/cm^3) from the full chemistry ACCMIP simulations (*AllVary*-ACCMIP, right column) at 950, 850 and 500 mb (from up to bottom).

4. Differences in the spatial distribution of methane, CO and OH:

Here, we show the influence of different scenarios on the spatial distribution of tropospheric methane, CO and OH.

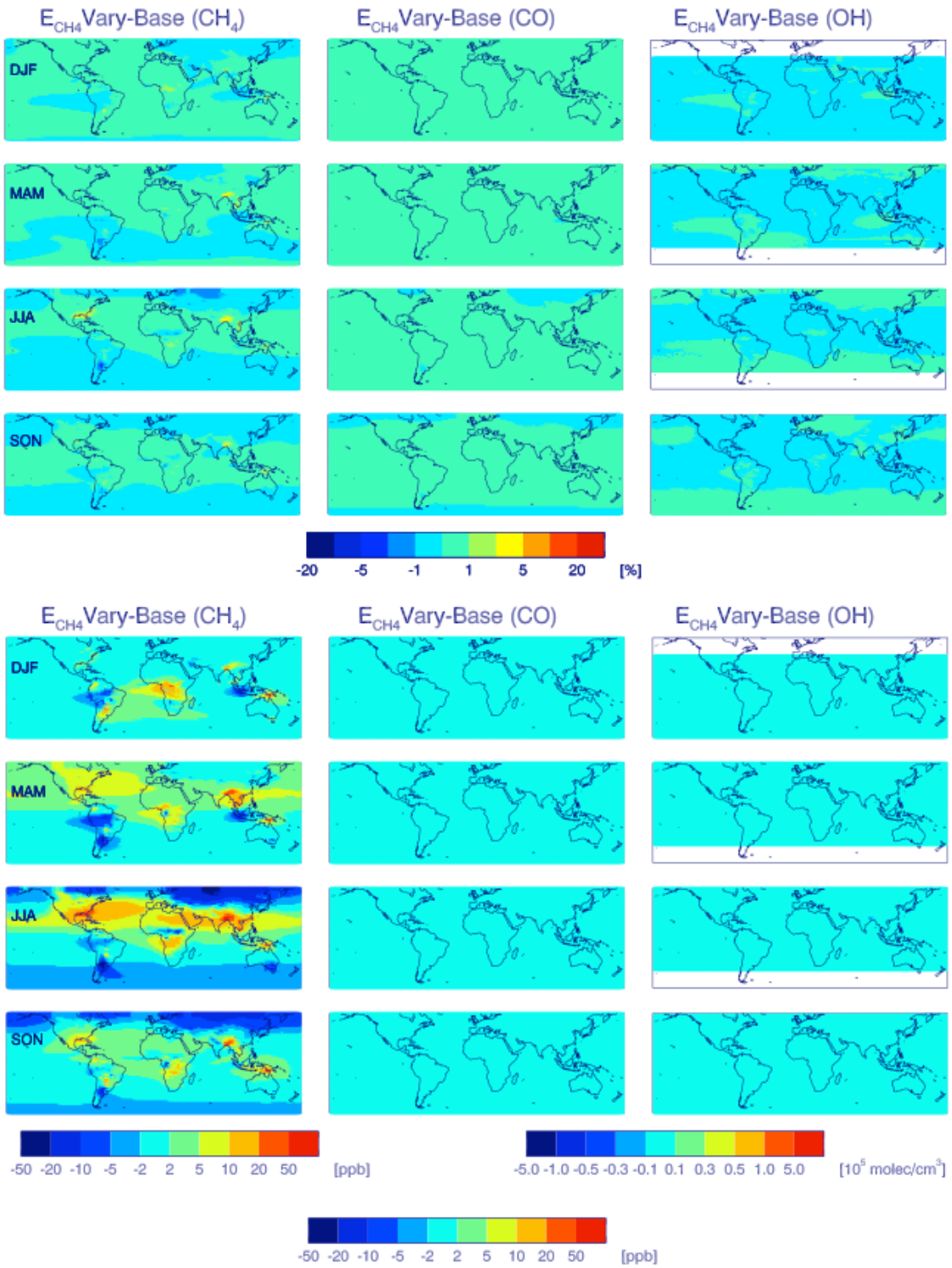

Figure S 19: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm^3) between the *E_{CH4}Vary* and *Base* scenarios.

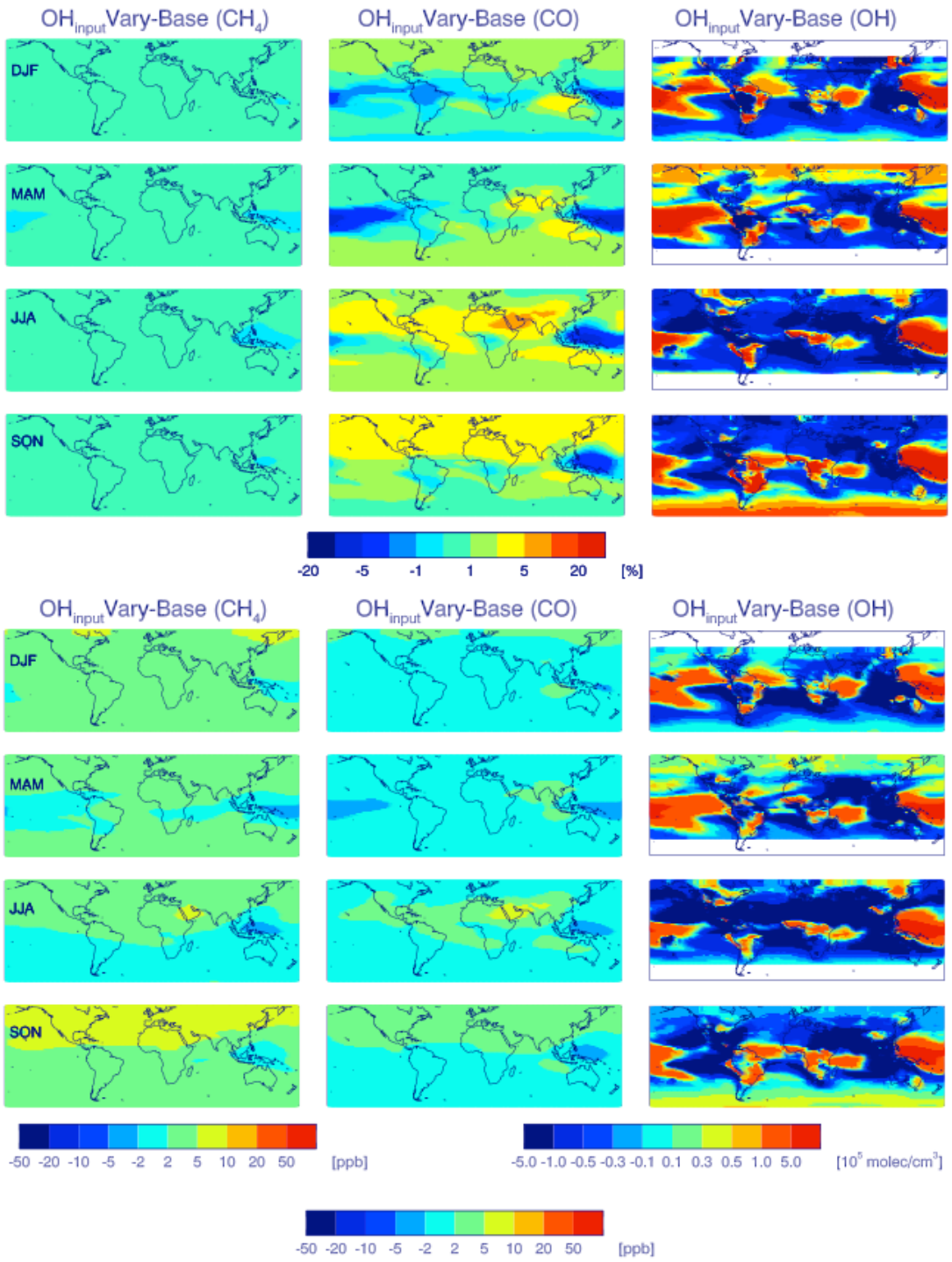

Figure S 20: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm^3) between the *OH$_{input}$Vary* and *Base* scenarios.

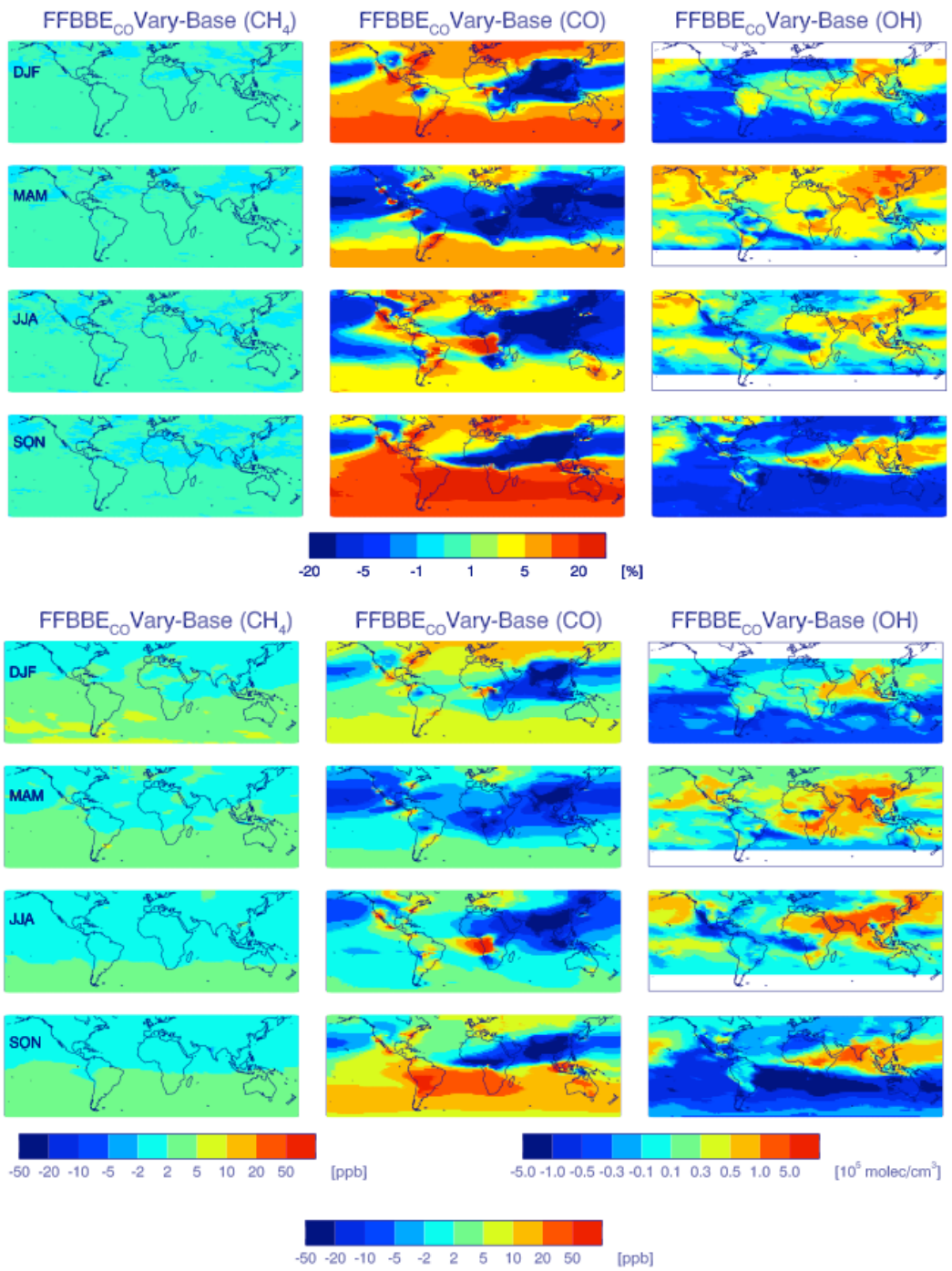

Figure S 21: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm3) between the *FFBBE$_{CO}$Vary* and *Base* scenarios.

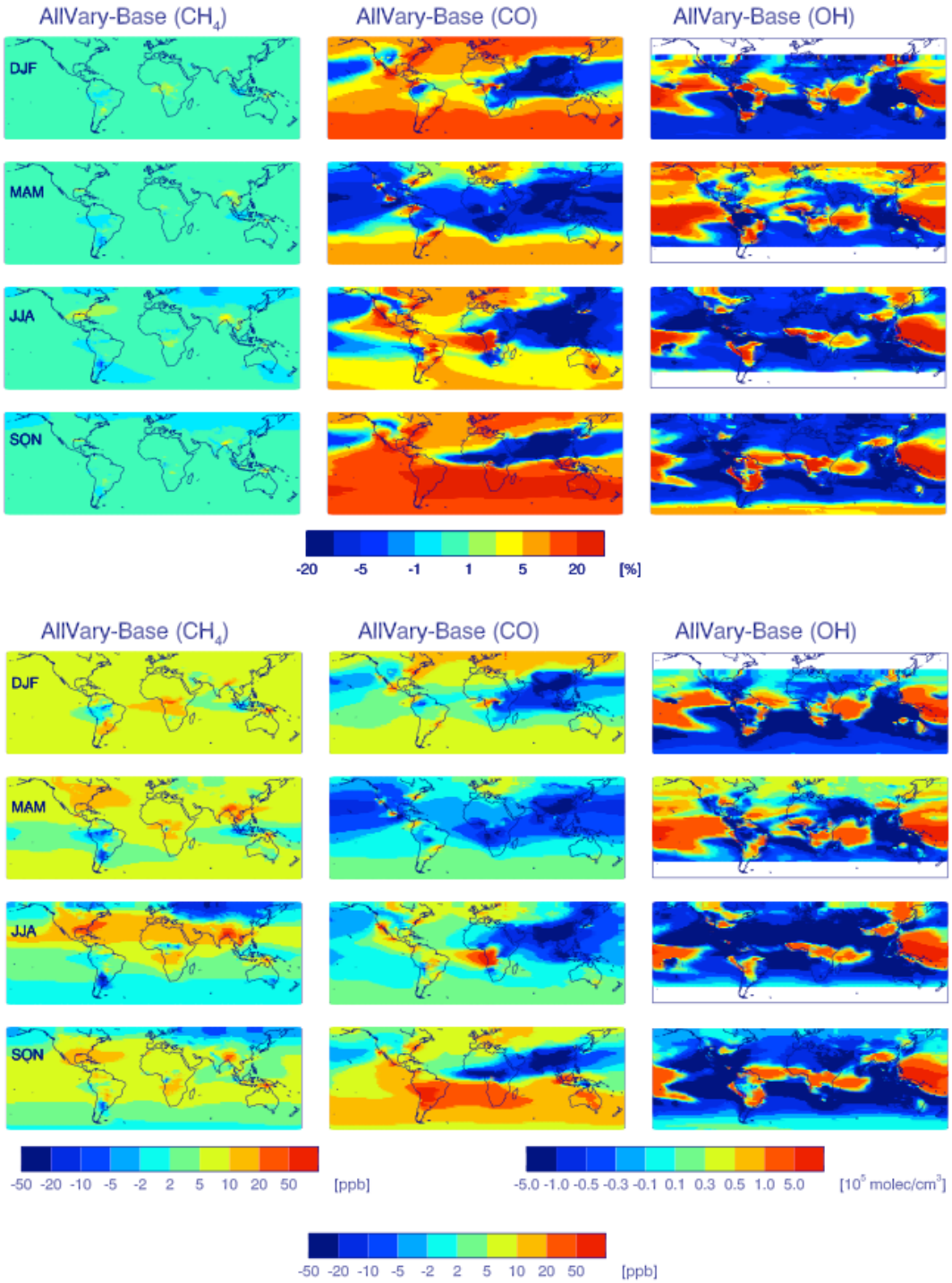

Figure S 22: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH ($x10^5$ molecules/cm^3) between the *AllVary* and *Base* scenarios.

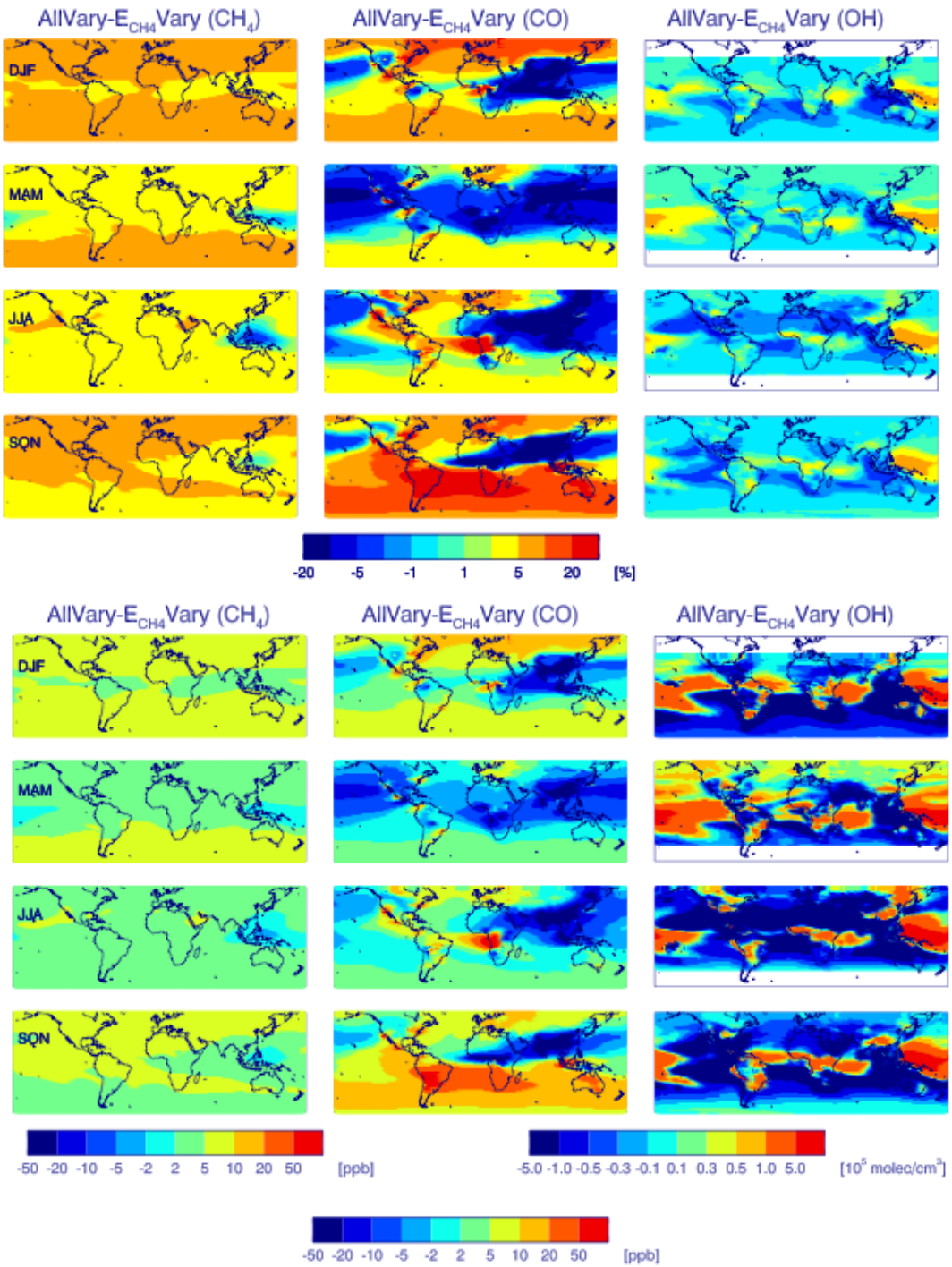

Figure S 23: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm^3) between the *AllVary* and E_{CH4}*Vary* scenarios.