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Supplement of

The description and validation of a computationally-Efficient CH_4-CO-OH (ECCOHv1.01) chemistry module for 3-D model applications

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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 (see Patra et al., 2011).

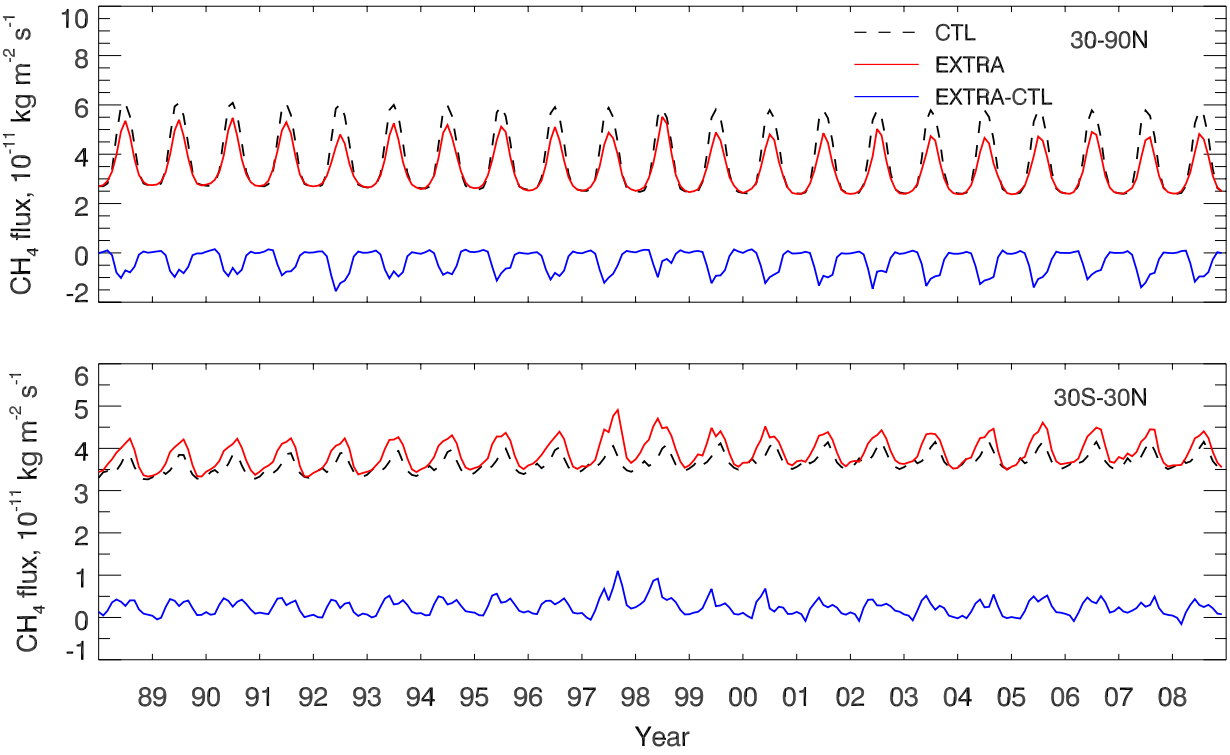

Figure S 1: Monthly methane emissions ($x10^{-11}$ $kg/m^2/s$) used in the *Base* and $E_{CH4}Vary$ scenarios.

1.2 CO

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

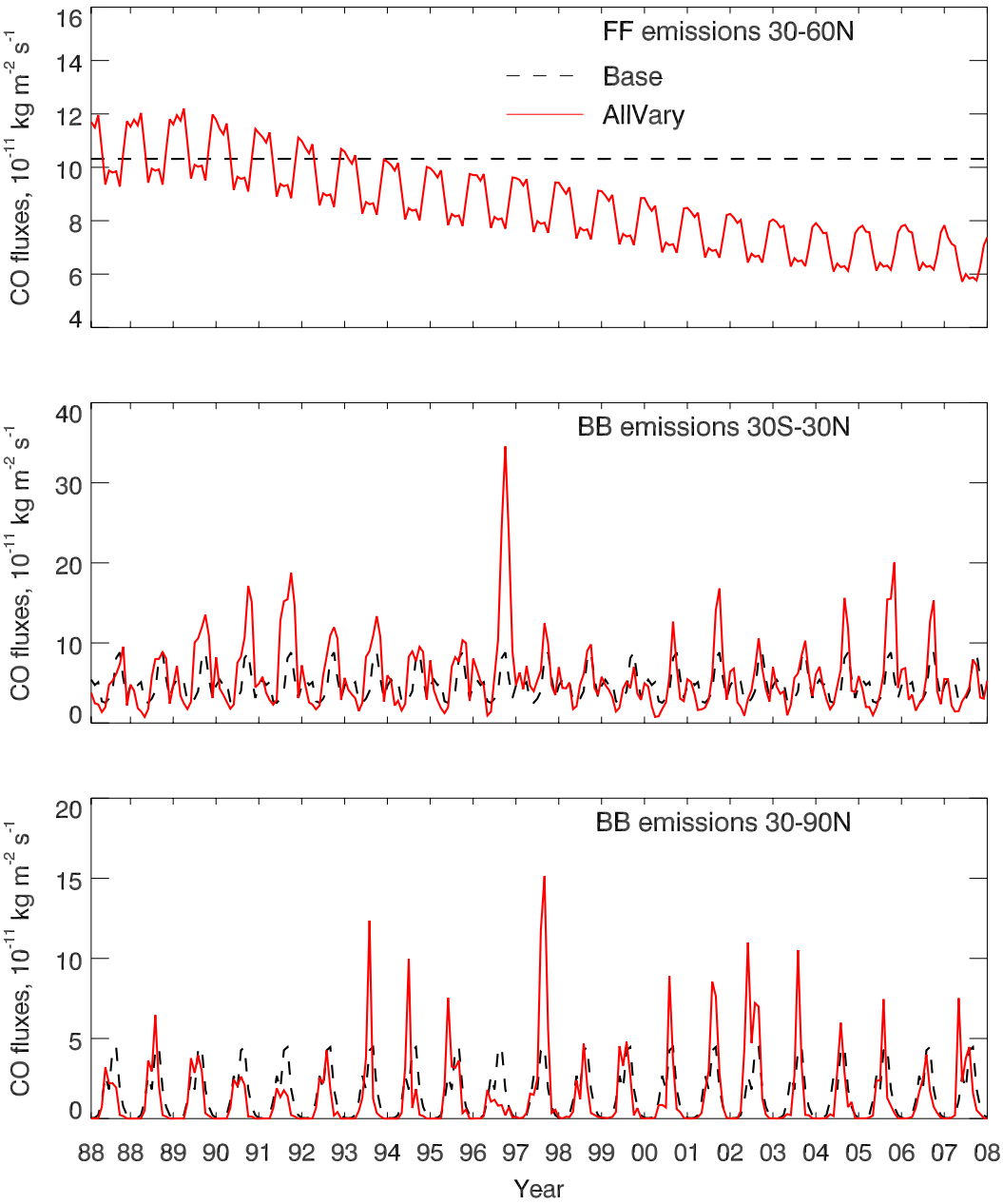

Figure S 2: Monthly CO emissions (x10^{-11} kg/m^2/s) used in the *Base* and *AllVary* scenarios.

2. Comparison to measurements

3.1 Methane

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.

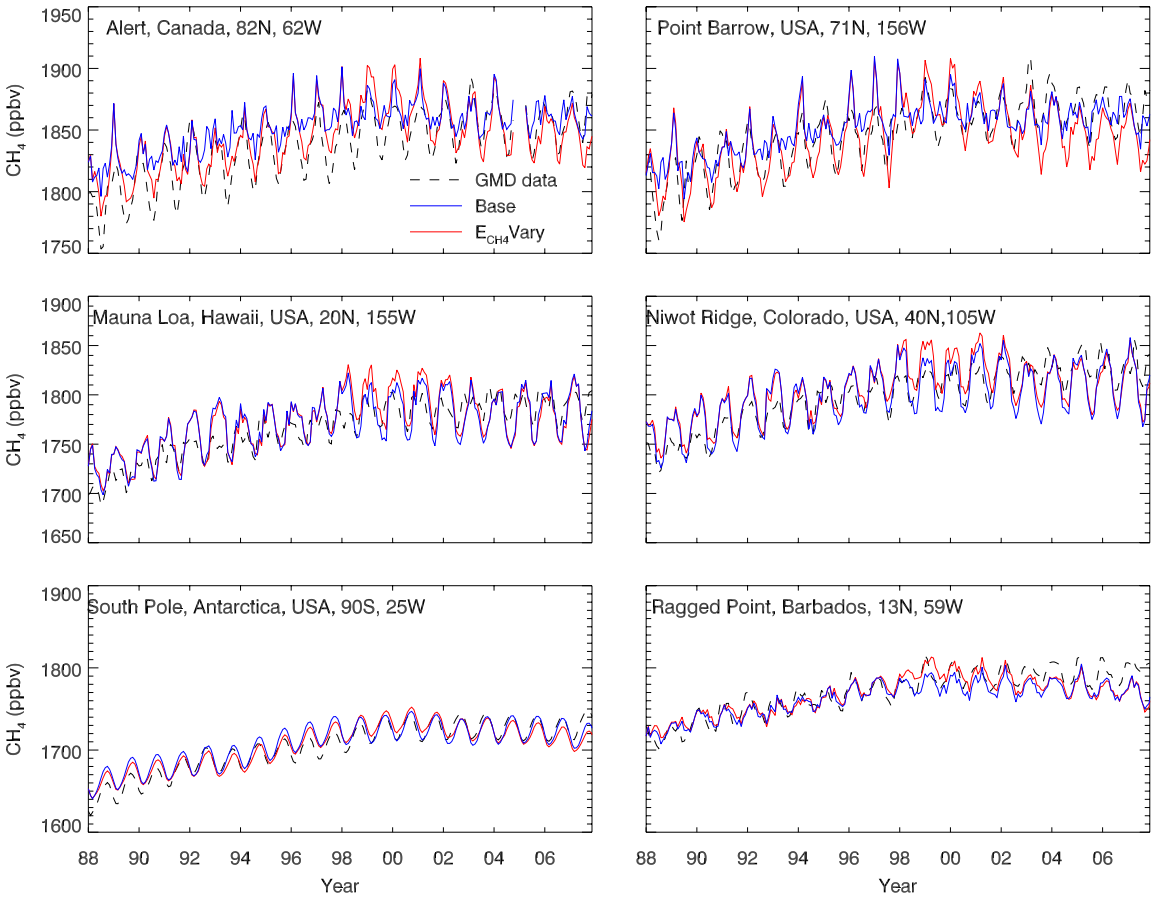

Figure S 3: Monthly methane (ppbv) from the *Base* and $E_{CH4}Vary$ scenarios and observations from six GMD stations.

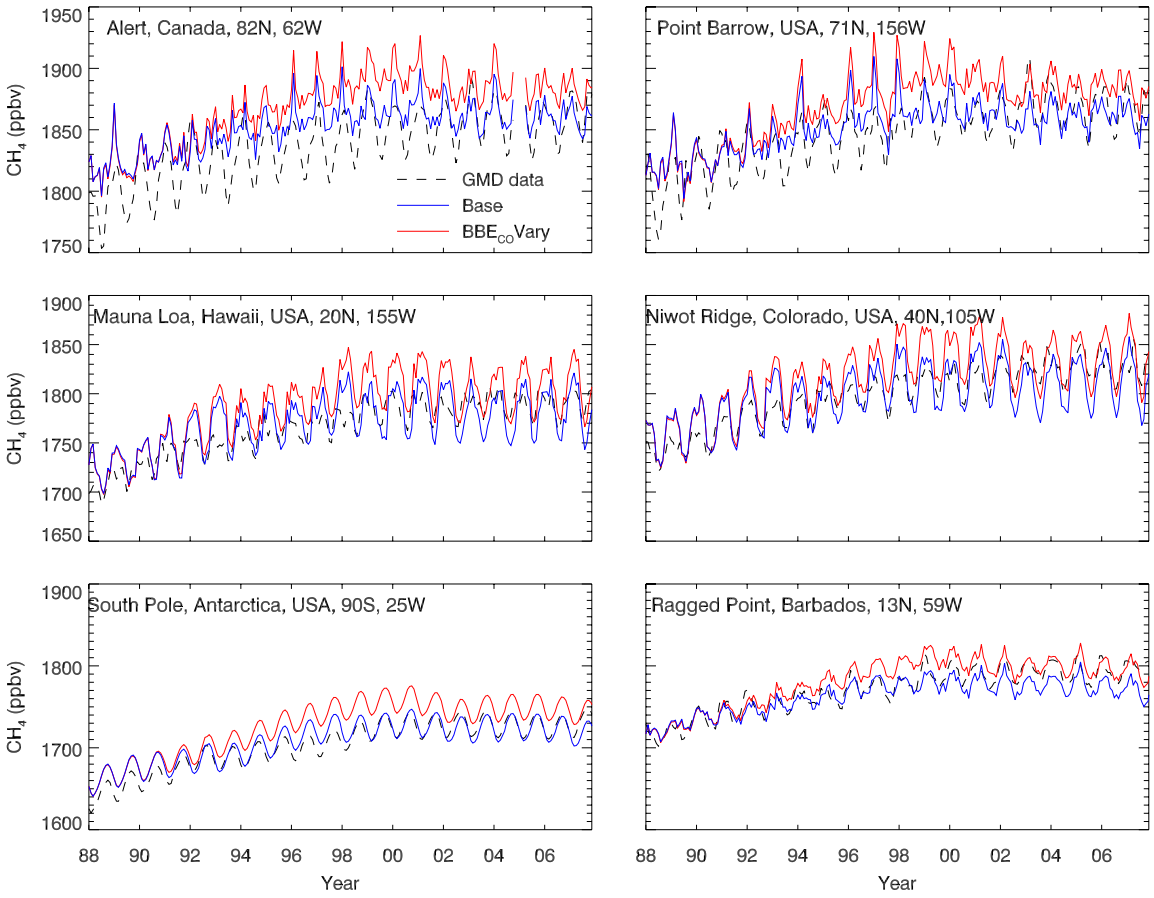

Figure S 4: Monthly methane (ppbv) from the *Base* and *BBE$_{CO}$Vary* scenarios and observations from six GMD stations.

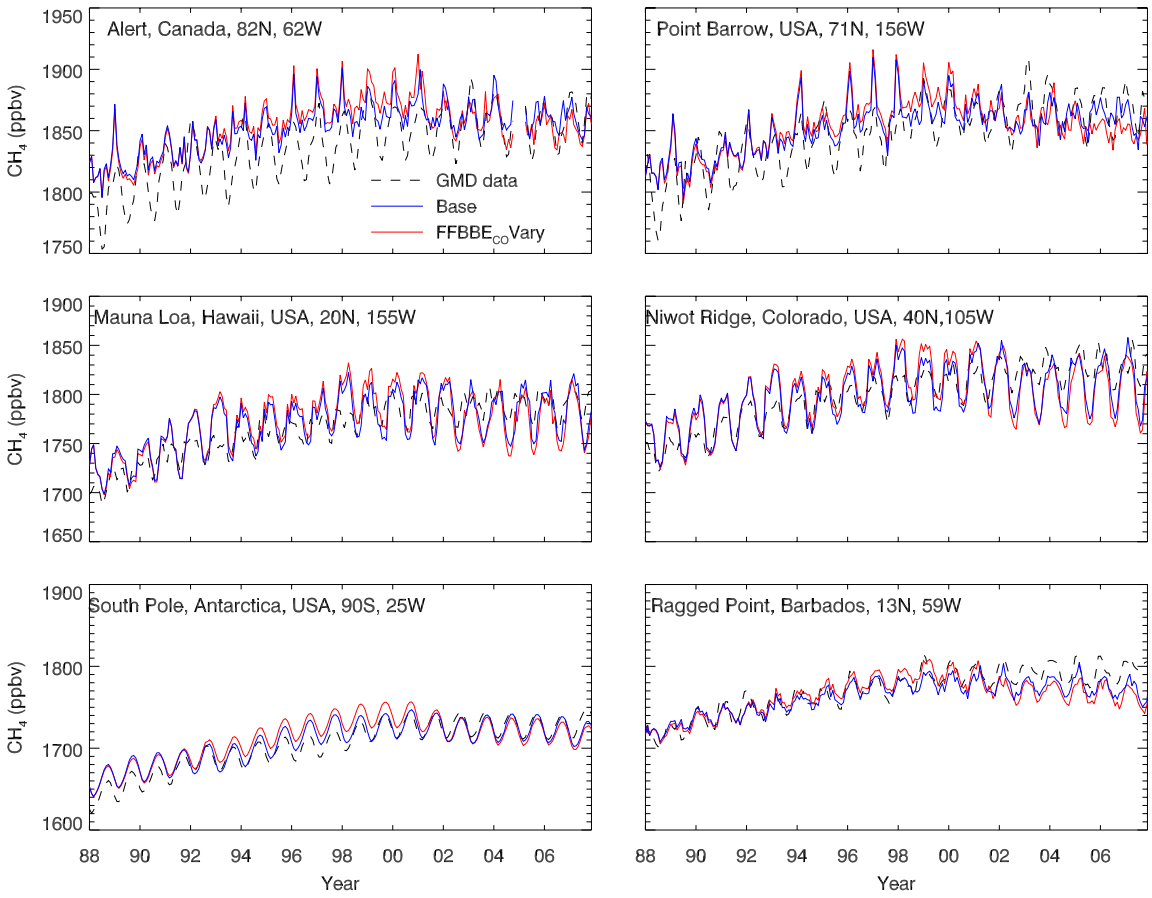

Figure S 5: Monthly methane (ppbv) from the *Base* and *FFBBE$_{CO}$Vary* scenarios and observations from six GMD stations.

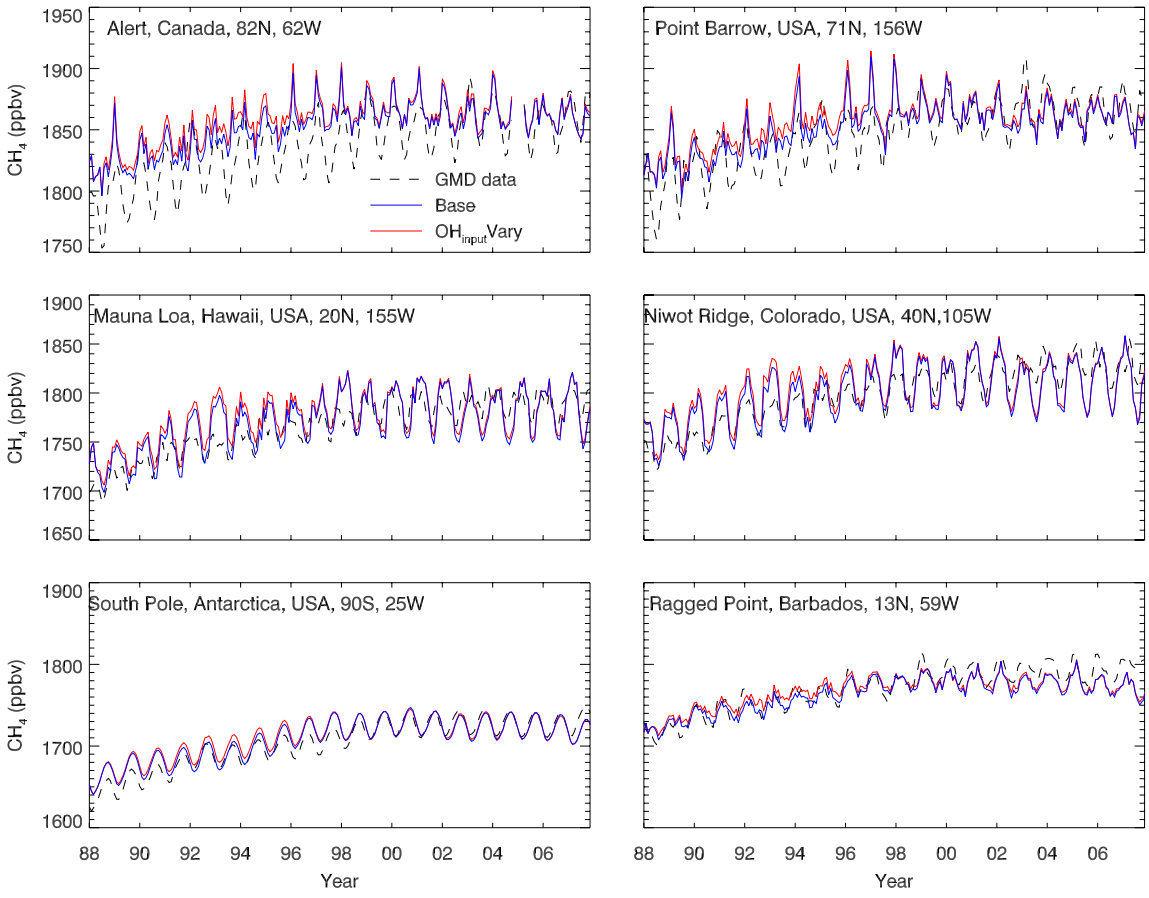

Figure S 6: Monthly methane (ppbv) from the *Base* and *OH$_{input}$Vary* scenarios and observations from six GMD stations.

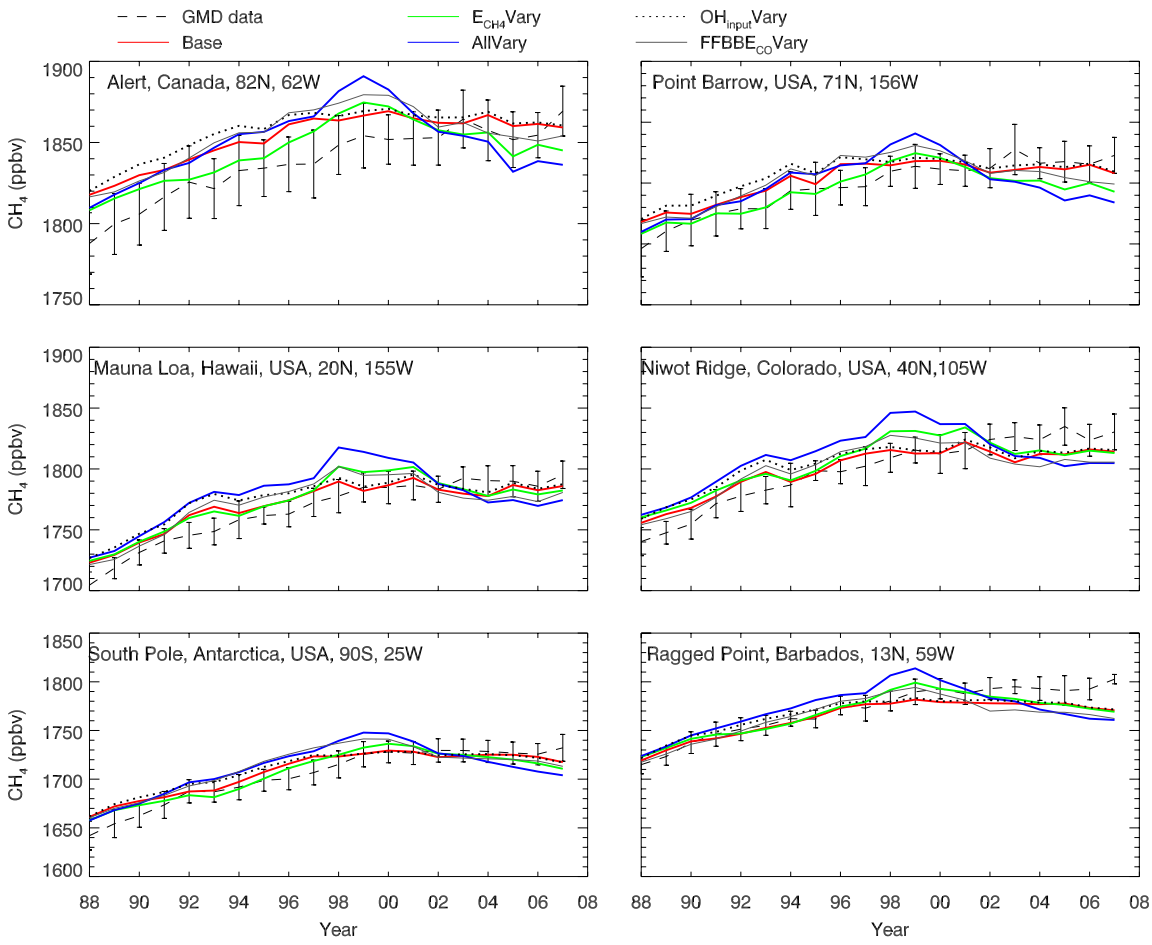

Figure S 7: Annual mean measured and simulated near-surface methane levels by different scenarios. Vertical lines represent the standard deviation of the measured annual mean.

SCIAMACHY comparison:

Here, we show the comparison between simulated (*AllVary*) methane dry column and that from the SCIAMACHY data.

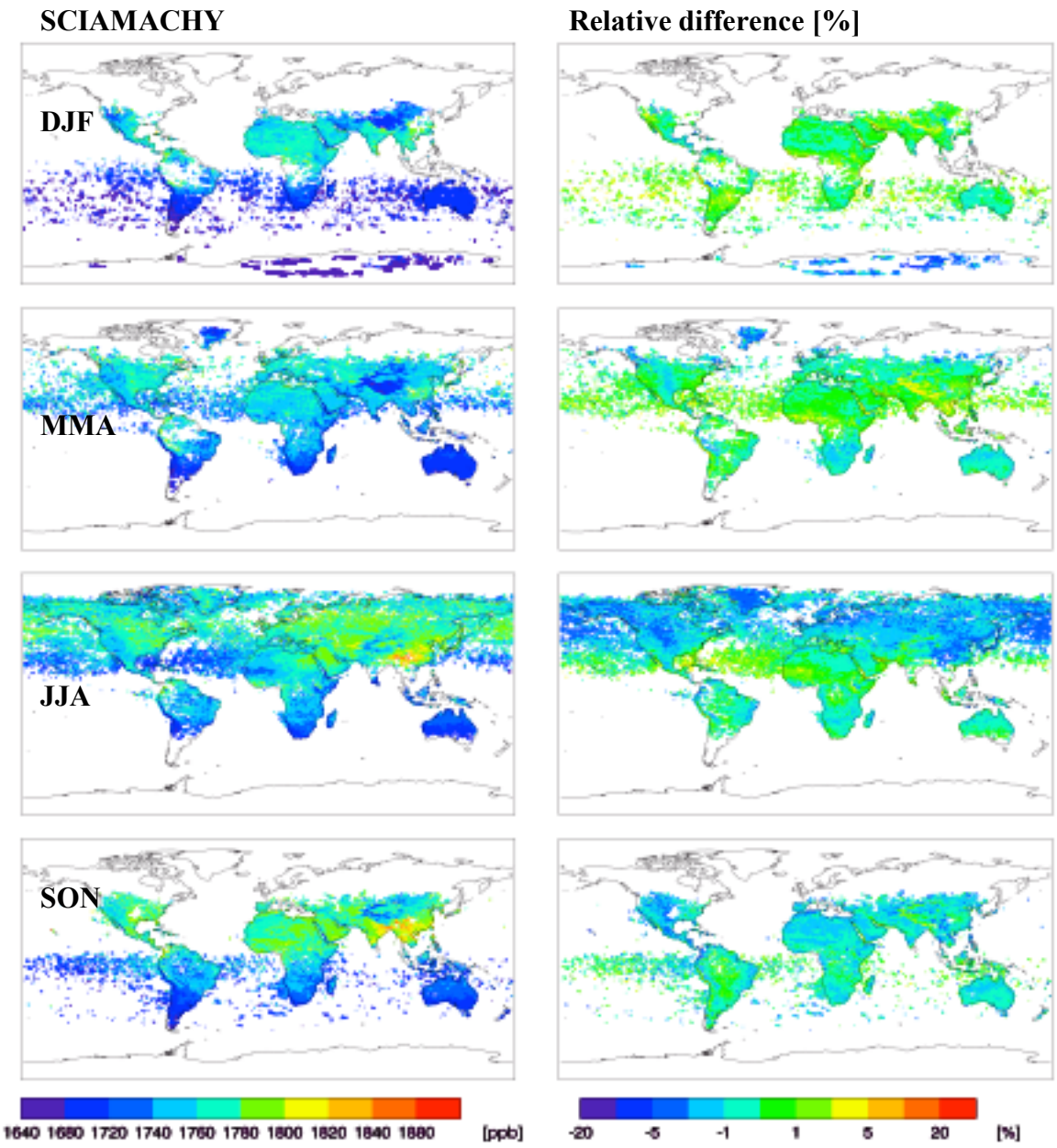

Figure S 8: Seasonal mean (2004) measured SCIAMACHY methane dry column (ppbv, left column) and the relative difference (%, (*AllVary*-SCIAMACHY)/SCIAMACHY, right column).

3.2 CO

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

GMD measurements:

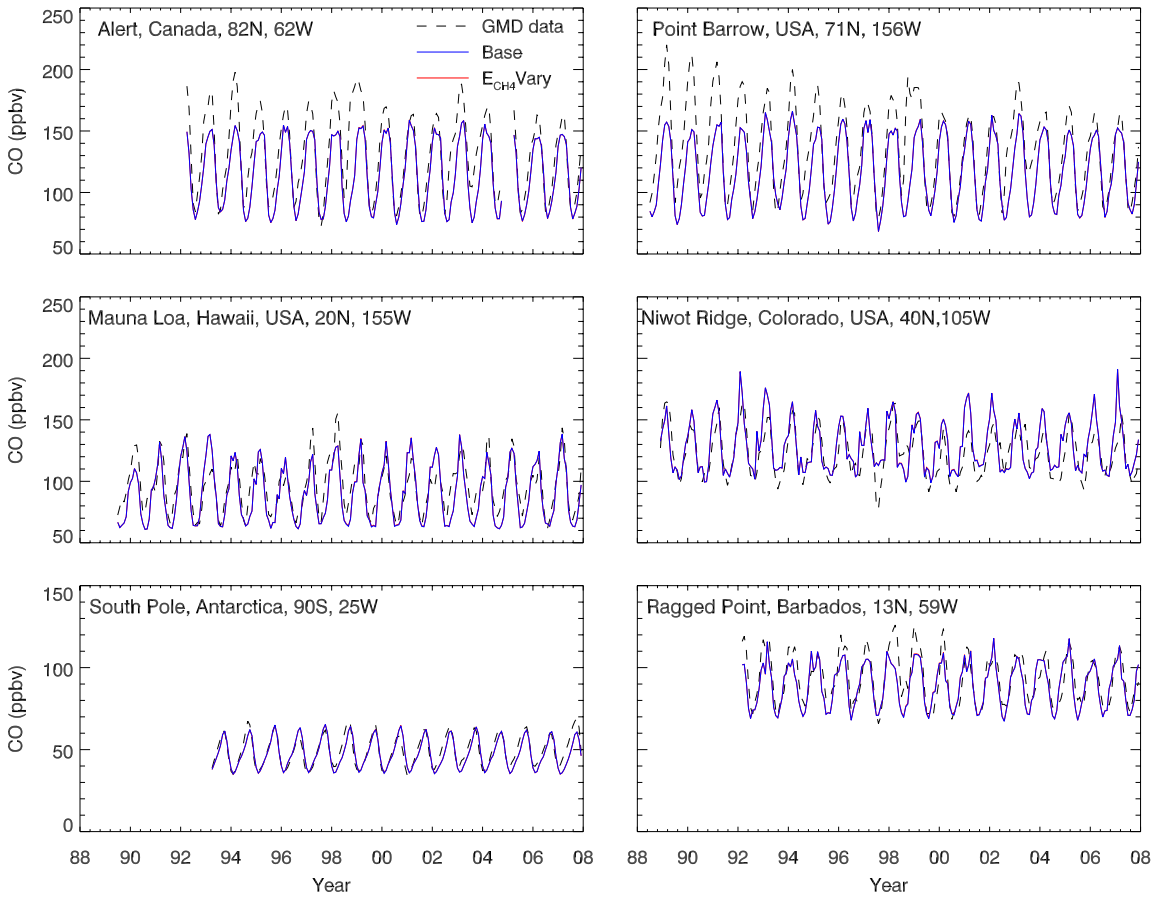

Figure S 9: Measured and simulated monthly near surface CO levels by the *Base* and E_{CH4}*Vary* scenarios.

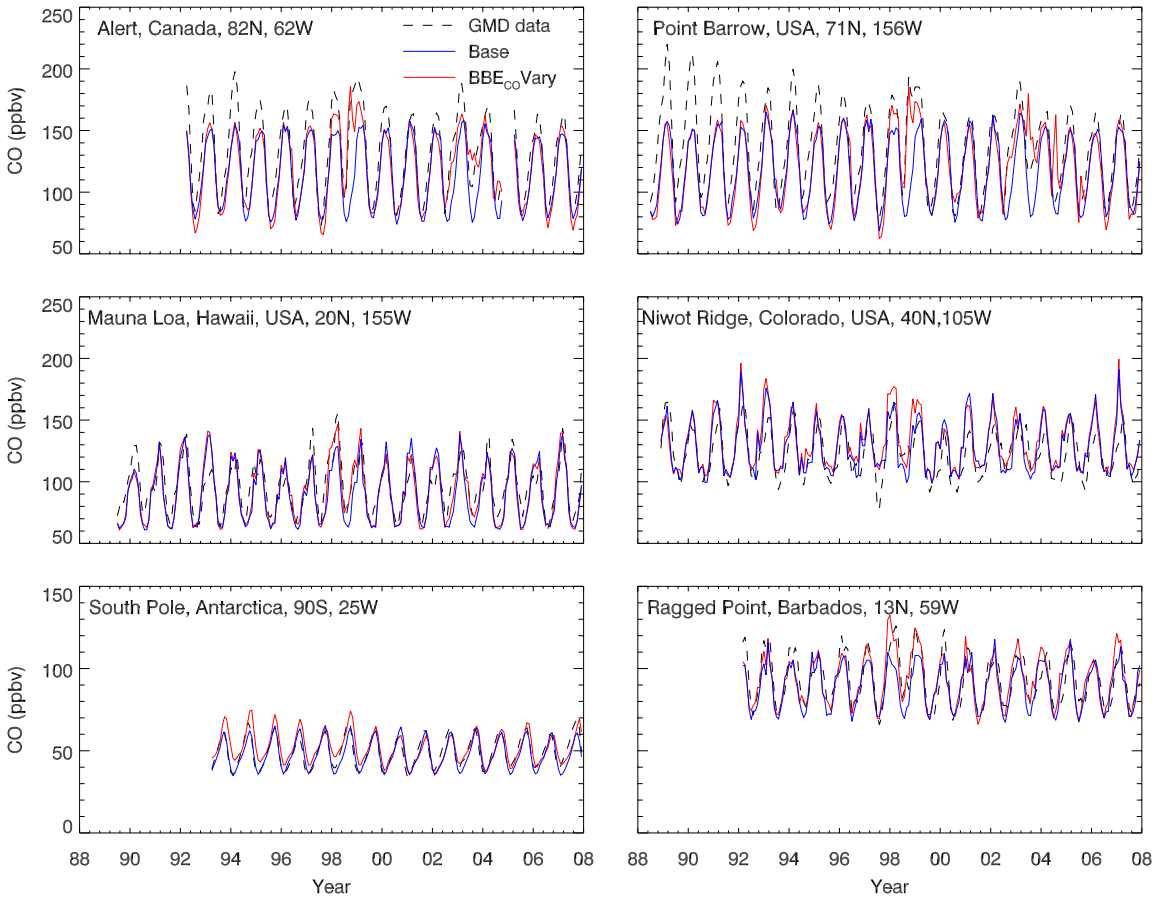

Figure S 10: Measured and simulated monthly near surface CO levels by the *Base* and *BBE$_{CO}$Vary* scenarios.

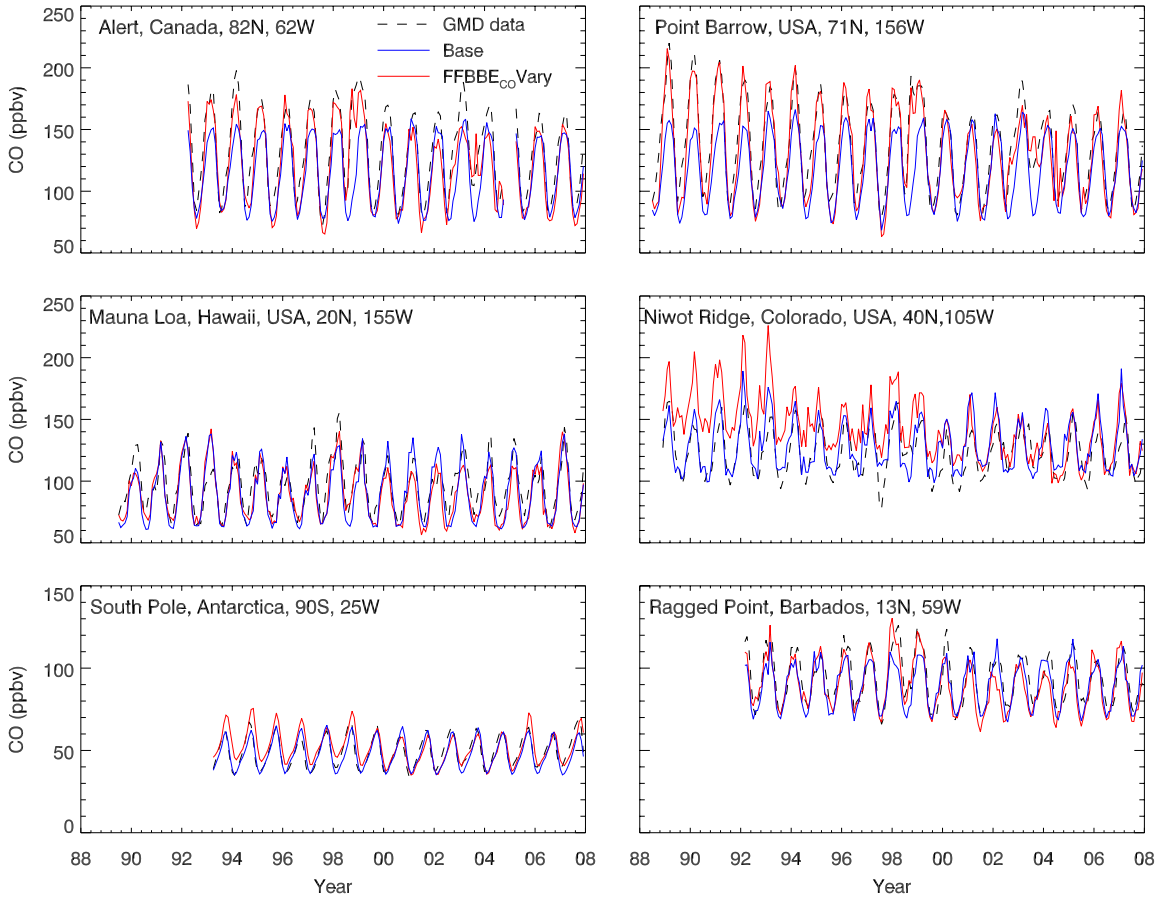

Figure S 11: Measured and simulated monthly near surface CO levels by the *Base* and *FFBBE$_{CO}$Vary* scenarios.

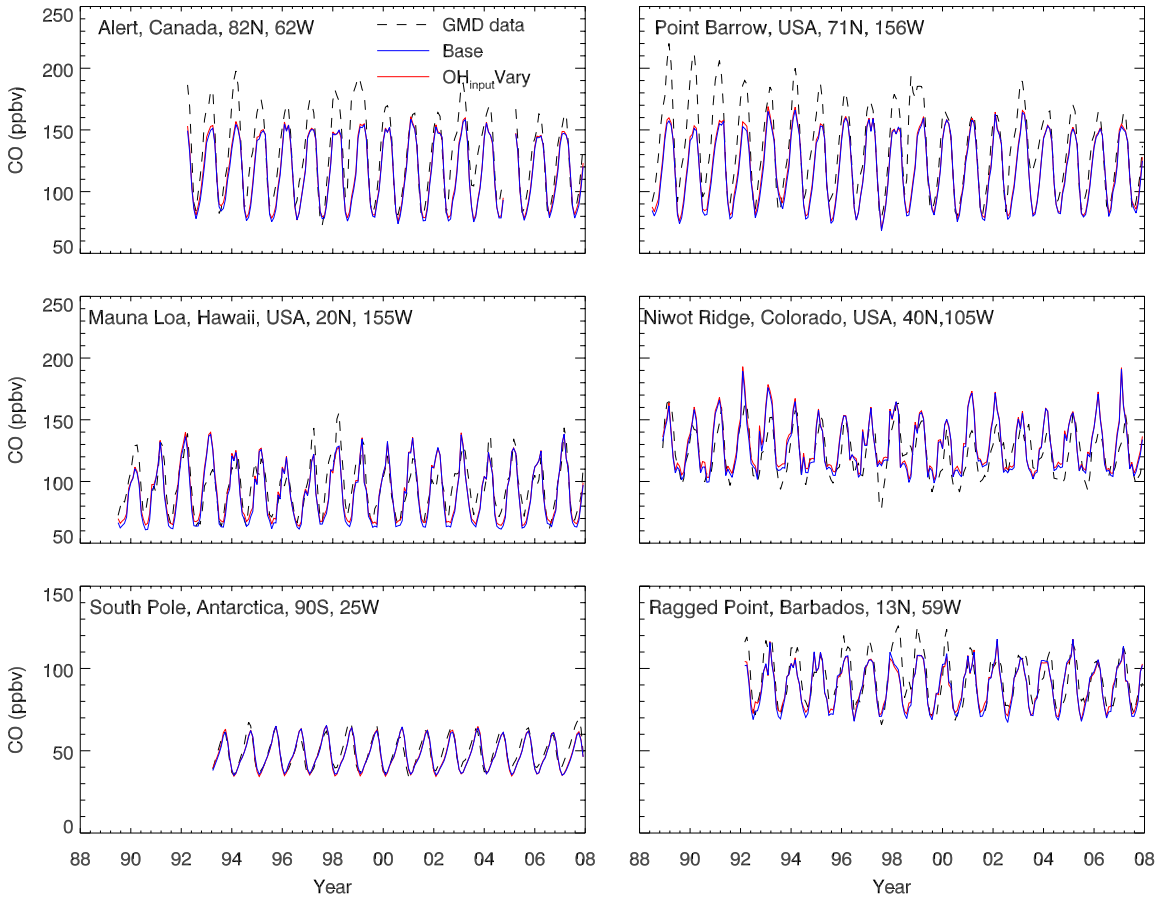

Figure S 12: Measured and simulated monthly near surface CO levels by the *Base* and *OH$_{input}$Vary* scenarios.

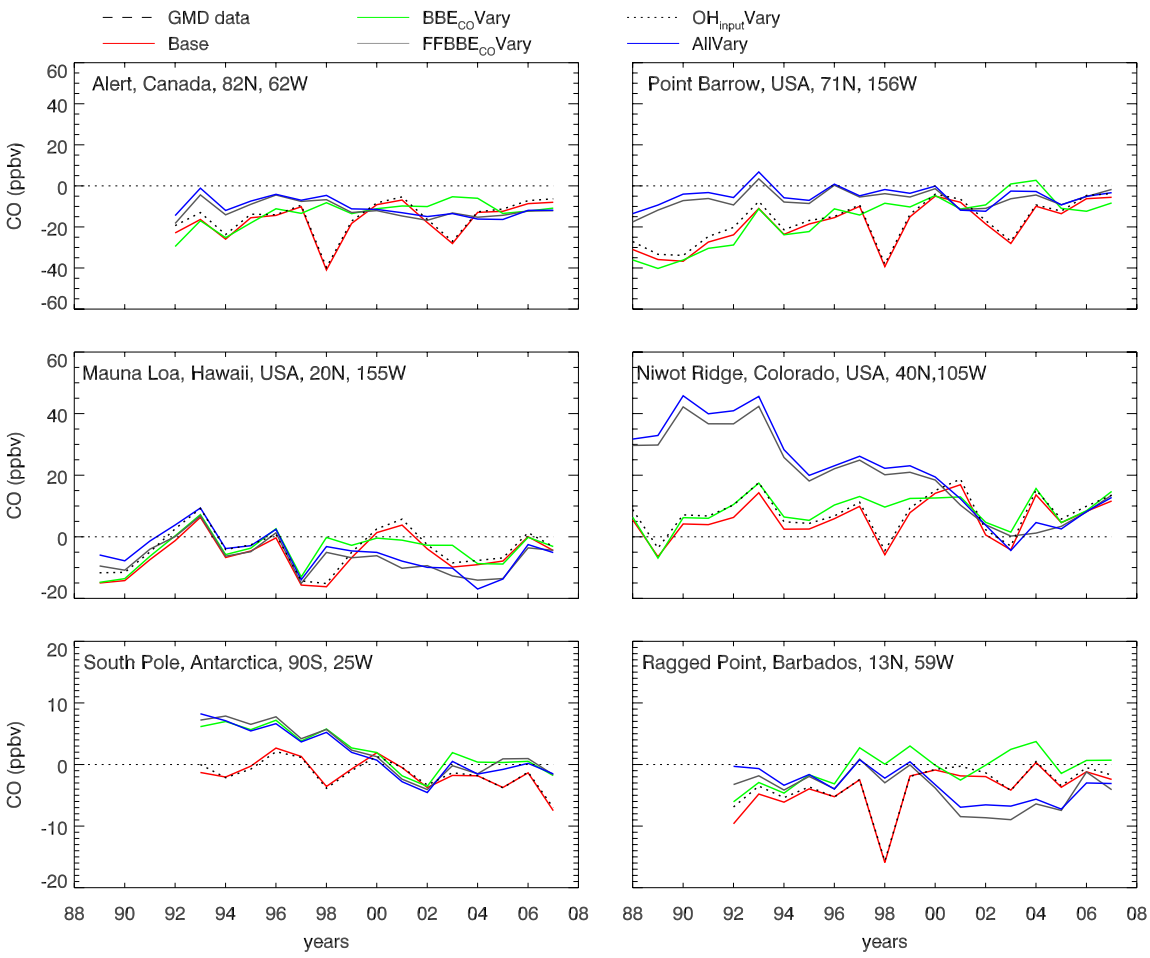

Figure S 13: Difference (simulated-measured; ppbv) of CO from GMD data and various scenarios at six GMD stations. Note the different scale on the y-axes.

TES/MLS comparisons:

1. **Base scenario**

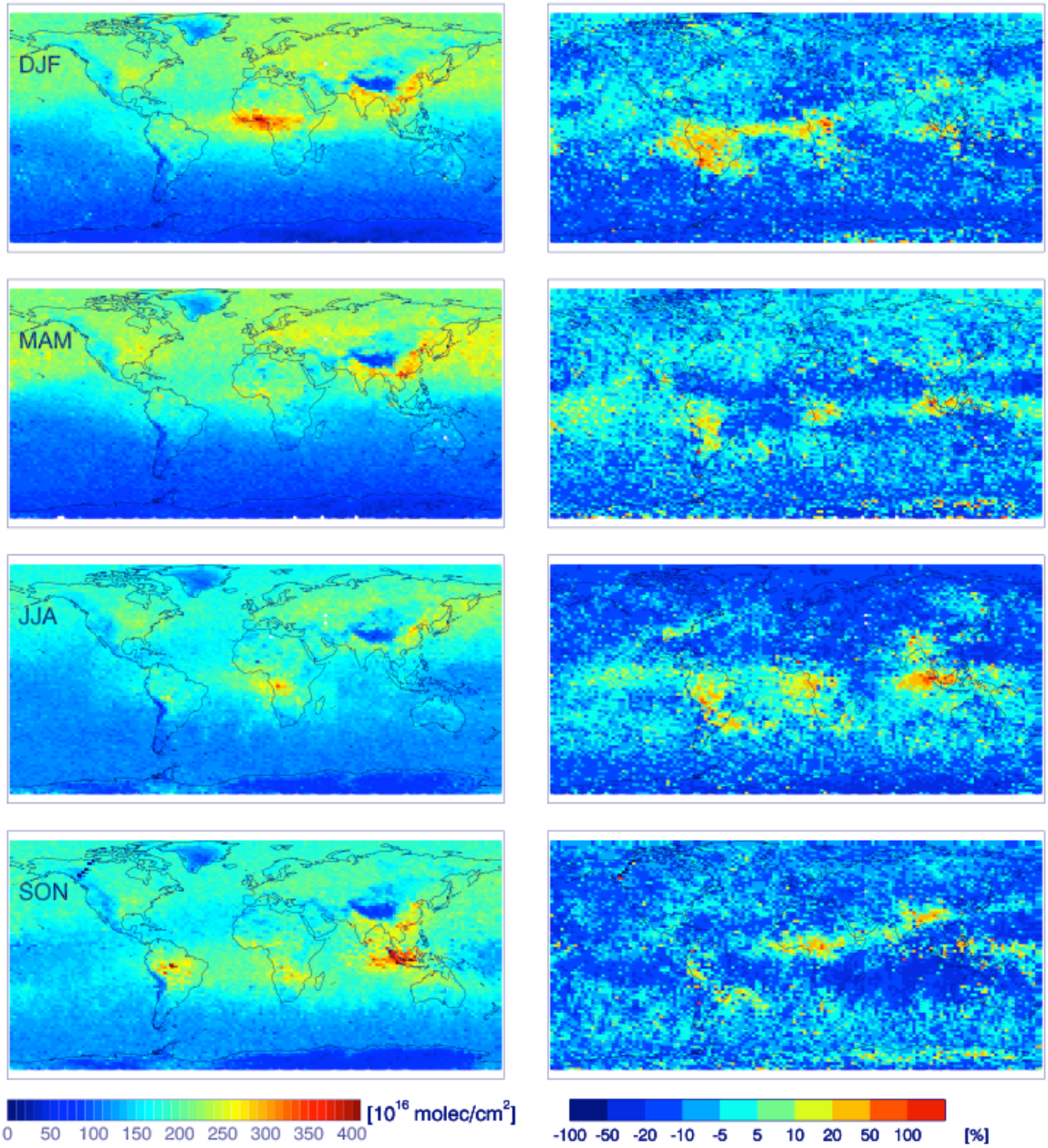

Figure S 14: Seasonal mean measured (TES/MLS, left column) and relative difference ((*Base*-TES/MLS)/TES/MLS, right panel) of the CO column for 2006-2007.

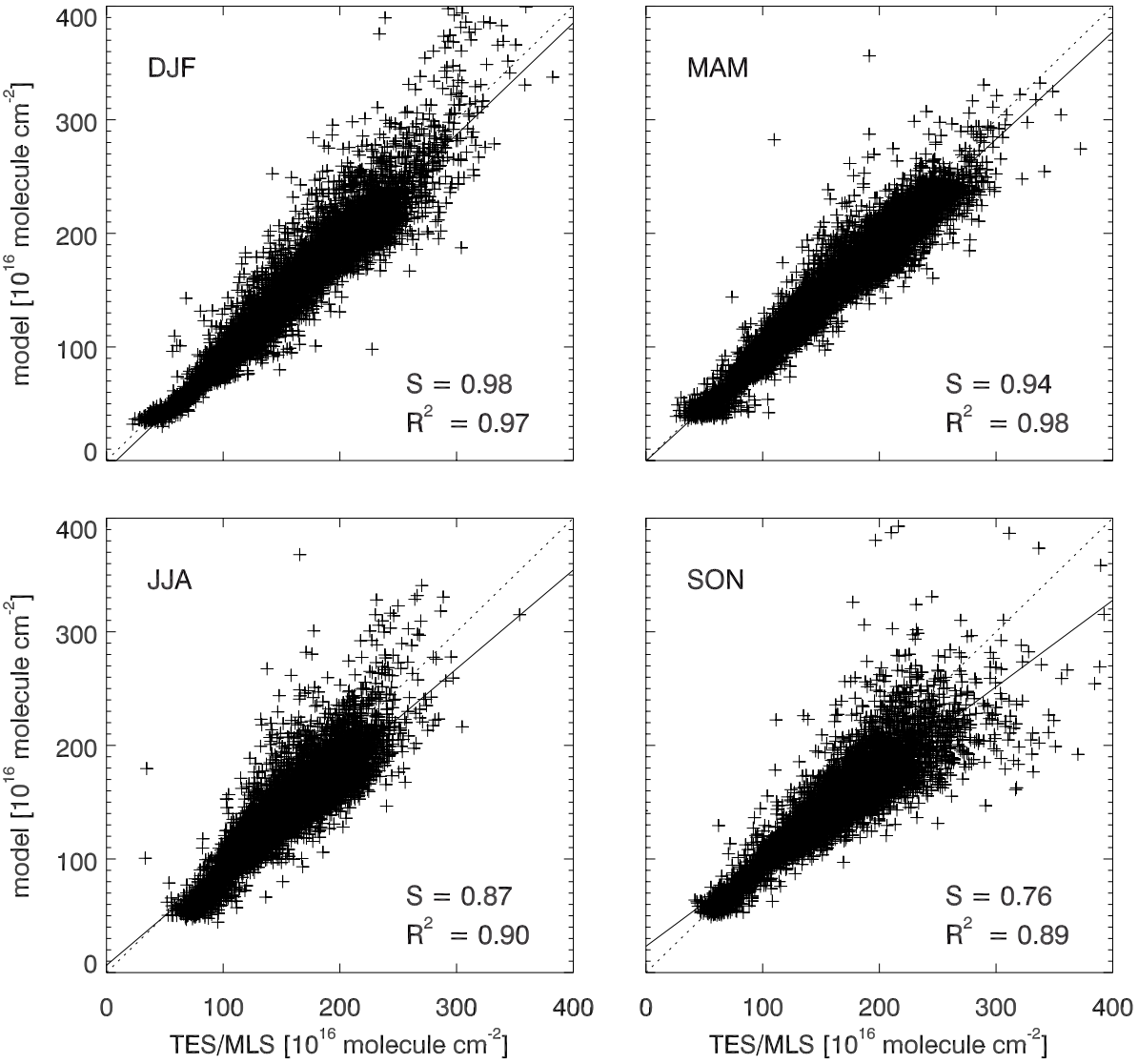

Figure S 15: Seasonal mean (2006-2007) CO columns (x10^{16} molecules/cm^2) from TES/MLS data and the *Base* scenario.

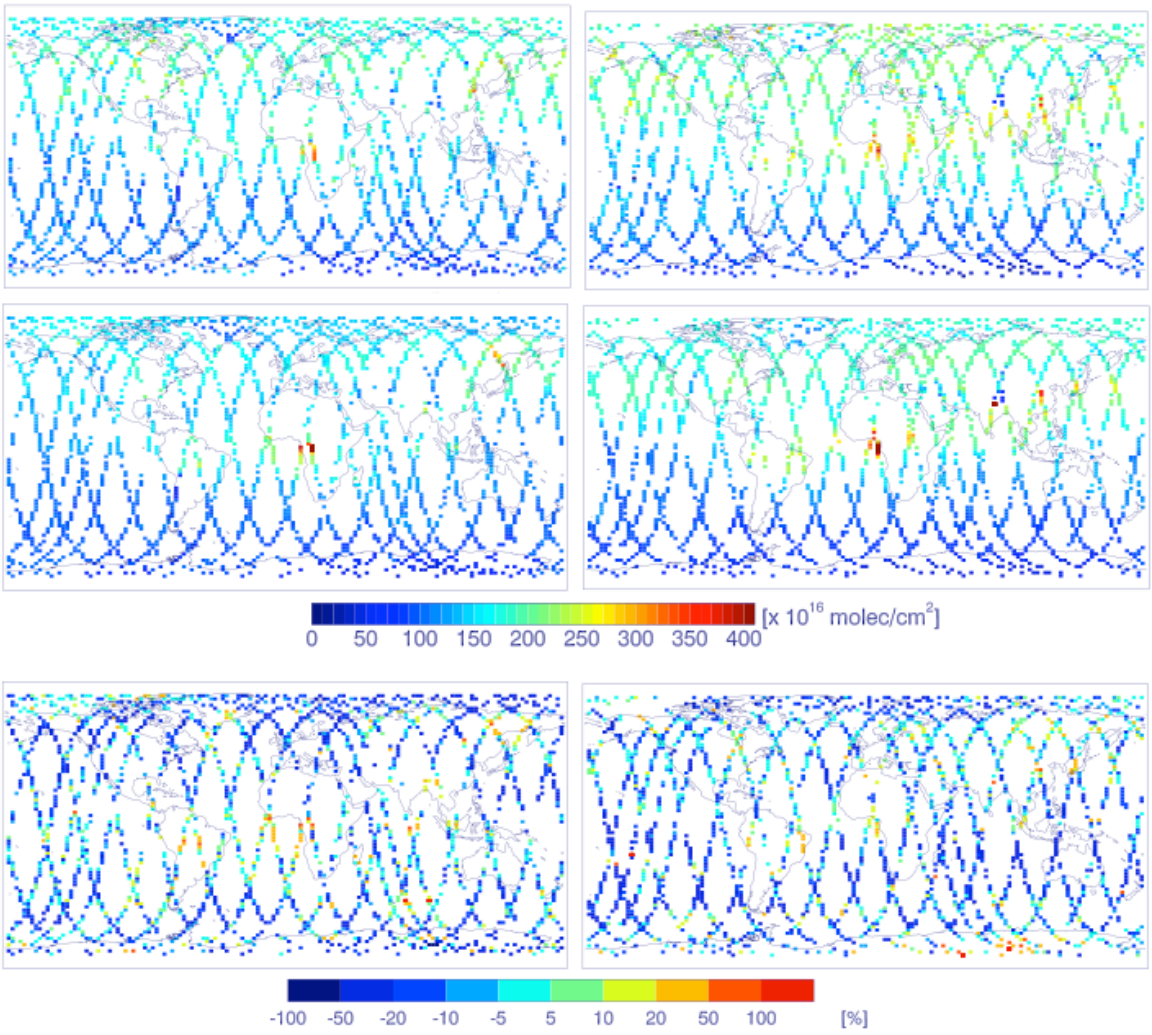

Figure S 16**:** Daily CO columns (x10^{16} molecules/cm^2) from TES/MLS data (top row) and the *Base* scenario (middle row), and their relative difference (%; (*Base*-TES/MLS)/(TES/MLS); bottom row) for July 1 (left column) and December 2 (right column) 2006.

MOPITT Correlations: Base scenario

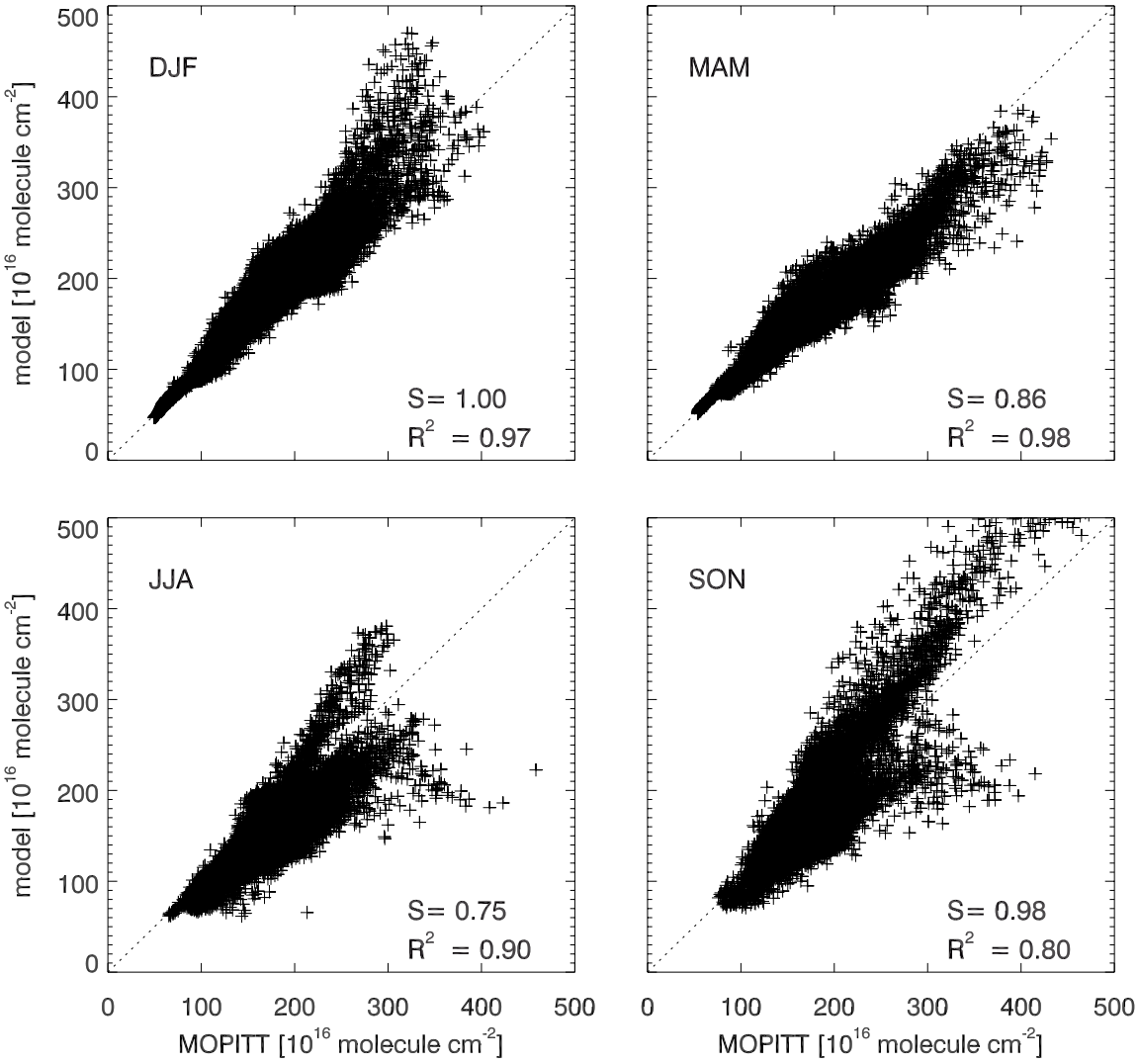

Figure S 17**:** Seasonal mean (2000-2007) CO columns (x10^{16} molecules/cm^2) from MOPITT data and the *Base* scenario.

TES/MLS: AllVary Scenario

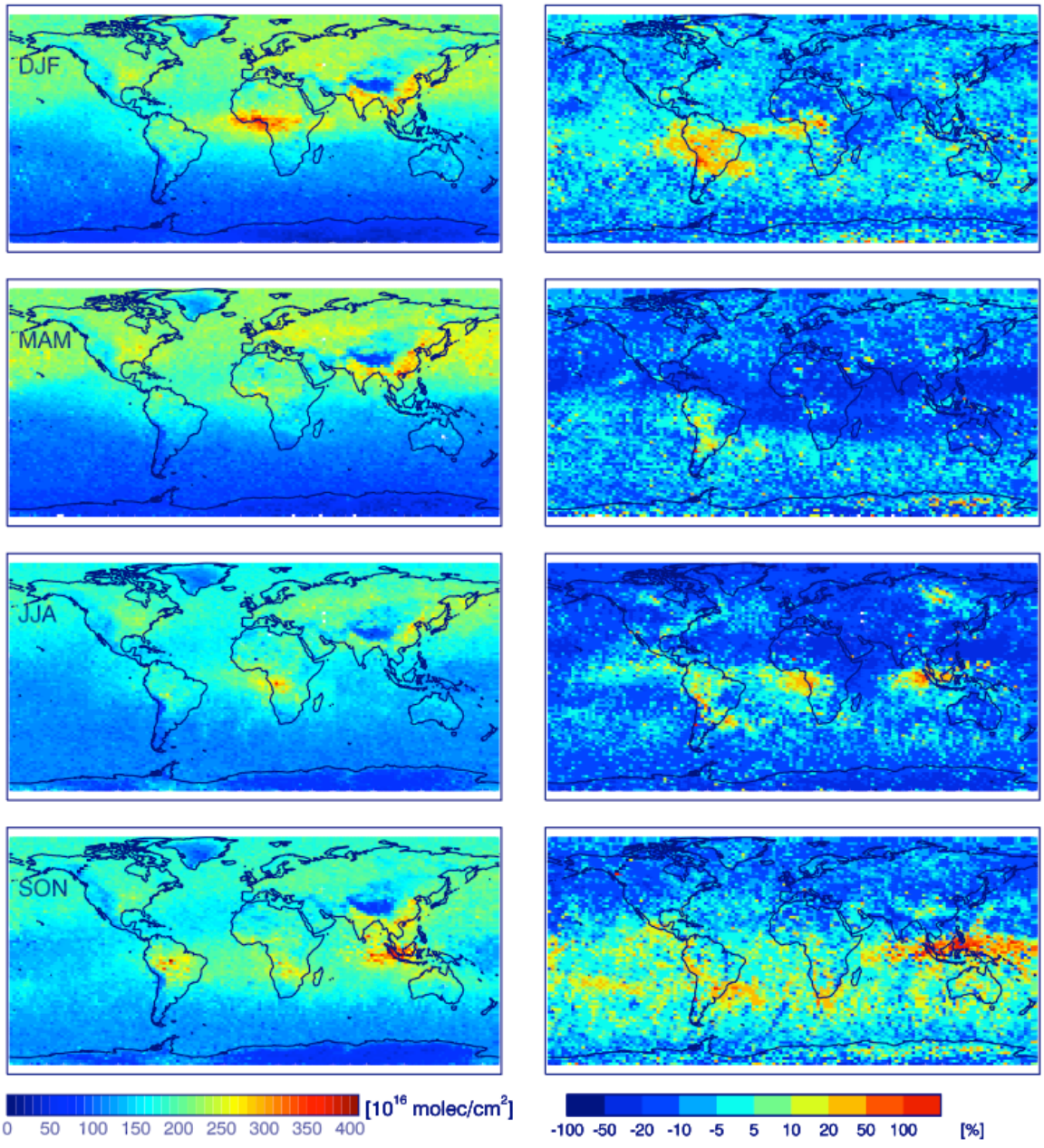

Figure S 18: Seasonal mean (2006-2007) TES/MLS (left column) CO columns (x10^{16} molecules/cm^2) and the relative difference (%) with the *AllVary* scenario ((*AllVary*-TES/MLS)/(TES/MLS), right column).

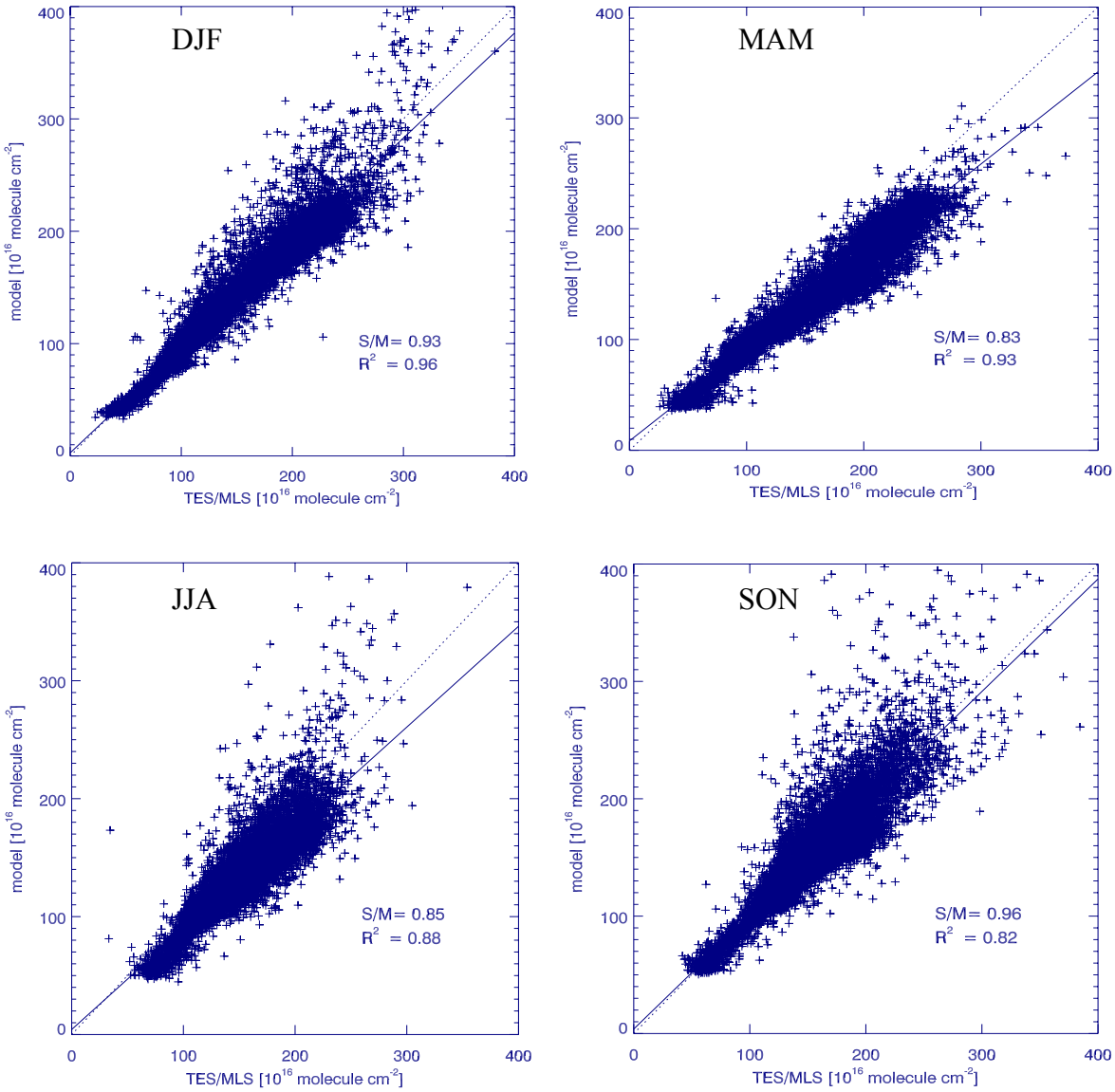

Figure S 19: Seasonal mean (2006-2007) TES/MLS and simulated CO column from the *AllVary* scenario.

MOPITT: AllVary scenario

In the figure below we show the seasonal distribution of the CO columns from the MOPITT (not shown in the paper) and the relative difference compared to the *AllVary* scenario.

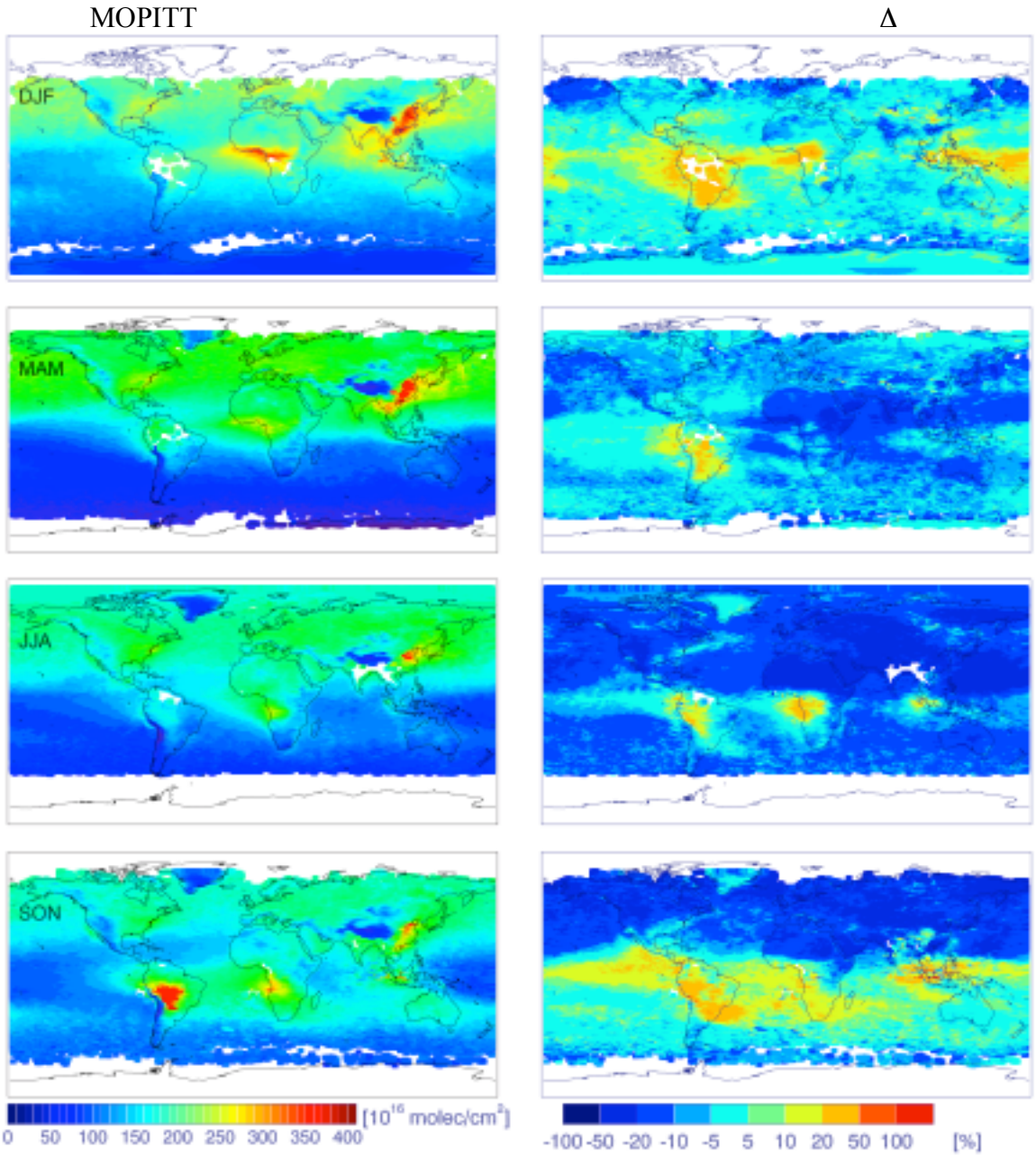

Figure S 20: Seasonal mean (2006-2007) CO columns from the MOPITT data (left column) and relative difference ((*AllVary*-measured)/measured, right column).

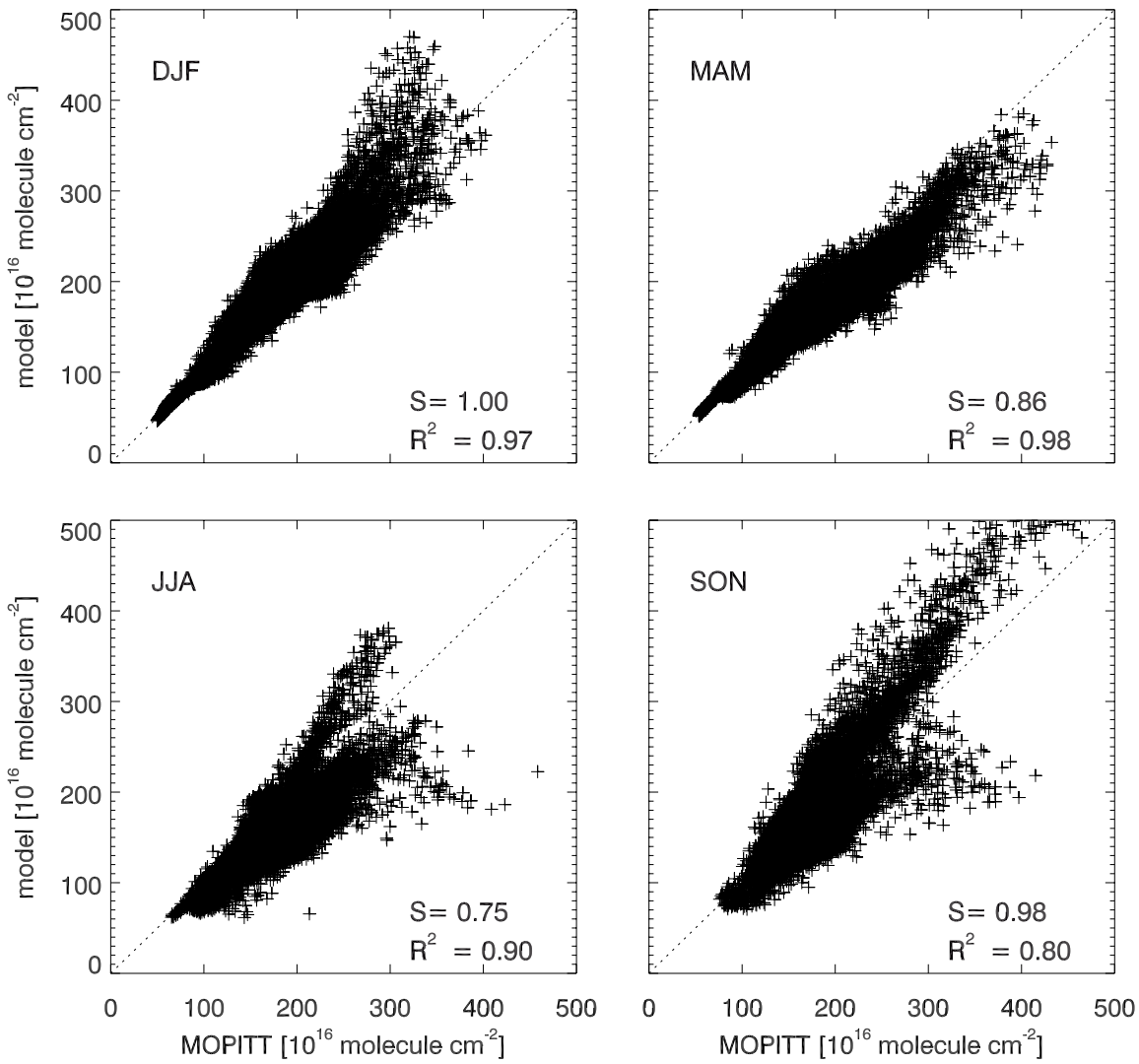

Figure S 21**:** Correlation plots of monthly mean (2006-2007) CO columns from the MOPITT data and *AllVary* scenario.

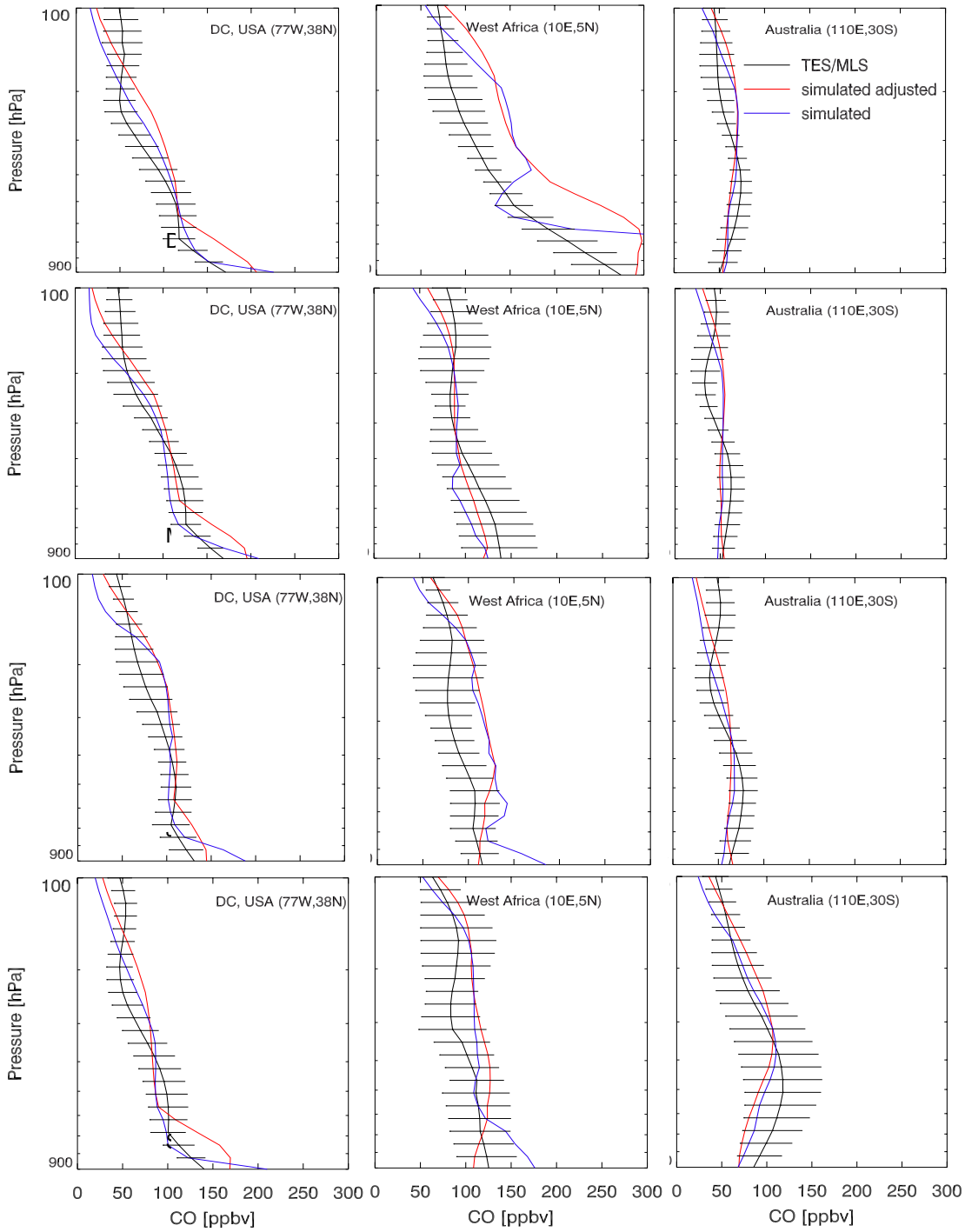

Figure S 22: Seasonal mean (2006-2007) Vertical profiles of measured (TES/MLS), simulated and simulated and adjusted with the averaging kernel of TES/MLS (labeled as 'simulated adjusted') of CO over selected locations using the *AllVary* scenario. The horizontal bars represent the standard deviation of the individual overpasses used to create the seasonal mean.

3. Comparison of simulated OH to full chemistry simulation.

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

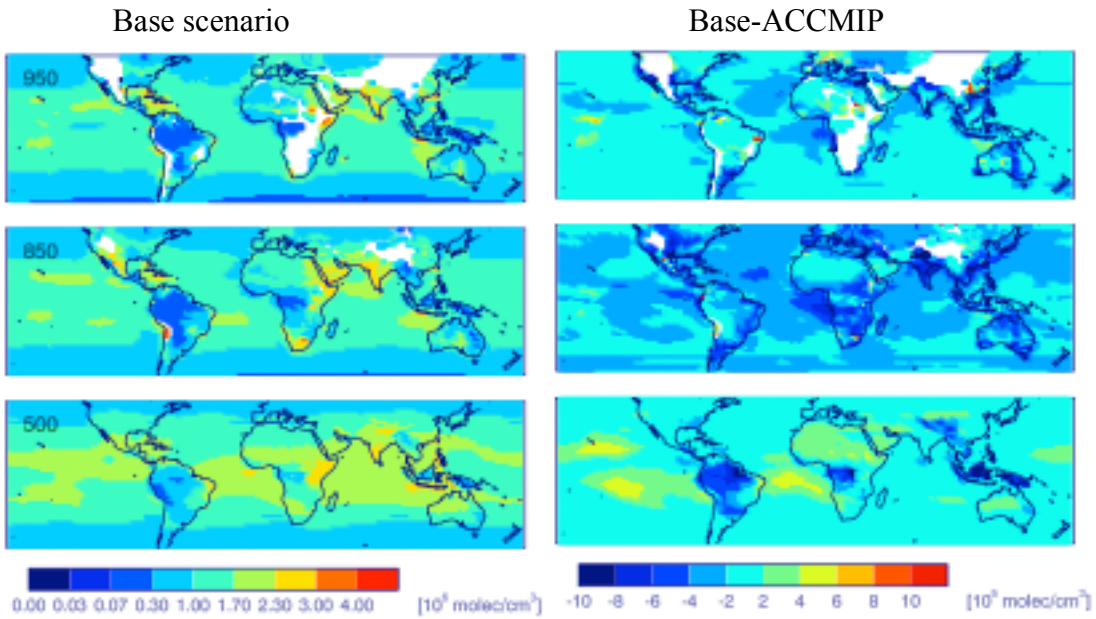

Figure S 23: Annual mean OH (left column; x10^6 molecules/cm^3) from 1999-2007 for the *Base* scenario and their corresponding difference (x10^5 molecules/cm^3) from the full chemistry ACCMIP (GEOS5CCM) simulation (*Base*-ACCMIP, right panels) at 950, 850 and 500 mbar (from up to bottom). White gaps indicate no model output at that pressure level.

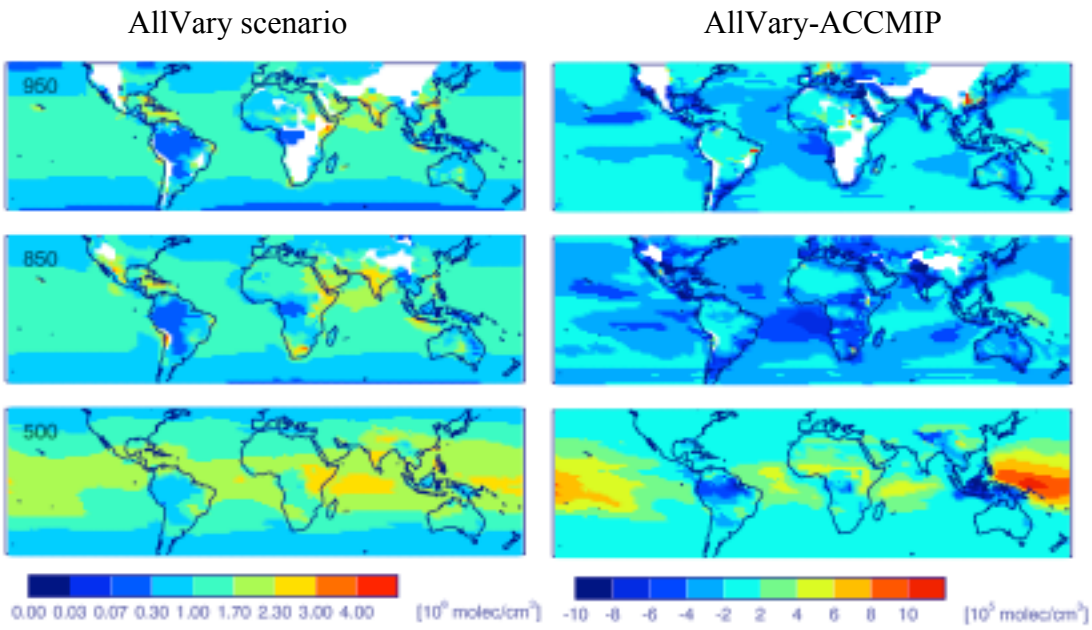

Figure S 24: Annual mean OH (left column, 10^6 molecules/cm^3) from 1999-2007 for the *AllVary* scenario and their corresponding difference (10^5 molecules/cm^3) from the full chemistry ACCMIP simulations (*AllVary*-ACCMIP, right column) at 950, 850 and 500 mbar (from up to bottom).

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

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

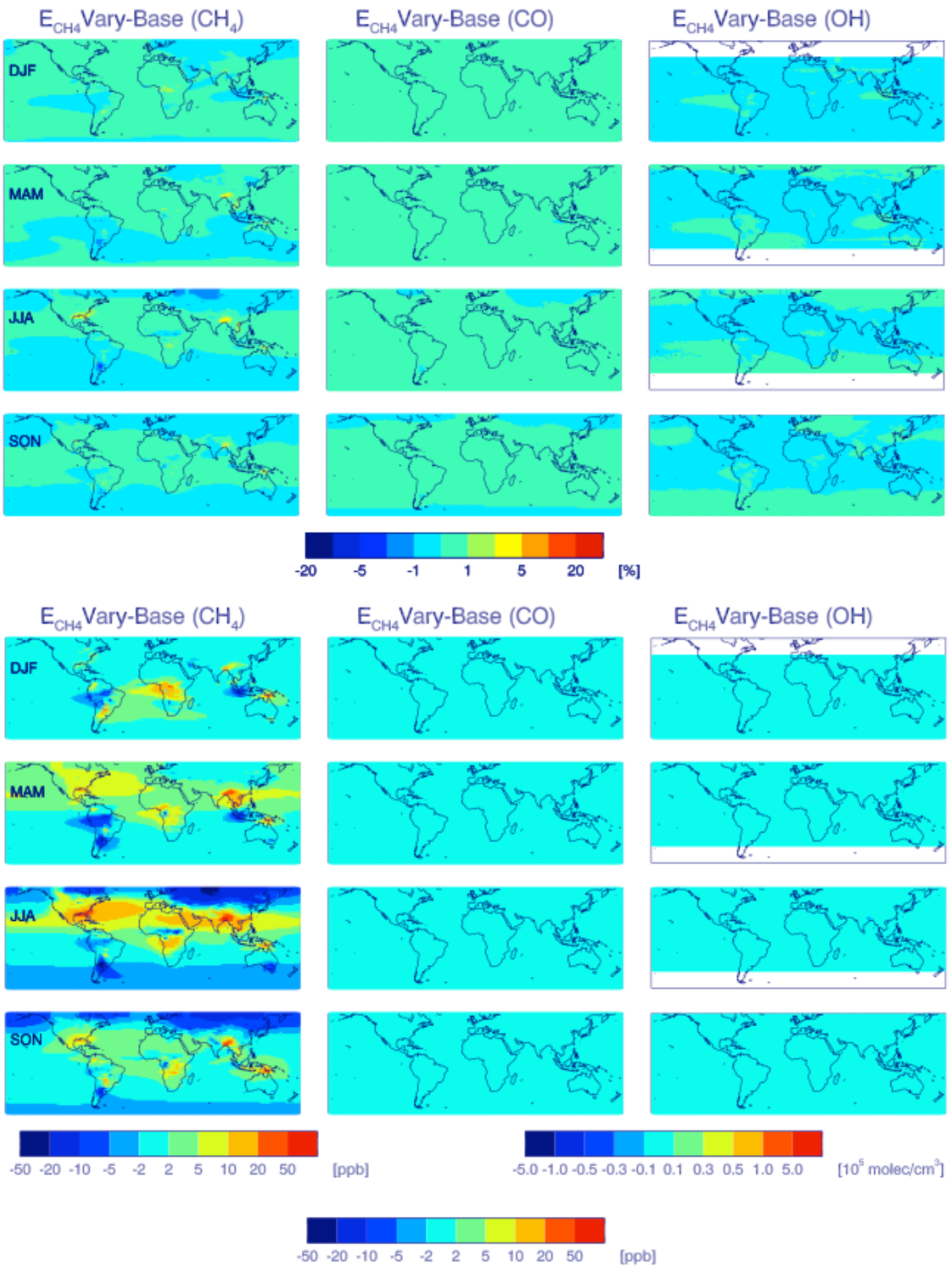

Figure S 25: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm^3) between the $E_{CH4}Vary$ and *Base* scenarios.

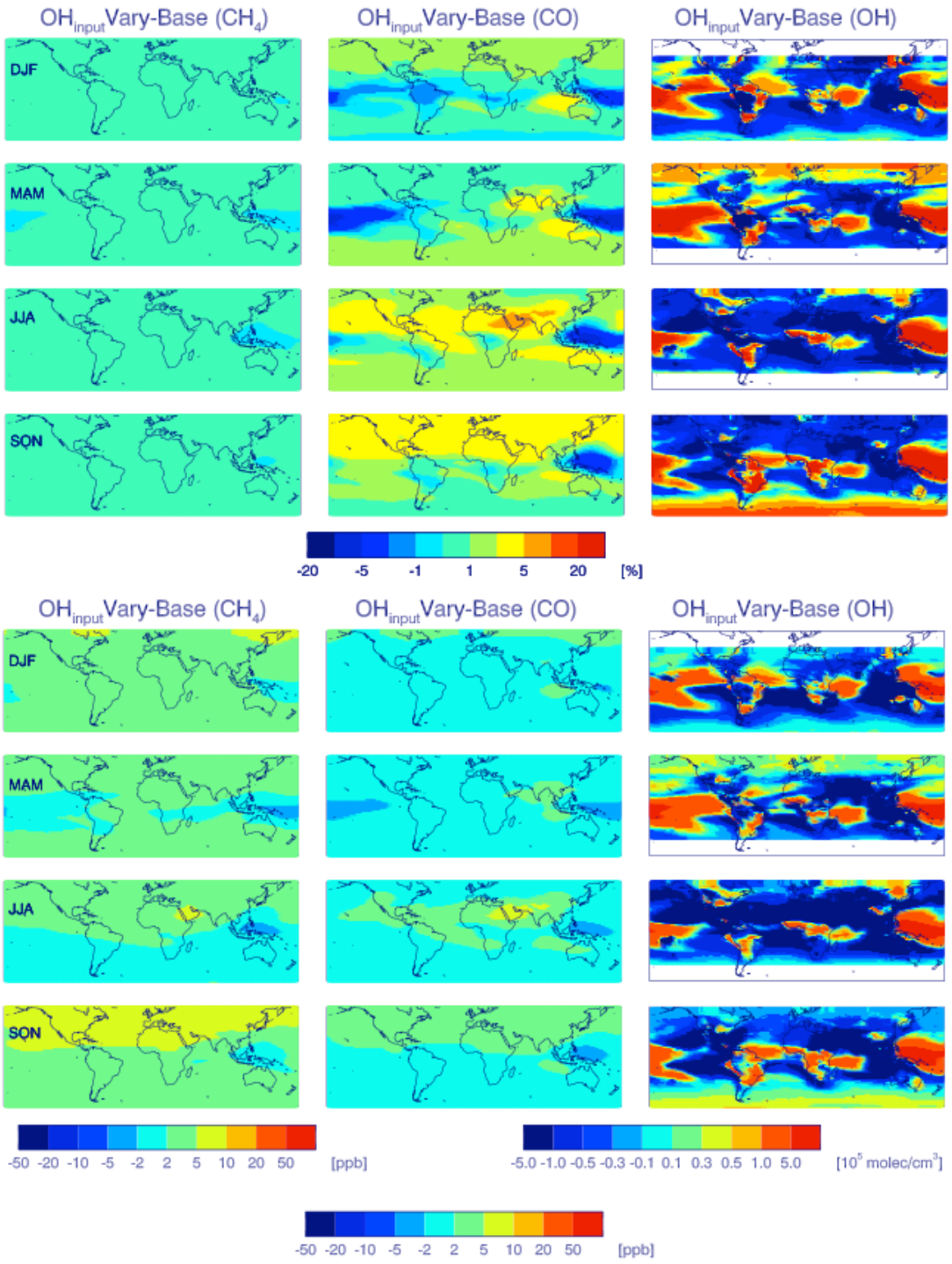

Figure S 26: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm^3) between the *OH$_{input}$Vary* and *Base* scenarios.

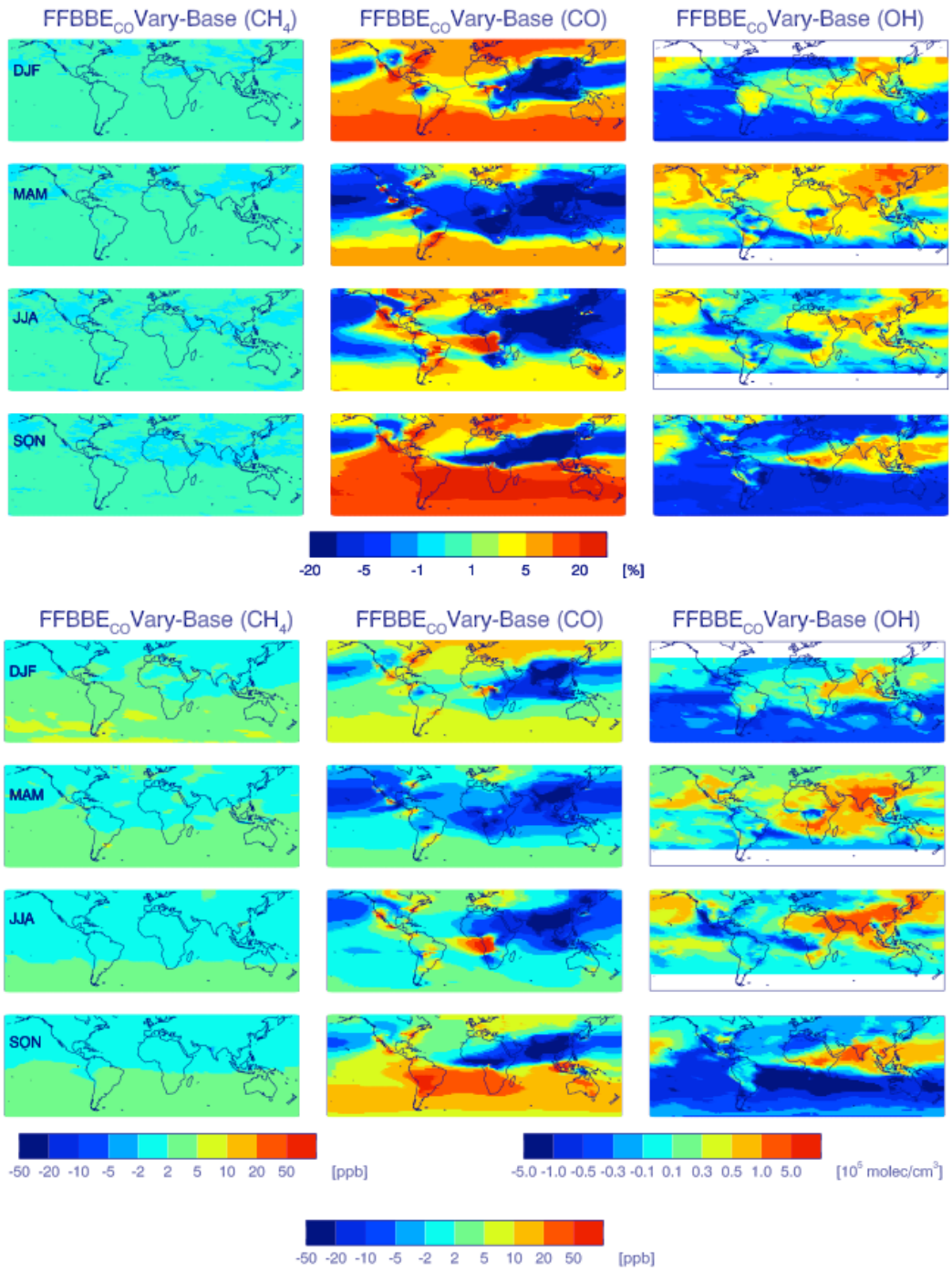

Figure S 27: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm^3) between the *FFBBE$_{CO}$Vary* and *Base* scenarios.

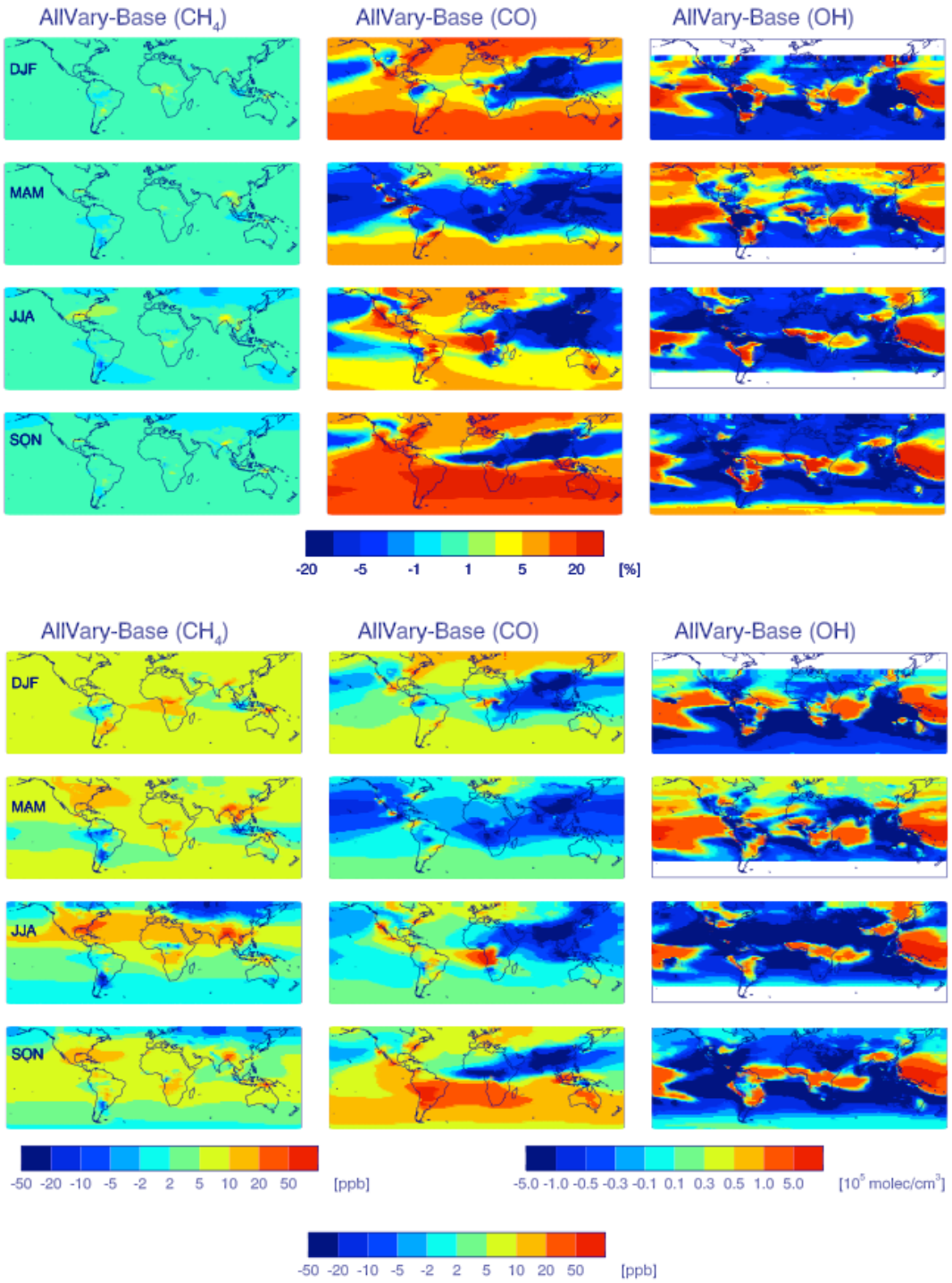

Figure S 28: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH ($x10^5$ molecules/cm^3) between the *AllVary* and *Base* scenarios.

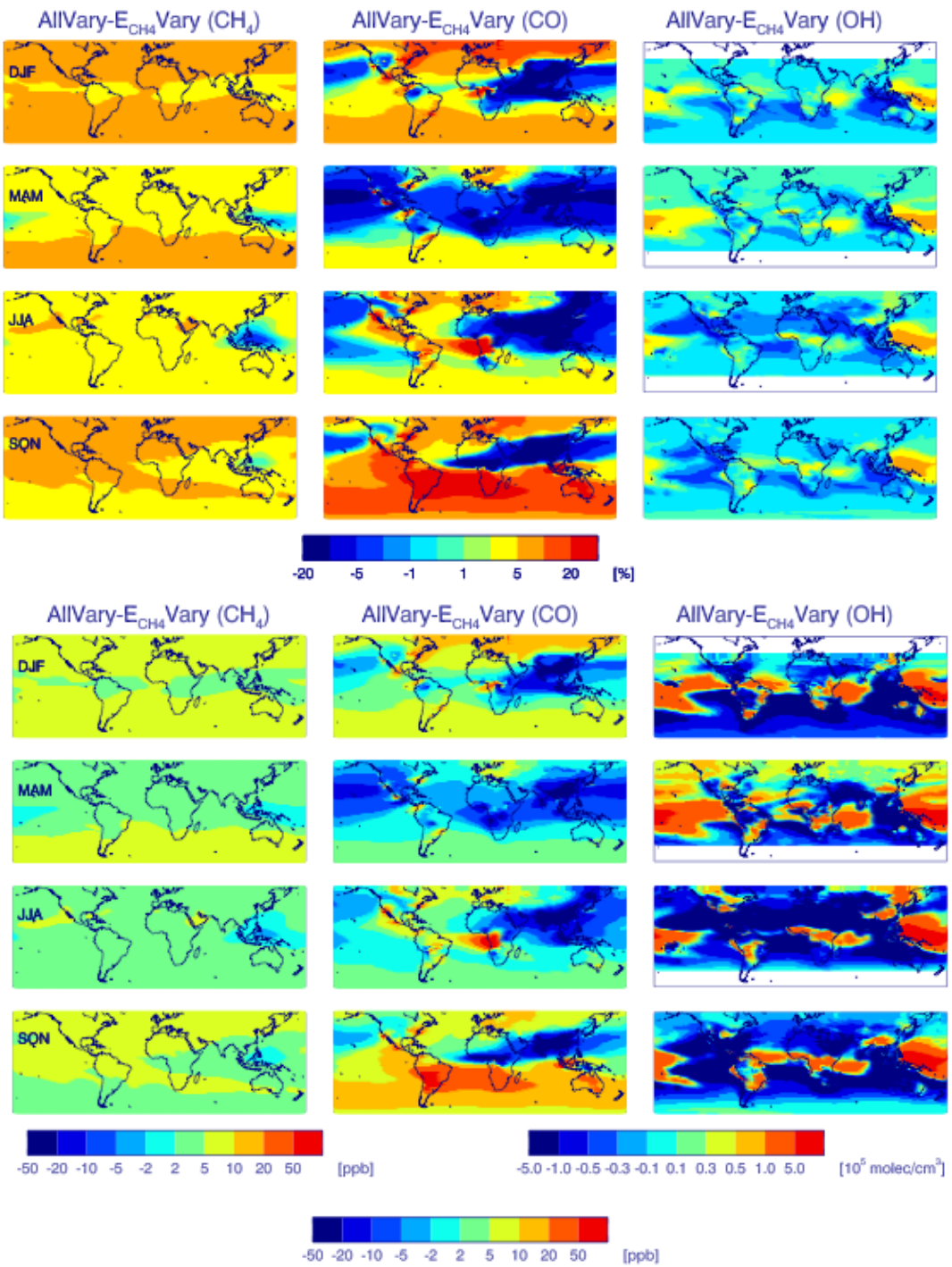

Figure S 29: Relative (%; upper panels) and absolute (lower panels) differences of seasonal, tropospheric methane (ppbv), CO (ppbv), and OH (x10^5 molecules/cm^3) between the *AllVary* and E_{CH4}*Vary* scenarios.

The figure below further demonstrate the importance of simulating interactive CH_4, CO , OH system. 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.

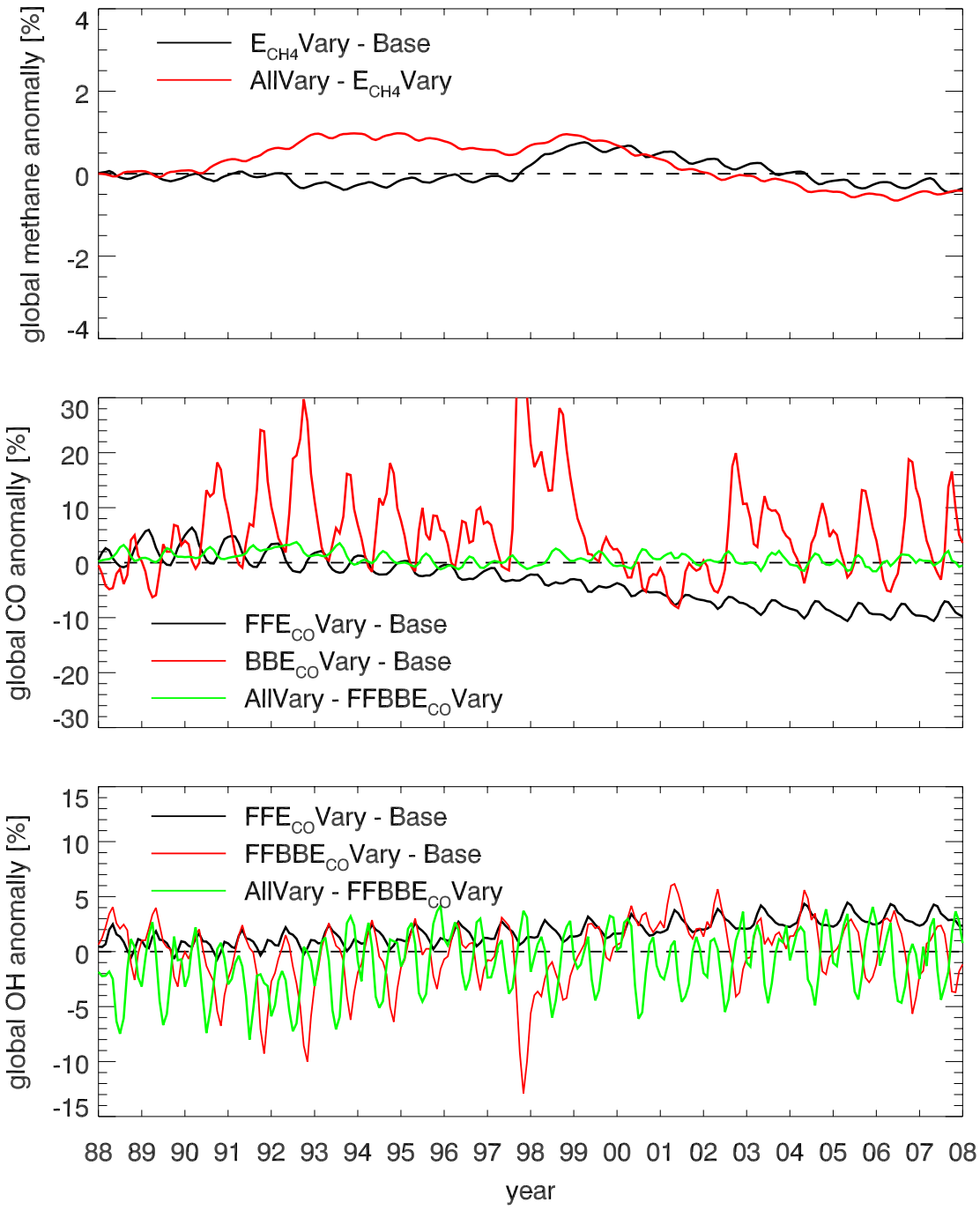

Figure S 30: Relative difference (%) of globally mass weighted tropospheric methane, CO, and OH (from up to bottom) between the different scenarios.