Interannual Variability and Trends of CH_4, CO and OH using the Computationally-Efficient CH_4-CO-OH (ECCOH) Module

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Introduction

- Methane (CH_4) is the second most important anthropogenic greenhouse gas (GHG). Its 100-year global warming potential (GWP) is 34 times larger than that for carbon dioxide. The 100-year integrated GWP of CH_4 is sensitive to changes in hydroxyl radical (OH) levels.
- Oxidation of CH_4 and carbon monoxide (CO) by OH is the main loss process, thus affecting the oxidizing capacity of the atmosphere and contributing to the global ozone background.
- Limitations of using archived, monthly OH fields for studies of methane's and CO's evolution are that feedbacks of the CH_4-CO-OH system on methane, CO and OH are not captured.
- In this study, we employ the computationally Efficient CH_4-CO-OH (ECCOH) module (Elshorbany et al., 2015) to investigate the nonlinear feedbacks of the CH_4-CO-OH system on the interannual variability and trends of the CH_4, CO, OH system.

Modelling Approach

The ECCOH module (Elshorbany et al., 2015) is implemented within the NASA GEOS-5 Chemistry Climate Model (Rienecker et al. (2008), Pawson et al. (2008), Ott et al. (2010), and Molod et al. (2012)):

Model Scenarios:

- ***Base:***
  Simulation period: 1988-2007
  Resolution: 2.5°×2° (longitude × latitude), 72 hybrid layers from the surface to 0.01 hPa.
  CH_4 emissions: Transcom (Patra et al., 2011) CTL scenario (only anthropogenic emissions vary)
  CO emissions: Annually-repeating emissions representative for year 2000.
  Chemistry: Fully interactive CH_4-CO-OH system, in which OH is accurately predicted by a set of high-order polynomials in meteorological variables (i.e., pressure, temperature, cloud albedo), solar irradiance variables (i.e., ozone column, surface albedo, declination angle, latitude) and chemical variables (e.g., CO, CH_4, NO_y, O_3, H_2O, and various VOCs). The computational cost of simulating tropospheric OH is reduced by about a factor of 500 when the full O_3-NO_x-VOC chemistry is replaced by the parameterization of OH (Spivakovsky et el., 1990; Duncan et al., 2000). The losses of methane and CO in the ECCOH chemistry module are determined by their reaction with tropospheric OH. Additional losses of methane in the stratosphere occur by reactions with OH, Cl and O^1D, whose distributions are simulated using archived, monthly fields.. CH_4, CO, and OH tracers are radiatively inactive.
- ***E_{CH4}Vary:*** Similar to *Base* but CH_4 natural emissions vary annually (TransCom EXTRA emissions scenario)
- ***BBE_{CO}Vary:*** Similar to *Base* but biomass burning (BB) CO emissions vary annually.
- ***$FFBBE_{CO}$Vary:*** Similar to *BBECOVary* but fossil fuels (FF) emissions vary annually.
- ***OH_{Input}Vary:*** Similar to *Base* but except the monthly, archived chemical variables (e.g., VOCs, NO_x) used as input to the parameterization of OH are annually varying.
- ***AllVary:*** Annually varying methane and CO emissions from all sources and annually-varying OH constraints.

Results and Discussion

Large Scale Interannual Variations in Methane, CO, and OH

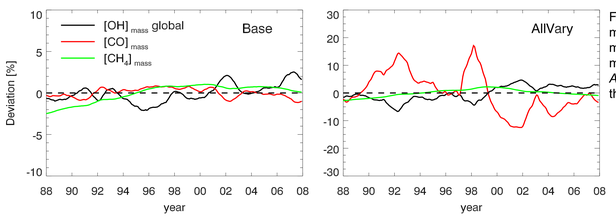

Fig. 1: Deviations of tropospheric, mass-weighted OH, CO and methane (12 month running mean) from the *Base* (left) and *AllVary* (right) scenarios. Note the different scales of the y-axes.

The magnitudes of the year-to-year deviations (relative to the mean (1988-2008)):

- CH_4: Small differences between the two scenarios since the *Base* scenario includes the important source of variation associated with anthropogenic methane emissions and methane's background is large. The nonlinear effects of the CH_4-CO-OH system on the temporal evolution of global mass-weighted methane are smaller, but significant, as compared to the effects of variations of methane emissions (see Fig. 2).
- CO: 10x greater in the *AllVary* scenario.
- OH: Increase by ±2% to ±5% in the *AllVary* scenario.
- Much larger variations on regional scales are masked in the global average

➢ Significant interannual variations in methane, CO, and OH.

Global Mean Growth Rates of Methane

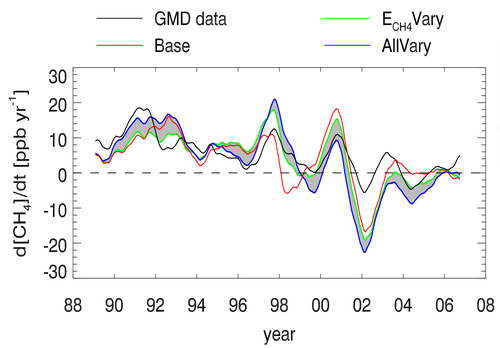

Fig. 2: Atmospheric methane growth rate (ppbv/yr, average of 92 GMD stations) from several scenarios. The shaded area is the difference between the E_{CH4}*Vary* and *AllVary* scenarios and it illustrates the combined effect of nonlinearities of the CH_4-CO-OH system on methane's growth rate.

- The simulated, interannual variation of methane's global growth rate agrees reasonably well with that estimated from GMD data.
- The nonlinear effects of the CH_4-CO-OH system on the temporal evolution of global mass-weighted methane are smaller, but significant, as compared to the effects of variations of methane emissions.

➢ **The nonlinear effects of the CH_4-CO-OH system cause significant fluctuations in methane's growth rate over our study period of ±4 ppb/yr.**

Methane Interannual Variability and trends

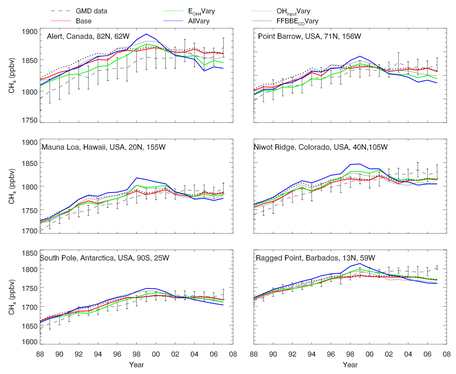

Fig. 3: Annual mean measured and simulated near-surface methane levels by different scenarios. Vertical lines represent the standard deviation of the measured annual mean.

- ➢ The *Base* scenario overestimates methane concentrations by 20-30 ppbv at the northern high latitude stations of Alert and Barrow during the 1980s and 1990s related to regional high biases in natural methane emissions
- ➢ Simulated methane improves in the northern hemisphere in the E_{CH4}*Vary* scenario, which includes annually-varying natural methane emissions.
- ➢ All scenarios capture the increasing observed methane trend in the 1990s, but under-predicts methane in the 2000 (Patra et al., 2011).

CO Interannual Variability and trends

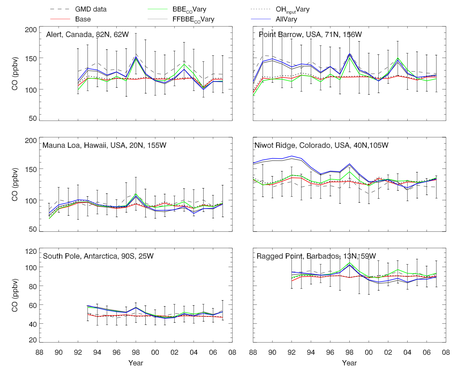

Fig. 4: Annual mean CO (ppbv) from several scenarios and observations at six GMD stations. Vertical lines represent the standard deviation of the observed annual mean

- ➢ the *Base* scenario does not capture the significant interannual variations associated with strong variations in emissions.
- ➢ Adding annually-varying anthropogenic and biomass burning CO emissions (*$FFBBE_{CO}$Vary*) improves the agreement, particularly in the northern hemisphere.
- ➢ Overall, annually-varying CO emissions (*$FFBBE_{CO}$Vary*) have a significant impact on the spatial distributions of tropospheric CO (±20%) and OH (±10%) relative to the *Base* scenario, and influence methane by ±1%.

Spatial and Temporal Distributions of the Loss Rates of Methane and CO

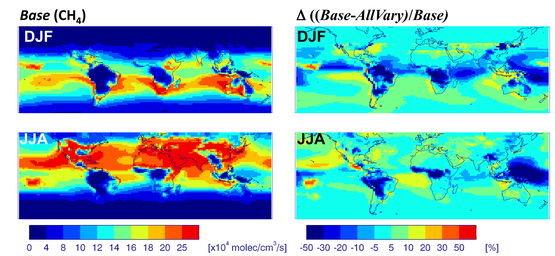

Fig. 5: Seasonal mean (1988-2007), mass-weighted tropospheric methane loss rate (left column; x10^4 molecules/cm^3/s) and the difference, relative (%) to the *AllVary* scenario ((*Base-AllVary*)/*Base*; right column).

- Most methane loss occurs between 30ºS and 30ºN since OH is most abundant in this region and methane's reaction with OH is temperature dependent.
- Methane's loss rates in the *AllVary* scenario are relatively higher, especially over biomass burning regions.

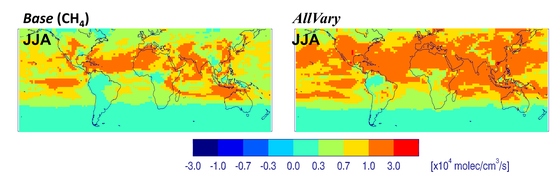

Fig. 6: Seasonal mean standard deviation of tropospheric methane loss rates (x10^4 molecules/cm^3/s) from the *Base* and *AllVary* scenarios.

- Interannual variability in the *AllVary* scenario are relatively higher.

➢ **Significant impact of non-linear chemistry on the IAV of methane loss rates**

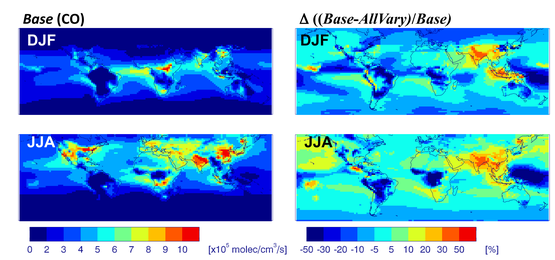

Fig. 7: Seasonal mean (1988-2007), mass-weighted tropospheric CO loss rates (left column; x10^5 molecules/cm^3/s) from the *Base* scenario and relative difference (%) between the *Base* and *AllVary* scenarios ((*Base-AllVary*)/*Base*; right column).

- In contrast to methane, a higher proportion of CO is lost at northern hemisphere mid-latitudes as the CO loss rate is less temperature dependent than methane's and the lifetime is shorter.
- CO loss rate from the *AllVary* scenario is relatively higher over biomass burning regions but lower over Asia

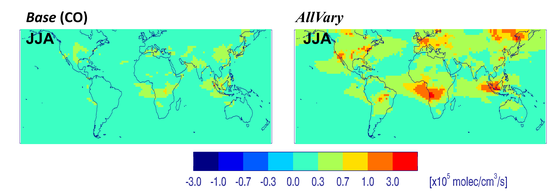

Fig. 8: Seasonal mean (1988-2007) standard deviation of tropospheric CO loss rates (x105 molecules/cm3/s) from the *Base* (left column) and *AllVary* (right column) scenarios.

- CO loss rate from the *AllVary* scenario show much higher variability that reaches up to ~20% compared to ~5% in the *Base* scenario.

➢ **IAV of CO loss rates is most sensitive to variability in CO emissions**

Conclusion

➢The nonlinear effects of the CH_4-CO-OH system cause significant fluctuations in methane's growth rate over our study period of ±4 ppb/yr.

➢ Significant impact of non-linear chemistry on the IAV of methane loss rates, while that of CO is most sensitive to variability CO emissions.

➢Future studies should consider the non-liner impact of the CH_4-CO-OH system when simulating methane growth rates and variability.

References

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