Introduction

- Methane (CH₄) 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 CH₄ is sensitive to changes in hydroxyl (OH) levels.

- Oxidation of CH₄ and carbon monoxide (CO) by OH is the main loss process, thus afflicting 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 CH₄-CO-OH system on methane, CO and OH are not captured.

In this study, we employ the computationally efficient CH₄-CO-OH (ECCOH) module (Elshorbany et al., 2015) to investigate the nonlinear feedbacks of the CH₄-CO-OH system on the interannual variability and trends of the CH₄-CO-OH system.

Modeling 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:

- Resolution: 2.5° x 2.5° (longitude, latitude); 72 hybrid layers from the surface to 0.1 hPa.
- CH₄ emissions: TransCom (Patra et al., 2011) CTL scenario (only anthropogenic emissions vary)
- Chemistry: Fully interactive CH₄-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, CH₄, NO, NO₂, O₃, H₂O, and various VOCs). The computational cost of simulating tropospheric OH is reduced by about a factor of 500 when the full NOₓ-VOC chemistry is replaced by the parameterization of OH (Sprykovskiy et al., 1999; Duscha et al., 2006). 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 with reactions by OH, Cl, and O_3. Their distributions are simulated using archived, monthly fields, CH₄, CO, and OH tracers are radially inactive.

- ECH4_Vary: Similar to Base but CH₄ natural emissions vary annually (TransCom EXTRA emissions scenario).
- ECH4-Vary: Similar to Base but biomass burning (BB) CO emissions vary annually.
- FFBBE-Vary: Similar to Base but except the monthly, archived chemical variables (e.g., VOCs, NOₓ) 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

The magnitudes of the year-to-year deviations (relative to the mean) (1988-2006):

- CH₄: 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 CH₄-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: 10% greater in the AllVary scenario.
- Methane loss rate is less temperature dependent than methane's and the lifetime is shorter.
- 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.

- Methane Intermural Variability and trends

The simulated, interannual variation of methane’s global growth rate agrees reasonably well with that estimated from GMD data.

- The nonlinear effects of the CH₄-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.

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

CO loss rate from the AllVary scenario is relatively higher over biomass burning regions but lower over Asia...

- CO loss rate from the AllVary scenario shows much higher variability that reaches up to 20% compared to ±3% in the Base scenario.

- Conclusion

The nonlinear effects of the CH₄-CO-OH system cause significant fluctuations in methane’s growth rate over our study period of ±4 ppb/yr.

- Significance impact of non-linear chemistry on the IAV of methane loss rates

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- CO Interannual Variability and trends

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- Methane Interannual Variability and trends

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