The Introduction

Methane (CH\(_4\)) is the second most important anthropogenic greenhouse gas (GHG). Its 100-year global warming potential (GWP) is 25 times larger than that for carbon dioxide. The 100-year integrated GWP of CH\(_4\) is sensitive to changes in hydroxyl (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 the feedbacks of the CH\(_4\)-CO-OH systems 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-Chem Climate Model (Riemer 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°-2.5° (longitude - latitude), 72 hybrid layers from the surface to 0.01 hPa.
  - CH\(_4\) emissions: TransCom (Pata et al., 2011) CTL scenario (only anthropogenic emissions vary)
  - 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., CH\(_4\), CH\(_3\)O, CO, H\(_2\)O, and various VOCs). The computational cost of simulating tropospheric OH is reduced by a factor of 500 when the CH\(_4\)-VOC chemistry is replaced by the parameterization of OH (Spracklen et al., 1999; 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 from reactions with OH, Cl, and O\(_3\); those distributions are simulated using archived, monthly fields. CH\(_4\), CO, and OH tracers are radially inactive.
  - \(\text{E}_{\text{CH}_4}\): Similar to Base but CH\(_4\) natural emissions vary annually (TransCom EXTRA emissions scenario)
  - \(\text{E}_{\text{CO}}\): Similar to Base but biomass burning (BB) CO emissions vary annually.
  - \(\text{BB}_\text{E}\): Similar to \(\text{E}_{\text{CO}}\) but except the monthly archived chemical variables (e.g., VOCs, NO\(_x\)) used as input to the parameterization of OH is annually varying.
  - \(\text{AllVary}\): 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)):

- 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: 3% greater in the AllVary scenario.
  - Decrease 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.**

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

- **Methane Interannual Variability and trends**
  - **Fig. 3** Annual mean measured and simulated near-surface methane levels by different scenarios. Vertical bar shows the standard deviation of the measured annual means.
  - **Fig. 4** Annual mean CO (ppbv) from several scenarios and observations at six GMD stations. Vertical bar shows the standard deviation of the observed annual means.

- **CO Interannual Variability and trends**
  - **Fig. 5**: Seasonal mean (1988-2007) mass-weighted tropospheric CO loss rates from several scenarios (ppm/yr, average of 92 GMD stations) (right column) compared to the base (left column).

**Conclusions**

- **The nonlinear effects of the CH\(_4\)-CO-OH system cause significant fluctuations in methane’s growth rate over our study period of 4 ppm/yr.**
- **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 show much higher variability that reaches up to 20% compared to 5% in the Base scenario.**
- **CO loss rate from the AllVary scenario is relatively higher over biomass burning regions but lower over Asia.**

**Future studies should consider the non-linear impact of the CH\(_4\)-CO-OH system when simulating methane growth rates and variability.**