Chemical Feedback from Decreasing Carbon Monoxide Emissions

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Motivations, data and methods

➢ **Global surface network from the 90s** (Novelli et al. 1998, 2003)

- ✓ Northern hemisphere dominated by anthropogenic sources and boreal fires, long term decreasing trends.
- ✓ Tropics and southern hemisphere variability governed by Biomass Burning emissions, large interannual variability
- ✓ CO sinks has also a strong seasonal cycle

Novelli et al. 2003

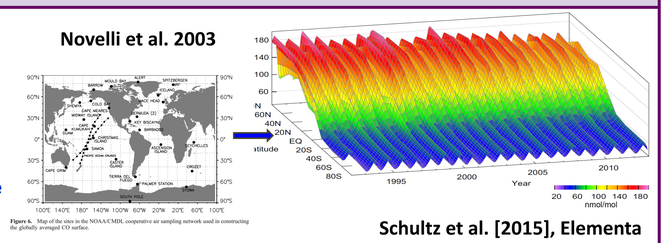


Figure 6. Map of the sites in the NOAA/CMDL cooperative air sampling network used in constructing the globally averaged CO surface.

Schultz et al. [2015], Elementa

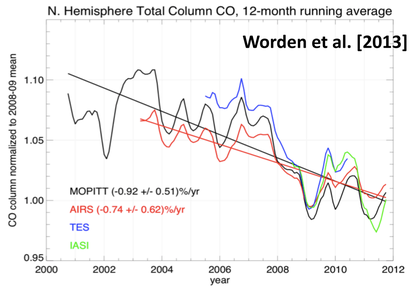
➢ **Satellites measurements since 2000s**

- ✓ Worden et al. [2013] compiled 4 different Nadir Earth Orbiting satellites retrieving CO in Thermal Infra-Red wavelengths.
- ✓ The main results are in accordance with the surface estimates

➢ **Reanalysis of satellite observations**

- ✓ Understanding the CO budget
- ✓ Developing pre-operational analysis and forecast system
- ✓ Explaining variability and long term trends

Worden et al. [2013]



12-month running averages for N. Hemisphere total column CO measurements normalized by the 08/2008–07/2009 average CO column for each instrument.

Prescribed stratosphere from previous WACCM simulation

CAM4-Chem
Ensemble Run : 30 CAM-Chem simulation

MOZART gas phase chemistry
Emission perturbation
- ✓ CO standard deviation 40 %, spatial correlation length (1000 km)
- ✓ VOC's standard deviation 30 %, spatial correlation length (1000 km)
- ✓ Prescribed Methane (surface level) standard deviation 10 %, spatially uniform

Meteorology assimilation, ensemble of initial conditions
- ✓ Space and time adaptive inflation

Ice model

Meteorological parameters ... Biogenic emissions

depositions ...

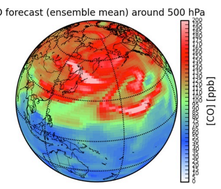
Ocean Model
SST climatology

Land Model (CLM 4)
Ensemble of deposition fields
Ensemble of biogenic emissions MEGAN V2.1

Goal:
Represent errors in modelling CO

1. **Emissions**
2. **Meteorology**
3. **Deposition**
4. **Initial conditions / chemistry**

CO forecast (ensemble mean) around 500 hPa



NCAR UCAR CESM COMMUNITY EARTH SYSTEM MODEL

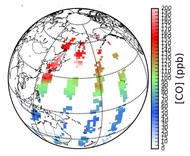
CESM/CAM-CHEM

- ➢ CESM122 / CAM4 / 1.9°x2.5°
- ➢ MOZART tropospheric chemistry (explicit OH calculation)
- ➢ Prescribed surface CH4
- ➢ MEGAN (biogenic) / FINN (fires) / RCP8.5 emissions (anthropogenic)

30 CAM-Chem forecasts
- ➢ Ensemble of emissions (and update of CO tags)
- ➢ Ensemble of transport
- ➢ Ensemble of deposition (land model)
- ➢ Ensemble of Chemistry

NCAR NASA ACOM

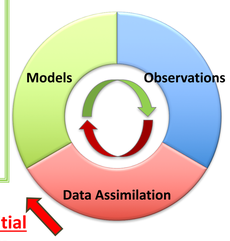
MOPITT observations around 550hPa



MOPITT

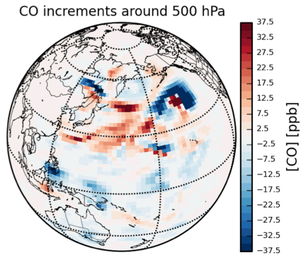
Observations

- ➢ **Meteorological observations**
  - ✓ DART/CAM (Raeder et al. 2012)
- ➢ **MOPITT V5J [Deeter et al., 2013]**
  - ✓ Daytime retrievals +/- 65°
  - ✓ Removed biased upper three levels
  - ✓ Super observations (horizontal): Error weighted average, no observation error correlation
  - ✓ Relocation according to the maximum of the averaging kernel



Ensemble of optimized initial conditions every 6 hours

CO increments around 500 hPa



Data Assimilation Research Testbed (DART)

- ➢ Assimilation of MOPITT and IASI described in Barré et al. JGR [2015]
  - ➢ [CO] inferred by MOPITT
  - ➢ P, T, U, V, Q inferred by Meteorological observations
- ➢ Space and time additive inflation / Spatial localization
- ➢ This reanalysis set-up is described in Gaubert et al. JGR [2016], the CO tags are scaled to conserve the total CO (actually assimilated).

$$CO^a_{i,j} = CO^f_{i,j} + \frac{CO^f_{i,j}}{CO^f_j}\Delta CO_j \qquad (5)$$

Annual and global tropospheric rates : the CO-OH-CH4 coupled chemical system

Context

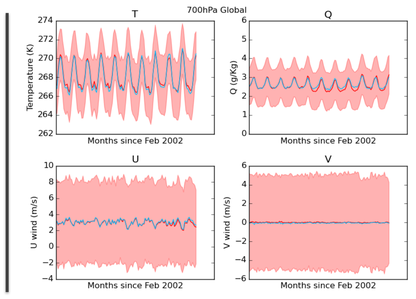
➢ **Simple chemical models for CH4/CO/OH interactions** (Prather 2007)

- ✓ CH4, CO and OH cannot be treated separately
- ✓ Perturbations in CO or CH4 affect the whole systems ('1ppb CO excites all the three modes')
- ✓ The CH4 lifetime is 40 % longer because of CO
- ✓ 'The atmospheric oxidation capacity is generally not sensitive to perturbations that may arise from variations or trends in emissions of natural and anthropogenic origin.' (Lelieveld et al. 2016)

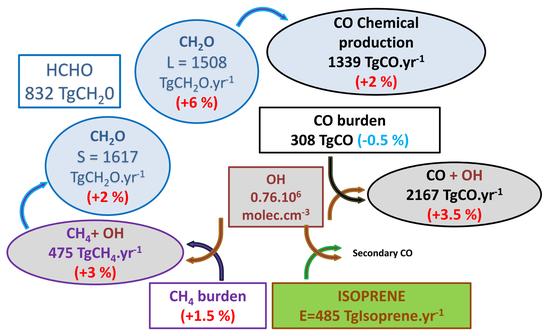
$$\frac{\mathrm{d}[CH_4]}{\mathrm{d}t} = S_{CH_4} - R_5$$

$$\frac{\mathrm{d}[CO]}{\mathrm{d}t} = S_{CO} + R_5 - R_6$$

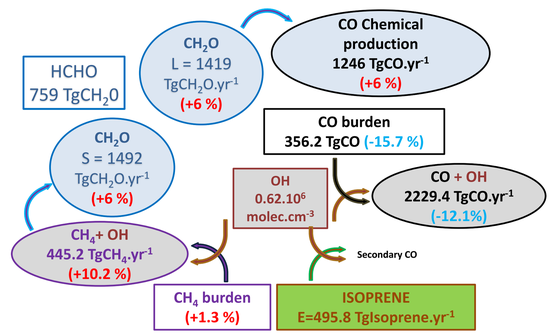
$$\frac{\mathrm{d}[OH]}{\mathrm{d}t} = S_{OH} - R_5 - R_6 - R_7.$$

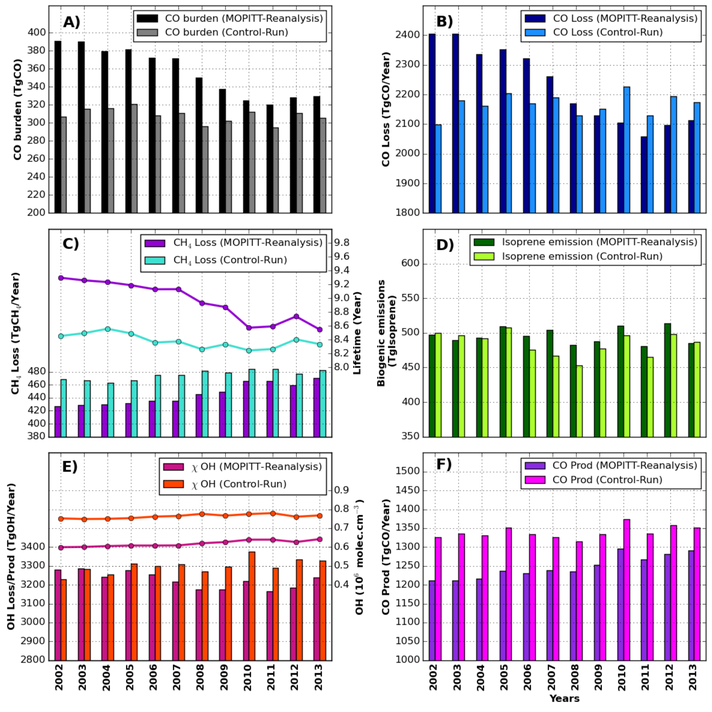


Control-Run



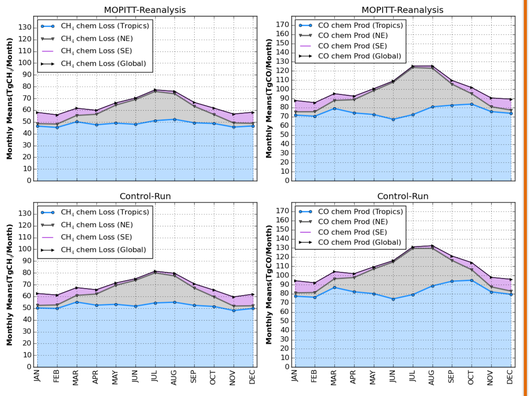
MOPITT-Reanalysis





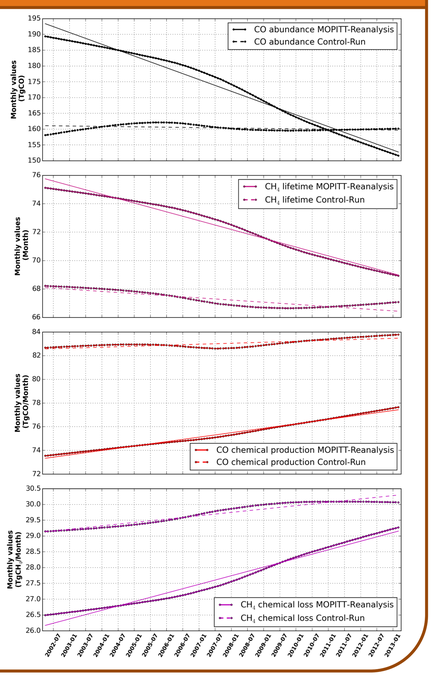
Impact on tropical CO chemicals trends

➢ **The tropics (between 30S and 30N) contributes to between 60 (summer) to 80 % (winter) of both the global tropospheric CH4 oxidation and the global CO chemical production**



➢ **We used the widely employed Seasonal Trend decomposition using LOESS (locally weighted scatterplot smoothing), or STL [*Cleveland et al.*, 1990].**

- ✓ This method is designed to identify the trend (T) and the seasonal (S) component from a given time series (Y), as well as a second order remainder (e). The general model is to decompose the time series into those 3 additive components as follow:
  - ➢ $Y(t) = T(t) + S(t) + e(t)$
- ✓ The reduction of [CO] across the period is remarkably well-correlated with the CH4 lifetime, confirming the mechanism presented above.
- ✓ The long-term trends provide strong evidence of a positive trend of the CH4 chemical loss and CO chemical production in the MOPITT-Reanalysis.



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