Review: A fifteen year record of CO emissions constrained by MOPITT CO observations, Z. Jiang et al., 2016, doi:10.5194/acp-2016-811

The work investigates the possible cause of the observed trend of a reduction of Carbon Mononixde (CO) emissions over the last 15 years over the northern hemisphere and parts of China. This trend is somewhat mitigated by an increased trend of CO emissions over India. The authors use global MOPITT remote sensing data of CO in the thermal infrared region to constrain model forecasts of CO concentrations and surface emissions. The model being used is the adjoint of the off-line global chemistry transport model GEOS-Chem.

The authors make 4 big assumptions: 1) Unknown model biases can be handled with by providing independent boundary conditions of CO concentrations over oceans each month from a Kalman Filter inversion run, 2) Local continental scale emissions can be estimated then by a 4dvar method constrained by MOPITT observations over land (and constrained by the boundary conditions of CO concentrations over the oceans), 3) The inversion system works best by removing a latitudional bias in MOPITT retrievals as derived from the HIAPER Pole to Pole Observations campaign (HIPPO), 4) The hydroxyl radical (OH) variability cannot explain the decrease in CO emissions if we put trust in the method of using MCF (methyl chloroform) measurements as a proxy for estimating atmospheric OH concentration change.

Comments:

- **Chapter: 2.1 MOPITT**

Did you do any data thinning on the MOPITT data and how did you screen the MOPITT data?

- line 176-178:

You need to describe the 4dvar adjoint method in more detail. What are typical numbers of N and it is not clear from the equation (line 178) or Figure 3 how you defined the length of the assimilation window in your 4dvar system. In GEOS-Chem met fields are typically updated every 6 hours – does this also correspond to your assimilation window (e.g. 6 hour window)? Or is your assimilation window a full month and observations are sampled every hour?

- line 186-189:

Cite: D.B.Jones, et al: Potential of observations from the Tropospheric Emission Spectrometer to constrain continental sources of carbon monoxide, doi:10.1029/2003JD003702, J. Geopys. Res, 2003

It is not clear to me why the authors cannot follow the method of constructing the observation error covariance matrix as outlined in the above paper (Dylan et al 2003). Of course TES and MOPITT are different products but as far as I remember MOPITT will also let you construct a retrieval error matrix as part of their released data products (they come with the averaging kernels). It is true that there is some vertical correlation in the averaging kernels but cannot account for the information loss of a uniform or flat construed observation error.

- 190-196

Reword and emphasise that posterior emissions estimates (e.g. Table 1) do not have uncertainty reduction error bars because of the way the adjoint method works and ask Daven Henze if there is a reference for that.

- line 194-196: As shown by Heald et al (2004), different assumptions about the inversion configuration can produce differences in the source estimates that are significantly larger than the a posteriori errors.

Is this statement related to the bias correction in the next paragraph (line 197-209)? Why is this important here?

- line 197-198: Removing the bias in initial conditions is essential for inverse analysis, and can be performed with various data assimilation techniques.

Have you got a reference for this? I have heard people claiming (I am not one of them) that in a good inversion system there is no bias correction needed. Have you tested your system without bias correction?

- line 218-220: They demonstrated that the systematic bias associated with North American CO emissions due to OH distribution can be reduced by up to 50% with optimised boundary conditions. Similar optimisation on the boundary condition can also be employed in global model, for example, Pfister et al. (2005) constrained biomass burning CO emissions from boreal North America with optimised CO fields outside the impacted region.

How does this relate to your work? Your are using pre-calculated OH fields from a full chemistry run. Are you making the point here that the influence of the badly understood OH bias can be reduced by optimised CO 3D boundary conditions (e.g. from your Kalman Filter at the beginning of each month)? Please clarify.

- Figure 3

This needs clarification in the Figure caption or text. If I am right to assume that your Kalman filter runs from 1st of March until 31nd December first and is

completely independet of the 4dvar inversion in the assimilation window? And there is no feedback of the 4dvar inversion results to the boundary conditions of the following months?

- 4.1 Long-term variation of global tropospheric OH

Krol et al. found a somewhat different result of OH trends based on MCF measurements and model studies. Admittedly for a different study period (1978-1998). You could (or should) cite that paper: M. Krol et al., 1998: Global OH trend inferred from methylchloroform measurements, 103, p.10,697—10,711, 1998, J. Geopys. Res.

- 4.2 Long-term variation of global CO emissions

It would be a good idea if you split the section into different smaller subsections:

4.2.1 Emissions US
4.2.2 Emissions EU
4.2.3 Emissions India + South East Asia
4.2.4 Biomas Burning Emissions

etc.

- line 425-427: In a recent study, Schnell et al. 92015) evaluate surface O3 concentrations simulated by multi-models for North America and Europe. They found most models can provide good simulations for the patterns of O3 but cannot reproduce the magnitude.

I do not think citing an ozone study supports your argument in terms of CO.

- line 466-468

Reformulate the part including 'MCF'. I do not think you have used MCF to 'evaluate changes in the sources and sinks of atmospheric CO … '.

- Table 1

Add a fifth column of global total posterior emissions to the 3 individual sub tables: 'MOPITT Columns (Tg/year)', 'MOPITT Profile (Tg/year)' and 'MOPITT Lower Profile (Tg/year)'.

Add a sixth column to the 3 individual sub tables for posterior CH4 and VOC production.

Also append 4 single columns for the global prior emissions in each year.

e.g.

Year,US,EU,China,India,CH4,VOC, US,EU,China,India,CH4,VOC, US,EU,China,India,CH4,VOC, PRIOR ANTHRO, PRIOR CH4, PRIOR VOC, PRIOR TOTAL

And comment on these global budgets in the main text.

- Figure 11

I am not convinced your method of singling out the meteorological effects works as intented. Firstly, what exactly is being defined as meteorological conditions? I think the accumulation of surface CO, especially over the tropical regions and to a lesser extened the slight loss of CO at higher latitudes is an artifact and CO builds up, unrealistically, in GEOS-Chem tagged tracer mode.

I am not asking you to conduct more model calculations. However, it would have beend interesting to see if a full global 4x5 GEOS-Chem CO chemistry run gives a similar answer than Figure 11a and 11b.