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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A fifteen year record of CO emissions constrained by MOPITT CO observations" *by* Zhe Jiang et al.

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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.

Thank you for your comments. Modifications have been made to improve this manuscript.

Q1: Chapter: 2.1 MOPITT: Did you do any data thinning on the MOPITT data and how did you screen the MOPITT data?

We employed the same data quality control as our previous studies. Detailed description has been added in Section 2.1.

Q2: 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?

Thank you for your suggestion! More description has been added.

In order to match model output, the high resolution MOPITT measurements are averaged temporally (one-hour resolution) and spatially (4°x5° resolution) to produce grid mean observations. The length of assimilation window is one month. The number (N) of grid mean observations in one month is around 10000.

Q3: 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.

Jones et al. (2003) used the NMC method to assess the model transport errors. This approach uses pairs of model forecasts, of different length, but which are valid for the same time, to characterize the model errors. We do not have such forecasts available during this analysis period.

We have compared the discrepancies associated with two types of error covariance matrix in the preparation stage of this work: 1) diagonal matrix (this work); 2) full error covariance matrix including vertical correlation, based on MOPITT error covariance. Our results show that the difference in the scaling factors is small, perhaps due to the large amount of satellite measurements in our global scale inversion.

Because we are focusing on the mitigation of effects of systematic errors, we used the diagonal matrix to keep consistency with our previous studies. However, as the reviewer indicated, a better description for the error covariance matrix is important. We will improve our methodology in our future study.

Q4: 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.

Thank you for your suggestion! The discussion has been modified.

Q5: 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?

We hope to demonstrate that the actual a posteriori uncertainty (including systematic errors) is much larger than the a posteriori uncertainty calculated based on Gaussian assumption (random errors).

Q6: 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?

We have tested the effects of initial condition in our previous study. As shown in Figure 4a of Jiang et al. (2013), there are large discrepancies between MOPITT and original model simulation due to the accumulation of model errors prior to the assimilation window. We cannot use the biased initial condition for the inverse analysis.

“a good inversion system there is no bias correction needed” is valid for the ideal condition. However, there are always systematic biases, and we cannot ignore them. For example, Figure 1 shows noticeable discrepancies between MOPITT and HIPPO. We have to mitigate these discrepancies using latitude dependent correction factors, although we know the best approach is an update of retrieval algorithm.

Q7: 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.

We hope to demonstrate that the influences of systematic errors can be mitigated by the optimization on the boundary condition. We have changed the statement to make it more concise.

As the reviewer indicated, the optimization on the boundary conditions (e.g. around North America) can really mitigate the influences of OH bias on a posteriori estimation of North American CO emissions. Although the OH distribution over North America continent is still biased in a reginal inversion, the adverse effects of biased OH distribution on the CO inflow from outside of North America can be significantly reduced.

Q8: 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?

Thank you for your suggestion! The Figure caption has been changed.

Q9: 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.10,697âĂŤ10,711, 1998, J. Geopys. Res.

The citation has been added.

Q10: 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 Emission;s US 4.2.2 Emis-

sions EU; 4.2.3 Emissions India + South East Asia; 4.2.4 Biomas Burning Emissions etc.

Thank you for your suggestion! Two subsections “Regional analysis for anthropogenic emissions” and “Regional analysis for biomass burning emissions” have been added.

Q11: 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.

This citation has been removed.

Q12: 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 . . . ’.

The statement has been modified.

Q13: 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.

Thank you for your suggestion! Three columns of global total anthropogenic emissions have been added in Table 1. A new table (Table 2) was added to show the annual variation of biomass burning emissions.

We didn’t provide values for CO sources from VOC and CH4 oxidization because our

results for these two sources are inconclusive. The values for a priori emissions are also excluded because the tables are already complex.

Q14: 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.

A very good question! Our forward model simulation, based on various versions of the meteorological fields (i.e. GEOS-4, GEOS-5 and GEOS-FP), is not an ideal tool for the analysis of influences of meteorological fields. We have modified the text to emphasize on this point.

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