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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Optimizing global CO emissions using a four-dimensional variational data assimilation system and surface network observations" *by* P. B. Hooghiemstra et al.

Anonymous Referee #1

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The authors have conducted in inverse modeling analysis of surface observations of CO using a 4-dimensional variational (4D-Var) data assimilation system. They showed that the surface data can reduce uncertainty on estimates of the CO sources by 60% in some regions, but that as a result of the limited information from the observation network it is difficult to separate anthropogenic and biogenic sources of CO. Furthermore, they show that assumptions about the OH distribution and the injection heights of the CO emissions from biomass burning can have a large impact on the source estimates. Although the atmospheric chemistry of CO is simple, there are still large uncertainties in the regional top-down estimates of the CO sources. Until there is a coordinated model intercomparison to develop a comprehensive understanding of the

impact of model and data errors and the top-down CO source estimates, studies such as this provide valuable insight into the possible factors behind the discrepancies in the top-down source estimates. The manuscript is well written and is appropriate for ACP. The authors have presented a detailed sensitivity analysis of the inversion system and I have no major concerns with the analysis. However, they are a number of places, noted below, where I think the discussion needs clarification. I recommend publication of the manuscript after the authors have addressed these comments.

Specific Comments

1) Page 344, lines 10-12: The statement that "the main sink of CO is the reaction with OH, the co-called cleansing agent of the atmosphere" is somewhat redundant with the first sentence, where it is stated that "by reaction with OH, CO influences the oxidizing capacity of the atmosphere."

2) Page 347, line 3: The reference for Fisher (1998) here is to an ECMWF seminar. Is this really a valid reference? Also, there is a typo in the date of the reference on page 370. The conjugate gradient approach was introduced decades ago, I would think that there are much more appropriate references for this than Fisher (1998).

3) Page 347, lines 11-15: I don't have access to the ECMWF technical report for Fisher and Courtier (1995). How is the Hessian approximated? What is the accuracy of the approximation –i.e. how well does it converge to the true a posteriori covariance? More information would be helpful here since the source uncertainty discussion requires confidence in the Hessian calculation.

4) Page 348, lines 12-15: What is the global, annual mean OH concentration? What is the estimated methyl chloroform lifetime?

5) Page 350, lines 20-23: The authors claim that their error estimates of 20-48% and 58-72% are realistic for the Western developed world and the developing world, respectively. Do they have a reference for this claim?

6) Page 351, line 3: On what analysis is the 1000 km correlation length scale based? Is there a reference for this?

7) Page 351, lines 3-6: The authors mentioned that they do not expect a pronounced seasonal cycle for anthropogenic emissions. Although this is typically assumed, Petron et al. (GRL, 2004) found that emissions of CO from fossil fuel and biofuel combustion were 30% and 200% greater, respectively, in winter than in summer. Similarly, The a posteriori results of Kopacz et al. (JGR, 2010) show a significant seasonality in CO emissions from North America and Asia. They found that North American emissions were 50% larger in winter than in summer, while Asian emissions were almost a factor of 2 larger in winter than in summer. It would be valuable if the authors were to assess the impact of their assumed temporal correlation on their results. Would a much shorter e-folding timescale significantly change the regional estimates?

8) Page 352, line 1: On what is the assumed 1.5 ppb measurement error based? Is there a reference for this?

9) Page 352, lines 3-4: A brief explanation of the model error approach used in Bergamaschi et al. (2010) would be helpful for the reader, especially since the Bergamaschi et al. analysis was for CH4 and this paper is focused on CO. A Figure similar to Figure 2 (top panel) of Bergamaschi et al., showing the representativeness error and the overall data uncertainty for a couple of selected stations would be helpful.

10) Page 352, lines 15-21: I do not understand the justification for throwing out 15-20% of the data as outliers. If those observations represent particular pollution events that the model cannot capture because of the coarse resolution, as the authors claim, then the representativeness errors should account for this. This seems quite arbitrary to me and represents a significant weakness in the analysis. Omitting this much data needs better justification.

11) Table 1: Although removing the outliers results in a better goodness of fit for the a posteriori CO field, removal of the outliers increases the mean a priori bias. For all

the stations shown in Table 1, the bias goes from -0.58 ppb to 1.53 ppb. For individual stations such as Alert (ALT), the bias increases from 0.94 ppb to 2.78 ppb. Again, what is the justification for removing the outliers given that doing so means that you are starting the inversion from a more biased a priori state? It would be helpful to see what is the impact of removing the outliers on the inferred sources – i.e. how do the regional source estimates compare in inversion cycles 1 and 2?

12) Page 353, lines 22-26: Large amounts of satellite data would not necessarily create a situation in which the inversion is not strongly dependent on the a priori. Ultimately, it will depend on the precision of the satellite data. Large amounts of imprecise data are not necessarily better than sparse but precise data.

13) Figure 3 and Table 2: With the exception of Asia and Europe, there is little reduction in the uncertainty of the anthropogenic emissions. However, Kasibhatla et al. (GRL, 2002) showed much greater uncertainty reduction for fossil fuel emissions from North America, Europe, and Asia in their inversion analysis of the surface CO data. The authors should comment on why their inversion results are so different from those of Kasibhatla et al. (2002).

14) Page 358, lines 13-17: The authors claim that the overestimate in May and June is due to an overestimate of the NMVOC source in the a priori, since the inversion particularly reduced the NMVOC in these months. However, as the authors acknowledge on page 355, lines 23-46, the data do not constrain the NMVOC source well. Furthermore, a tight a priori constraint was imposed on the NMVOC source in the inversion. As a result, the NMVOC source should have remained close to the a priori. Also I find it suspicious the bias is confined to just May and June, given that the summertime maximum in the NMVOC source is broad and does not peak as early as May. It seems likely that the large reduction in the NMVOC source reflects the impact of other biases being projected onto the NMVOC source. Indeed, the authors noted on page 361, lines 15-18, that the decrease in the inferred NMVOC source from 2003 to 2004, could be an artifact of the inversion.

15) Page 359, lines 14-16: I disagree with the claim that the "comparison with NOAA aircraft profiles showed however that the vertical transport in TM5 is reasonable." The aircraft data were mainly in the northern hemisphere, whereas the bias with respect to MOPITT is pronounced in the southern hemisphere (at middle and high latitudes). In the absence of more independent aircraft data in the southern hemisphere, over a range of longitudes, one cannot rule out a bias in vertical transport in the model.

16) Page 359, lines 18-19: It is not clear to me how any putative issues with the MOPITT retrievals over deserts is relevant here? Indeed, the bias seems to be larger on land over the deserts, where one would expect the retrievals to be challenging, but the model-MOPITT bias is largest over the oceans in the southern hemisphere. Furthermore, the CO inversion of Jones et al. (ACP, 2009) showed that both TES and MOPITT data resulted in an overestimate of surface CO in the midlatitudes of the southern hemisphere. It is unlikely that the TES and MOPITT data are similarly biased. Can the authors comment on this? My guess is that the model used here as well as the model used in Jones et al. are biased in their vertical transport.

17) Page 361, lines 23-26: Are the correlation coefficients of -0.29 and -0.23 statistically significant?

18) Page 363, Section 5.1: It would be helpful to show how the regional estimates respond to the different sensitivity tests. Showing only the global totals in Table 5 is less informative.

19) Tables 4 and 5: The error for the NMVOC-CO source in Table 5 is 10% for S3 (81 out of 812 Tg CO), whereas in Table 4 the error is listed as 16%. Which is correct?

20) Page 364, lines 1-4: I do not understand the discussion here. What do the authors mean when they say that the "in sensitivity study S1 the NMVOC-CO prior error dominates, resulting in less reduction in this source"?

21) Page 366, line 3: Please change "increase with 75 Tg CO" to "increase by 75 Tg

CO".

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