Atmos. Chem. Phys. Discuss., 8, S7822–S7827, 2008
www.atmos-chem-phys-discuss.net/8/S7822/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sources of carbon monoxide and formaldehyde in North America determined from high-resolution atmospheric data" *by* S. M. Miller et al.

S. M. Miller et al.

Received and published: 1 October 2008

We would first like to thank the reviewers and editor for their time and effort: we appreciate any comments to help make the manuscript better, and we are grateful for the recommendation for publication. We have made changes to the manuscript below to reflect comments and suggestions made by the reviewer. These changes will be included in future versions of the manuscript submitted to the editor.

Comments:

Comment 1: We have included a table in the results sections that lists the RMSE and correlation coefficient for the STILT-CO model at Argyle during summer months, WLEF during summer months, and WLEF during spring months. To make the results section

easier to read and interpret, we have also removed the posterior scaling factors for the model from the text itself and have put the inversion results instead in a table.

P11396 – Much of the focus of the paper is for emissions over the United States, and therefore we feel it is appropriate and important to at least briefly mention the US policy context. We have clarified the section to hopefully make the passage easier to read for those not familiar with US air management policies. We have changed the text as follows: "Carbon monoxide is a key species for both atmospheric chemistry and public health. In the United States, it is one of the original six criteria air pollutants in the Clean Air Act of 1970, and many urban areas remain either in violation of ambient CO air quality standards or at risk of violation (US EPA, 2007b). Effective emissions control strategies require accurate emission inventories and models that can forecast concentrations across the North America."

P11397, line 10; P11398, line 4: This has been changed in the text (see the responses to other comments).

P11400, line 15: We feel that the lateral tracer boundary condition as used in the current STILT-CO model better represents the boundary than would using a global CTM and makes the model more stand-alone. The STILT-CO model includes a boundary condition for CO and CH4 but not for HCHO. CO has an average lifetime of approximately two months (Pfister et al., 2004). Some CO will therefore decay in transit from the boundary condition, but this decay has been easily incorporated into the model. The boundary condition within the STILT model has the advantage of being based upon several empirical measurement stations. In contrast, the GEOS-Chem model has a significant global offset (20 ppb) during some seasons at northern latitudes. STILT model results cannot be thrown off by any erroneous modelled events or offsets that could occasionally be given by a CTM. The use of an empirical boundary condition also makes the STILT model more stand-alone or self sufficient. We do not want the STILT model results to be entirely the product of the results given by a CTM.

ACPD

8, S7822–S7827, 2008

P11402, line 23: We agree with the reviewer's suggestion for Equation 4. We have changed this equation in the text as per the reviewer's suggestion.

P11404: We do neglect natural CH4 emissions in the model. For an in-depth discussion of natural methane sources, please refer to point (2) in the reply to Referee #3. Hudman et al. (2008) estimate that anthropogenic NMVOCs account for 12.1% of the total CO source over the continental United States. Most of the anthropogenic VOC source is over urban areas. We use receptor points that are in relatively rural areas. The short lifetime of many VOCs (particularly isoprene) means that most anthropogenic NMVOC emissions will decay before reaching the tower. Therefore, we expect a model error from the exclusion of anthropogenic NMVOCs likely in the range of ˜2-4ppb for CO. A caveat to this effect has been added to the paper.

P11413, line 11 and further: The number of particles used in simulations does seem to make a larger difference (up to ˜20ppb) for higher modeled PM concentrations. We ran model simulations at Argyle Tower during the summer months using both 100 and 500 particles. 500 particle simulations at the Argyle Tower do appear to smooth out some possible noise in the posterior model result. However, the 500 particle simulations do not necessarily improve model-measurement fit for Argyle for the first two weeks of the summer (We have added a graphical display of these Argyle results to the final paper). STILT simulations with 500 particles entail enormous computational costs. Results from Argyle Tower suggest that any increases in model performance are far outweighed by the increased computational cost.

P11414, line 3: We agree with the reviewer's suggestion and have changed the figure accordingly.

P11414, line 14: We have included in the final paper a table that displays the correlation coefficient and RMSE of posterior model results at WLEF using the different emissions inventories displayed in Fig. 9.

P11417, line 11: The reviewer makes an interesting point. Heating and power gener-

ACPD

8, S7822–S7827, 2008

ation would likely also account for the majority of the increase in CO emissions during the wintertime over North America. The EPA-1999 inventory might overestimate the relative increase in non-road sources from winter to summer (i.e. tractors, diesel from construction, etc.). Alternately, the EPA-1999 inventory could also underestimate the relative increase in area sources (home heating, fireplaces, etc.) or increases in CO emissions from power generation from summer to winter. We have added the following explanation to line 7: "The EPA-1999 inventory might overestimate the relative increase in non-road sources from winter to summer (i.e. tractors, diesel from construction, etc.) and/or underestimate the relative increase in area sources (home heating, fireplaces, etc.) or increases in CO emissions from power generation from summer to winter."

P11417 Section 3.3: We have changed Section 3.3 in order to reflect the reviewer's suggestions. The last paragraph of the section has been re-written as follows:

"Issues affecting very large biomass sources very far away represent a major challenge to any modelling framework, and their resolution lies beyond the scope of this paper. The lack of pyro-convective injection in the model might, in part, account for why the model performed very well on relatively small fires in the near field but showed mixed performance in capturing the influence of very large fires at a long distance. Time periods affected by long distance biomass burning emissions are not included in the assessment of source inventories."

P11419, section 3.5.1: The STILT model at Argyle Tower often detects periods of large anthropogenic emissions influence that are not measured at the tower. Similarly, the model often misses episodes of high pollution that are likely caused by the influence of emissions from urban areas on the East Coast. These false pollution episodes are independent of changes in the modeled boundary condition. We have added to the final paper an illustrative graphic from July 17, 2004 at Argyle Tower, a period when the model misses a high concentration event. Air parcels traveling from Argyle stagnate in a low pressure system over the Atlantic Ocean. Most likely as a consequence, the influence footprint misses large pollution sources in either Boston or New York

ACPD

8, S7822–S7827, 2008

City. Problematic modeling periods like July 17 could very well be due to problems in modeling the planetary boundary layer (PBL). For example, the modeled PBL over the ocean may be too low on July 17. If this were the case, the modeled particle ensemble may not have been sufficiently dispersive. The modeled particles may miss large coastal sources because of some other problem in the modeled meteorological fields. We have added a sentence to the second paragraph of section 3.5.1 to suggest that modeled PBL could be a possible source of the problem as well.

P11421, line 8: Hudman et al. (2008) predict a higher fraction of CO from NMVOCs than does this paper. However, there are a few important differences. First, Hudman et al. (2008) include methane sources in boundary condition calculations and do not include methane when determining the relative importance of different sources. Secondly, Hudman et al. (2008) include the entire continental United States in their source calculations. Therefore, their calculations include high biogenic NMVOC source regions such as the southeastern US. Our results, in contrast, represent the upper Midwest, a region with much lower biogenic NMVOC emissions. Page 11415, lines 20-25, list the relative uncertainties associated with the posterior model sources. These uncertainties provide an idea of the possible margin of error in the source budget calculations. For example, there is a reasonable range of uncertainty in the posterior VOC inventory scaling factor. Errors in modelled chemistry would likely be corrected or adjusted by the posterior inventory scaling factors. Therefore, uncertainties in the posterior inventory scaling factors best represent uncertainties in the budget calculations.

P11422, line 7: STILT-CO shows high model-measurement fit for the WLEF tower and for INTEX-A aircraft flights. Even though the model shows relatively poor performance at one of the receptor sites (the Argyle Tower), we believe this conclusion is still warranted.

P11396, line 8-9: We have changed line 9 to read "from a tall tower." We believe that the model capability to match hourly data merits the description "high fidelity", notwithstanding the fact that not all receptors are simulated with such accuracy.

ACPD

8, S7822–S7827, 2008

Interactive Comment

P11397, line 18: This reference has been fixed in the text.

p11404, Line 23: We have changed this citation in the text.

p11405, Line 15: We have changed this citation in the text in accordance with the referee's suggestions.

P11409, line 11: This parenthetical citation has been fixed in the text, as suggested.

P11410, line 2-4 : We have fixed this typo.

P11421, line 22: We have fixed this typo.

References

Hudman, R.C., Murray, L.T., Jacob, D.J., et al: Biogenic vs. anthropogenic sources of CO over the United States, Geophys. Res. Lett., 35(4), L04801, doi:10.1029/2007GL032393, 2008.

Pfister, G., Petron, G., Emmons, L.K., et al: Evaluation of CO Simulations and the Analysis of the CO Budget for Europe, J. Geophys. Res., 109, D19304, doi:10.1029/2004JD004691, 2004.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 11395, 2008.

ACPD

8, S7822–S7827, 2008

Interactive Comment