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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.

Anonymous Referee #3

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General comments:

This study attempts to apply the model-data assimilation approach using the STILT model to optimize the local/regional sources of CO from anthropogenic emission, biogenic VOCs and biomass burning. Using the STILT framework to optimize CO sources is a novel approach and may provide new insights and constraints on CO sources. The model includes the chemical breakdown of biogenic VOCs and CH4 to HCHO and then CO, as well as the reaction of CO to CO2. The model-data assimilation was applied to the data from two tower sites and aircraft measurements. The results showed some agreement for the WLEF tower site and poor agreement at the Argyle tower site. This

study based on limited data (essentially from the WLEF site) is useful in illustrating the extension of the STILT model to another compound and give regional flux estimates. Therefore this study is of interest to the scientific community and worthy of publication, however, there are many (possibly major) questions on the model and source functions with possible (major) impact on the results, hence the questions should be addressed (possibly with new model simulations and model-data assimilations) before this manuscript is accepted.

The following are comments in addition to those from the other reviewer.

Specific comments:

The authors make the (acceptable) distinction between VOC and CH4. In their usage, VOCs refer to "biogenic VOCs"; (the distinction is noted at numerous places: page 11397, lines 2-3 and line 25, "methane and VOC"; page 11405, line 25, VOC refers to isoprene, monoterpenes, acetone and higher order alkenes; and page 11047, line 5 states "In addition to VOC chemistry, the model incorporates chemistry from CH4 loss ..."). This review accordingly assumes that CH4 and VOC are distinct and treated separately.

The CO sources used in this study (Section 2.1.4, pp 11404-11405) are: NEI-1999 for anthropogenic CO, MEGAN for VOC, EDGAR 1995 for anthropogenic emission of CH4 and satellite estimates of biomass burning from Wiedinmyer et al. (2006). Are CO emissions zero for Canada and Mexico in NEI-1999 and hence in the model? Although anthropogenic CO emissions may be smaller from Canada and Mexico than from the United States, they are not negligible. Are non-anthropogenic CH4 sources ignored (wetland is a significant CH4 source in the growing season)?

The results of this study raise questions about the initial assumptions of the study, particularly the assumption that the major sources of CO are fossil fuel combustion, biomass burning and biogenic VOC oxidation. At least at the WLEF site, Section 3.6 reports: the decomposition of CH4 contributes 35.3% to CO concentration, compared

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to 31% for anthropogenic emission, 21.2% for biogenic VOCs and 12.7% biomass burning. These results if true, would suggest that CH4 is a major source of CO and the model should be modified to include all (not only EDGAR) significant CH4 sources in the source optimization procedure.

Pages 11400, lines 24-25 states "Because the lifetime of formaldehyde is a few hours or less, we set formaldehyde to zero at the boundary." Can the breakdown of CH4 and acetone (with long lifetimes) to HCHO be ignored at the boundary?

Page 11402, lines 22-23, Equation 4, if the 2nd term represents the chemical change (through HCHO and hence HCHO chemistry is implicitly included in Equation 4) of VOC to CO (since Fb is flux of VOC according to line 12), then I do not understand the meaning of "An analogous approach is taken for formaldehyde" (lines 18-19). The 3rd term accounts for the CO loss due to chemistry. Why does the loss apply only to the first term (direct CO emissions at the surface with flux Fa) and not the 2nd term (CO from VOC)? This model description is quite confusing, raising doubts on the validity of the model.

Page 11406, Equations 5 and 6, Equation 6 is the solution for Equation 5 (I believe there is a minus sign missing in Equation 6) for the special case with no VOC addition or flux for $t>0$, and no transport (similar problems for Equation 8). A clearer and more complete description of the chemistry model is needed here to show how flux and transport are treated.

Page 11413, lines 14-15, "we tested model simulations using both 100 and 500 particles to determine the number required for accurate simulations." The difference appears small for the WLEF area, what is the effect of more particles for more difficult sites like Argyle (where model performance is poor)?

Page 11414, lines 26-27, " ... optimized only for biomass burning scaling factors", what is the resultant biomass burning scaling factor?

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Section 3.3: The discussion on biomass burning and STILT-CO is a bit confusing. Figure 15 claims to show good simulation of the CO peak on Aug 17, 2004 at WLEF from distant forest fires "near Great Slave Lake, in the Yukon Territories and in eastern Alaska", then next paragraph states: "The lack of pyro-convective injection in the model may account, in part, 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". Where are the results showing "model performed very well on relatively small fires in the near field"?

Page 11451, Figure 20. The top figure shows CH4 in yellow color, is this an error (as the figures below use yellow for "other VOC")?

Technical comments and corrections

Page 11396, lines 8-9, "In most cases, the model demonstrates high fidelity simulations of hourly surface data from tall towers ..", this description appears exaggerated. "The model simulation shows some agreement with hourly data from a tall tower .."; may be a more suitable description.

Page 11399, line 20: add "biomass burning" to the list of sources treated by the Bayesian optimization for completeness and consistency.

Page11401, line 9 and page 11402, line 8: delete "incremental", since "concentration changes"; are not always incremental. Also on page 11402, line 6, replace "concentration increment" with "concentration change".

Page 11402, lines 5 and 7, replace "t"; with the Greek letter tau; for the time step of the model to be consistent with Equation 3. Also replace "yi"; with "yj"; on line 7.

Page 11402, line 13, replace "tr" with "t";.

Page 11402, line 20, "mair"; is not defined.

Page 11403, line 3, delete "they".

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Page 11405, line 12, "midday ... over North America", North America has many time zones, does midday refer to WLEF site?

Page 11405, line 23, to to.

Page 11406, line 14, change "HCHO" to "HCHO and CO".

Page 11410, line 1, the subscript "2" should be superscript.

Page 11413, line 2, change "half" to "third".

Page 11414, line 23, change "MEGHAN" to "MEGAN" and "Co" to "CO".

Page 11414, line 20, what is the exact time period for "early summer"?

Page 11414, line 24, what is the exact time period for "summer"?

Page 11420, line 7, delete "be".

Page 11421, line 13, Hudman et al. (2008) Hudman et al. (2008)

Page 11421, line 22, "VOC (CH4 and biogenic compounts)" CH4 should be separated from VOC to be consistent with the rest of the manuscript. Change "compounts" to "compounds".

Page 11422, line 4, change "a variety of different tall tower sites" to "a tall tower site".

Page11422, line 25, change "ahd" to "and".

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