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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.

S. M. Miller et al.

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We would like to thank the reviewers and editor for their time and effort. 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. The reviewer's concerns are mainly questions of clarity in the exposition, not major omissions from the model (e.g. Canadian/Mexican sources, treatment of the chemistry), which we have rectified with appropriate changes in wording to be included in the final paper.

Specific comments:

1) The NEI–1999 inventory includes CO sources both from Canada and Mexico. Cana-

dian sources, for example, can significantly influence CO concentrations at the WLEF tower as modeled by STILT. The top panel of Fig. 6 shows a time period in which CO concentrations are significantly influenced by urban emissions from Chicago, Detroit, and Toronto. We have corrected the manuscript to clarify that both sources from Canada and Mexico are included in the model. Beginning with line 16 on page 11404, the manuscript will read: "The STILT–CO model utilizes a variety of different emissions inventories for the purpose of comparing different source estimates. This paper primarily relies upon the US EPA's 1999 National Emissions Inventory (NEI–1999) for anthropogenic CO and formaldehyde emissions over the US, Canada, and Mexico (US EPA, 2004; Frost and McKeen, 2007)."

2) The EDGAR methane inventory includes emissions from fossil fuels, biofuels, waste production, animals, agriculture, and human waste (ex: landfills and wastewater treatment) (Netherlands EAA 2005). Natural wetland sources were not included in the inventory. Kort et al. (submitted) used STILT to model methane emissions for COBRA–North America aircraft measurements taken during the summer of 2003. Kort et al. (submitted) found that natural wetland sources over North America contributed to only 3% of the total model enhancement (the model prediction minus the advected boundary condition). In our simulations, methane influences CO and HCHO concentrations predominantly through the decay of the methane boundary condition. During the summer months at WLEF, approximately 97.5% of all methane–derived CO came from the methane boundary condition while only 2.5% of all methane–derived CO came from continental methane sources. Similarly, 94% of all methane–derived formaldehyde came from the methane boundary condition while only 6% of all methane–derived HCHO came from continental methane sources. Local variations are small because the lifetime of CH_4 is greater than 10 years. Therefore it is important and useful to model variations in CH_4 emissions in the domain, but STILT does not need to represent every local variation. In light of results from Kort et al. (submitted), we estimate a possible uncertainty of 3–5% in the boundary layer near wetland source regions. A caveat to that effect will be added to the paper.

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3) Pages 11400, lines 24–25: We assume that formaldehyde (HCHO) is equal to zero at the boundary because it has a very short lifetime. HCHO at the boundary decays before reaching the North American continent. However, we do include the breakdown of the advected CH_4 boundary condition as an HCHO source. We do not include an advected acetone boundary condition in the model. First, acetone is a very small source of CO and HCHO in the model (0.3% and 0.5% respectively); including an acetone boundary condition would have a negligible impact on the results. Second, most acetone from Asian sources likely decays prior to reaching North America, and any chemistry or ocean source is likely very small. Millet et al. (2004) measured a range of VOCs at Trinidad Head, CA. They found an average acetone concentration of 0.6ppb from air transported over the Pacific. If this acetone took four days to reach the WLEF tower and if the resulting HCHO had a constant lifetime of four hours, then the approximate HCHO contribution from an acetone boundary condition would be 0.07–0.08ppb. In order to clarify this point in the text, we have changed page 11404, line 9 as follows: "The formaldehyde model incorporates HCHO from anthropogenic formaldehyde sources, from the decay of biogenic VOCs, and from methane decay (both from continental sources and from the methane boundary condition). We do not include any VOC boundary conditions. Measurements from Millet et al. (2004) provide an estimate of VOC concentrations in air at Trinidad Head, CA , advected from the Pacific Ocean. We estimate that the lack of an acetone boundary condition within the model contributes 0.07–0.08ppb uncertainty in formaldehyde model results."

4) Page 11402, lines 22–23, Equation 4: The model does include the decay of CO from all CO sources, not just CO from direct emissions. We have corrected Eq. 4 as follows and changed the description of the equation accordingly:

$$\Delta C_{CO}(\vec{x}_r, t_r) \; = \; \Sigma_{i,j,m}\{f(x_i, y_j, t_m)F_{CO}(x_i, y_j, t_m)$$

$$+f(x_i, y_j, t_m)F_{[VOCs,CH_4]}(x_i, y_j, t_m) \int_{t_m}^{t_r} R(x_i, y_j, t_m|\vec{x}_r, t)dt -$$

$$f(x_i, y_j, t_m)F_{[CO,VOCs,CH_4]}(x_i, y_j, t_m) \int_{t_m}^{t_r} k_{OH}[OH]dt\} \quad (1)$$

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In addition, we also add an equation after Eq. 4 to more clearly describe how formaldehyde concentrations are calculated:

$$C_{HCHO}(\vec{x}_r, t_r) \; = \; \Sigma_{i,j,m}\{f(x_i, y_j, t_m)F_{HCHO}(x_i, y_j, t_m)$$

+f(x_i, y_j, t_m)$F_{[VOCs,CH_4]}(x_i, y_j, t_m) \int_{t_m}^{t_r} R(x_i, y_j, t_m|\vec{x}_r, t)\,dt$

-f(x_i, y_j, t_m)$F_{[HCHO,VOCs,CH_4]}(x_i, y_j, t_m) \int_{t_m}^{t_r} j_{HCHO}\,dt\}$(2)

We have deleted the phrase "An analogous approach is taken for formaldehyde," (page 11402, line 18–19) and instead have added the following description: "Equation 5 describes the similar approach taken for formaldehyde. The HCHO signal at the tower from surface sources $(\Delta C_{HCHO}(\vec{x}_r, t_r))$ equals the influence of formaldehyde surface sources (1st term), the influence of VOC and CH_4 surface fluxes that decay to formaldehyde (2nd term) and the decay of formaldehyde given by the decay rate j_{HCHO} (described in more detail in section 2.1.5) (3rd term). The emissions from each back trajectory location and time (x_i, y_i, t_m) are summed to find the influence of advected continental sources $(\Delta C_{HCHO}(\vec{x}_r, t_r))$."

4) Page 11406: Eq. 6 is the solution to Eq. 5, and Eq. 8 is the solution to Eq. 7. In order to make this clearer, we have changed page 11406, line 19 as follows: "Eq. (5) describes the decay of VOCs to HCHO (1st term) and the decay of HCHO to CO (2nd term). Solving Eq. (5) for HCHO gives an expression (Eq. 6) for the increment of HCHO at the receptor after the gases have been transported for time t. Similarly, Eq. (7) expresses the creation of CO from decaying formaldehyde (1st term) and the loss of CO to oxidation (2nd term). Solving this equation produces Eq. 8: the increment of CO at the receptor after gases have been transported for time t from the source (x_i, y_j, t_m) to the receptor $(\vec{x}_r, t_r)$." The equations include both VOC addition and transport of pollutants from the source to the receptor. Additionally, there is a typo in Eq. 6: in the second half of the equation, k_1 and j_2 should switch positions. This has been corrected in the manuscript.

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5) Page 11413, lines 14–15: We ran model simulations at Argyle Tower using 500 particles - a graphical time series of these results has been added to the paper. Simulations with 500 particles at Argyle Tower appear to display less noise but do not improve model–measurement fit. We have added the following sentence to line 26 on page 11413: ”In addition, we ran 500 particle simulations at Argyle Tower but found no improvement in model–measurement fit over 100 particle simulations.”

6) Section 3.3: The model performs relatively well for fires in the near field. During the year 2004, most emissions influence at WLEF Tower from forest fires came from Canada. However, simulations from the spring months (see Fig. 12) report influence from continental sources over Missouri and Arkansas. Although these biomass burning sources are relatively small, the inclusion of these sources affords a better model–measurement fit. These biomass burning events are described from line 16, pg. 11417 to line 2, pg. 11418: ”The time series from WLEF Tower in spring 2004 (Fig. 12) shows that even during the spring months, biomass burning can substantially infuence pollution levels at the tower site. The influence of biomass burning events in Missouri and Arkansas were accurately characterized by STILT–CO during this time period.”

7) Page 11451, Figure 20: The yellow color on the top panel of the figure is not an error. However, we have changed this color in the top panel from yellow to grey in order to match the color coding in the lower panels. This will hopefully make the graphic less confusing for readers.

Technical Comments and Corrections

Page 11396, lines 8–9: 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.

Page 11399, line 20: We feel it is best not to include biomass burning in this sentence. The model does not produce a reliable scaling factor for biomass burning emissions. Because we do not use the Bayesian inversion to refine biomass burning sources, we

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do not feel it would be appropriate to list biomass burning in this sentence.

Page11401, line 9 and page 11402, line 8: We have deleted the word ”increment” from pg. 11401, line 9, pg. 11402, line 6 ,and line 8.

Page 11402, lines 5 and 7: We have made the suggested change.

Page 11402, line 13: We have made the suggested change.

Page 11402, line 20: m_{air} is the molar mass of air. We have added the following sentence to line 8, pg. 11402: ”m_{air} is the molar mass of air.”

Page 11403, line 3: We have made the suggested change.

Page 11405, line 12: For this graphic, we plotted mean July VOC emissions for 1pm CST. This equates to 2pm EST or 11am PST. We have clarified this point in the text (page 11405, line 12) as follows: ”Figure 1 displays a map of mean midday biogenic VOC fluxes (at 1pm CST/11am PST) over North America from 1 June to 15 August from the MEGAN inventory (Millet et al., 2004; Hudman et al., 2008).”

Page 11405, line 23: We have corrected the typo.

Page 11406, line 14: We have made the suggested change.

Page 11410, line 1: We agree with the reviewer and have made the suggested change.

Page 11413, line 2: We have made the suggested correction.

Page 11414, line 23: We have corrected these typos.

Page 11414, line 20: We have changed this line as follows: ”For early summer (June 1 – July 23)....”

Page 11414, line 24: We have changed the text as follows: ”The scaling factor for VOC emissions in summer (June 1 – July 23)....”

Page 11420, line 7: We have corrected this mistake in the text.

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Page 11421, line 13: We have corrected this typo.

Page 11421, line 22: We have made the changes suggested by the reviewer.

Page 11422, line 4, has been changed to ”at a tall tower and from aircraft.”

Page11422, line 25: We have made the change as suggested.

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