Atmos. Chem. Phys. Discuss., 8, S7709–S7717, 2008
www.atmos-chem-phys-discuss.net/8/S7709/2008/


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: 30 September 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.

Page 11369, Line 20: We changed the word "was" in this line to "is" in order to avoid confusion.

Page 11397, Line 10: The latest IPCC (2007) report says,

"Other significant precursors for tropospheric ozone are CO and NMVOCs, the most important of which is biogenic isoprene. Satellite measurements of CO from the Measurements of Pollution in the Troposphere (MOPITT) instrument launched in 1999 (Edwards et al., 2004) have provided important new constraints for CO emissions, pointing in particular to an underestimate of Asian sources in current inventories (Kasibhatla et al., 2002; Arellano et al., 2004; Heald et al., 2004; Petron et al., 2004), as confirmed also by aircraft observations of Asian outflow (Palmer et al., 2003a; Allen et al., 2004)" (549).

In light of the latest IPCC report, we have revised the line in question as follows in order to better reflect the exact content of the report: "Despite a long history of emissions estimates, substantial uncertainty remains in knowledge of carbon monoxide sources. IPCC (2007) indicates that remote sensing efforts have helped constrain CO emissions, but several recent studies suggest that EPA's 1999 National Emissions Inventory (NEI-1999) may overestimate anthropogenic CO emissions by 50% (Parrish, 2006; Turnbull et al., 2006; Warneke et al., 2006; Hudman et al., 2008)."

Page 11397, Line 16-18: This passage does, in fact, refer to global emissions. We have changed this sentence to read as follows: "VOCs are emitted by anthropogenic and biogenic sources, but biogenic VOC emissions, particularly isoprene and monoterpenes from plants, constitute 80% of the total global source (Oliver et al., 2001)."

Page 11398, Line 4: The newest IPCC report does not detail the importance of biomass burning as a CO source. However, several other studies released since the 2001 IPCC report make estimates of the contribution of biomass burning to global CO emissions. Petron et al. (2004) estimate that biomass burning accounts for ˜15% of the global CO source. Müller and Stravrakau (2005) estimate 12%, Arellano and Hess (2006) predict ˜20%, and Duncan et al. (2007) estimate ˜20-23%. In order to include the findings of more recent papers, we have changed the passage as follows: "Sources of CO from biomass fires are also poorly constrained. Biomass burning contributes 15-30% of all global CO emissions (IPCC 2001; Petron et al., 2004; Mueller



**ACPD**

8, S7709–S7717, 2008



and Stravrakau 2005; Arellano and Hess, 2006; Duncan et al., 2007)."

Page 11398, Line 15: On page 3430, Wiedenmyer et al (2006) explain that "inherent uncertainties" limit knowledge of emissions factors. In order to recognize this source of error in fire estimates, we have modified the last sentence of the paragraph as follows: "Uncertainties in emissions estimates arise from uncertainties in fuel loadings (estimates of biomass per area), in emissions factors (volume of emissions per mass burned), in combustion efficiency (fraction of biomass burned), and from the inability of satellites to detect fires through cloud cover (Wiedinmyer et al., 2006)."

Page 11398, Line 25: In response to the reviewer's comment, we have revised the sentence as follows: "The background arises because CO has an atmospheric lifetime of about two months - enough time to transport the pollutant over long distances, but not enough time for the pollutant to build up to very high levels in the absence of intense localized sources (Pfister et al., 2004)."

Page 11399, Line 12: We have changed this sentence as per the suggestion of the reviewer. The sentence now reads, "The present paper describes the Stochastic Time-Inverted Lagrangian Transport Model for CO (STILT-CO)...."

Page 11403, Line 7: We have corrected this typo in the paper.

Page 11403, Line 15: The mass violation derives mostly from the output functions of the meteorological models: winds are not given at each grid point and time step, and are often interpolated. The problem is largely solved by modifying the model to report mass fluxes on the native grid, averaged over the time between model outputs. We feel that discussion of this problem is beyond the scope of the current paper.

Page 11404, Line 13: HCHO loss rates in the STILT-CO model are taken from GEOS-CHEM a 2-hourly global grid. The loss rates include photolysis and loss via the hydroxyl radical but do not include HCHO deposition. Using a photochemical dispersion model for northern Italy, Liu et al. (2007) found that dry deposition of HCHO was unimportant



**ACPD**

8, S7709–S7717, 2008



compared to other processes; the change in HCHO concentration brought about by deposition was usually two orders of magnitude less than the effect of chemistry. Because HCHO deposition is generally very small, we assume that any errors that caused by excluding HCHO deposition will also be negligible. We do not consider uptake or emission of CO by soils; uptake is a very slow process occurring at high concentrations, and emissions are weak (Conrad and Seiler 1982).

Page 11404, Line 17: The EPA has released a more recent national emissions inventory, EPA NEI-2002 (EPA 2008). EPA has also produced several reduced form emissions inventories through 2006 (EPA 2007a). We use the NEI-1999 inventory because it allows direct comparison with a wide range of other studies that have also used this same inventory (ie: Parrish 2006, Warneke et al. 2006, Hudman et al. 2008). Additionally, in comparison with the inventory error estimated by this study, EPA's emissions estimates have not changed significantly since 1999. EPA estimates 1999 total national emissions at 114,541 short tons of CO and 2006 emissions at 100,552 tons, mostly due to reductions in emissions from highway vehicles (EPA 2007b). Our study demonstrates that EPA overestimates 1999 emissions by up to a factor of three. In order for the 2006 inventory to be correct, actual CO emissions would needed to have increased by a factor of 2.6 between 1999 and 2006; this is surely not the case. In order to incorporate data from the 2006 inventory, we have changed the passage as follows: "This paper primarily relies upon the US EPA's 1999 National Emissions Inventory (NEI-1999) for anthropogenic CO and formaldehyde emissions (US EPA, 2004; Frost and McKeen, 2007). Newer less detailed EPA inventories through 2006 are relatively similar in magnitude: EPA estimates that 2006 emissions are a modest 14% lower than in 1999, due mostly to reductions in highway vehicle emissions (EPA 2007b)."

Page 11407, Line 15: Lagrangian models like STILT estimate trace gas concentrations at much finer temporal and spatial scales than large-scale Eulerian models. This creates a highly coupled and transparent source-receptor relationship. Lagrangian models can therefore more easily identify both spatial and scalar inaccuracies in the emissions



**ACPD**

8, S7709–S7717, 2008



inventory, especially at the regional level. This can be a main advantage over large-scale Eulerian models.

Page 11409, Line 11: There is a "chain of custody" for carbon atoms such that uncertainties in the CO yield directly translate into uncertainties in the inferred fluxes. This type of uncertainty is not properly included in a Bayesian framework because it is not random - it is a single number affecting only the scaling of the prior, not its error covariance. We have added the following sentence to line 14 in order to clarify this issue: "We do not include uncertainty from CO/HCHO yields or chemistry in Sprior because this error is not random - it is a single number affecting only the scaling of the prior, not its error covariance."

Page 11415, Line 22: "95% CI" refers to the 95% Confidence Interval. We have clarified this in the text.

Page 11416: We had intended to point out the discrepancy between the EPA-99 regional scaling factors and the scaling factors derived from our Bayesian inversion. The EPA-99 inventory estimates an increase in anthropogenic CO emissions from early spring to summer, but our inversion results suggest the opposite to be true. In order to better clarify this point and clear possible confusion, we have modified the passage as follows: "The NEI-99 inventory data is only available for typical mid-week summer days and typical mid-week winter days. We use the summer inventory as the a priori for all simulations. We note that our scaling factor for the NEI-1999 inventory for simulations in the summer (June 1 – August 15) makes a much larger reduction than the scaling factor for simulations in the spring months (March 1 – April 30). During colder months in the upper Midwest, CO emissions could be higher because of less efficient combustion from mobile sources, plus sources from home heating using wood fuel. In contrast to our model results, the NEI-99 winter inventory predicts that total national CO emissions will be slightly lower during winter months than during the summer. On-road sources are predicted to be 5% higher during winter months and area sources such as home fuel burning are about three times higher during the winter. But non-road sources such



**ACPD**

8, S7709–S7717, 2008



as tractors and construction equipment are estimated to be 97% higher during summer months (Frost and McKeen 2007). Our results suggest that anthropogenic CO emissions are higher in spring months than summer months. We therefore cast doubt on the seasonal adjustments used in NEI-1999."

Page 11416, Line 12: We did intend to refer to Figure 12, the a posteriori time series for model results at WLEF. Hopefully a more lucid explanation in this section makes the figure references less confusing and difficult to follow.

Section 3.4: The disagreement between NCAR and URI INTEX-A measurements does appear somewhat befuddling. GEOS-Chem simulations from Millet et al (2006) capture 70% of the variability in NCAR formaldehyde measurements but capture only 42% of the variability in URI measurements. We therefore use NCAR measurements for the model optimization. We have clarified this in the text (pg. 11419, line 1): "GEOS-Chem simulations from Millet et al (2006) capture 70% of the variability in NCAR formaldehyde measurements but capture only 42% of the variability in URI measurements (along with a 34% bias). We therefore use NCAR measurements for the model optimization."

Page 11421, Line 17: Our relative estimated VOC contribution is lower than that estimated by Hudman et al. (2008). Hudman et al. (2008), however, compute the relative contribution of VOCs to CO over the entire continental US. Their calculations include the American Southeast, a region of the country that can have up to four times the mid-day summer biogenic VOC emissions of Wisconsin. Because of spatial heterogeneity in biogenic VOC emissions, our computed contribution of VOCs to CO will be lower than in Hudman et al. (2008). We change the passage as follows: "Hudman et al. (2008) found that biogenic VOCs contributed 56% of the CO source over the continental US during the summer. We found a lower relative VOC contribution likely because our study sites did not experience significant influence of VOCs from high biogenic source regions such as the American Southeast."

Page 11422: The Bayesian analysis predicts a very small uncertainty for the a poste-



**ACPD**

8, S7709–S7717, 2008



riori constraints on anthropogenic CO emissions. Because a posteriori anthropogenic CO emissions are so highly constrained, uncertainties in a posteriori VOC emissions appear unlikely to significantly effect knowledge of anthropogenic CO, certainly not enough to complicate the estimated seasonal trends in anthropogenic CO.

References

Arellano, A.F., and Hess, P.G.: Sensitivity of top-down estimates of CO sources to GCTM transport, Geophys. Res. Lett., 33, doi:10.1029/2006GL027371, 2006.

Conrad, R. and Seiler, W.: Utilization of traces of carbon monoxide by aerobic oligotrophic microorganisms in ocean, lake and soil, Arch. Microbio., 132(1), 41-46, 1982.

Duncan, B.N., Logan, J.A., Bey, I., et al: The global budget of CO, 1988-1997: Source estimates and validation with a global model, J. Geophys. Res., 112, D22301, doi:10.1029/2007JD008459, 2007.

Frost, G., and McKeen, S.A.: Emissions inventory mapviewer,US Department of Commerce, National Oceanic and Atmospheric Administration, National Satellite, Data, and Information Service, National Geophysical Data Center, Boulder, C.O., http://map.ngdc.noaa.gov/website/al/emissions/viewer.htm, 2007.

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.

Intergovernmental Panel on Climate Change (IPCC): Climate change 2001: The science of climate change, Third assessment report of the Intergovernmental Panel on Climate Change, Hougton, J.T., Ding, Y., Griggs, D.J., et al, ed., Cambridge University Press, New York, 2001.

Intergovernmental Panel on Climate Change (IPCC): Climate change 2007: The Physical Science Basis, Fourth assessment report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., et al., Cambridge Univer-



**ACPD**

8, S7709–S7717, 2008



sity Press, NewYork, 2007.

Liu, L., et al.: A photochemical modeling study of ozone and formaldehyde generation and budget in the Po basin, J. Geophys. Res., 112, D22303, doi:10.1029/2006JD008172, 2007.

Millet, D.B., Jacob, D.J., Turquety, S., et al: Formaldehyde distribution over North America: Implications for satellite retrievals of formaldehyde columns and isoprene emissions, J. Geophys. Res., 111, D24S02, doi:10.1029/2005JD006853, 2006.

Müller, J.-F., and Stavrakou, T.: Inversion of CO and NOx emissions using the adjoint of the IMAGES model, Atmos. Chem. Phys., 5, 1157-1186, 2005.

Oliver, J. G. J. and Berdowski, J. J. M.: Global emissions sources and sinks, in: The Climate System, edited by: Berdowski, J., Guicherit, R., and Heijj, B. J., Balkemea/Swets and Zeitlinger, Lisse, The Netherlands, 33–78, 2001.

Petron, G., Granier, C., Khattatov, B., et al: Monthly CO surface sources inventory based on the 2000-2001 MOPITT satellite data, Geophys. Res. Lett., 31, L21107, doi:10.1029/2004GL020560, 2004.

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.

Turnbull, J.C., Miller, J.B., Lehman, S.J., et al: Comparison of 14CO2, CO, and SF6 as tracers for recently added fossil fuel CO2 in the atmosphere and implications for biological CO2 exchange, Geophys. Res. Lett., 33, L01817, doi:10.1029/2005GL024213, 2006.

US EPA: Emissions modeling clearinghouse related spatial allocation files: "New" surrogates, http://www.epa.gov/ttn/chief/emch/spatial/newsurrogate.html, 2004.

US Environmental Protection Agency: 2005 National emissions inventory data & doc-



**ACPD**

8, S7709–S7717, 2008

umentation, http://www.epa.gov/ttn/chief/net/2005inventory.html, 2007a.

US Environmental Protection Agency: 1970-2006 Average annual emissions, all criteria pollutants, http://www.epa.gov/ttn/chief/trends/index.html, 2007b.

US Environmental Protection Agency: National emissions inventories for the US, http://www.epa.gov/ttn/chief/net/index.html, 2008.

Wiedinmyer, C., Quayle, B., Geron, C., et al: Estimating emissions from fires in North America for air quality modeling, Atmos. Env., 40, 3419-3432, 2006.

---

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