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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 #1

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General Comments

The paper describes the use of the lagrangian model STILT in a special version adopted for CO and its atmospheric chemistry, in order to derive updated emission maps for CO and formaldehyde in a Bayesian framework deploying CO observational data from two tall tower sites and aircraft campaigns. The approach followed here is quite novel and a good extension of the STILT inversion framework, that was developed for CO2. The study makes a valuable contribution to the progress in the use of the atmospheric signals contained in diurnal timescale variability to improve our knowledge in sources and sinks of atmospheric constituents. The paper is well written and

concise, but misses out on quite some details. Next to the comments of the other two reviewers I would like to note the following major and minor points. After the following major observations have been addressed I would recommend the paper for publishing.

Specific comments:

Model performance vs. observations (before and after flux optimisation) is not discussed systematically, and a long list of numbers in the text next to an alphabet soup of acronyms makes hard reading. Despite this only some metrics (r, rmse) are given at some occasions. A good place to insert a table would be P11414 line 17 to show model performance for both sites and the different seasons before and after inversion.

P11396, although the title and abstract indicate that the focus of the paper is North America, this should be made clear in the Introduction as well. The "original six criteria air pollutants"; and "non-attainment status"; is only clear for those familiar with the USA (Clean Air Act?) context.

P11397, line 10; P11398, line 4: update statement to 4th IPCC AR

P11400, line 15: The STILT lateral boundary condition seems applicable for a component like CH4 with a high background concentration and low source strengths over the ocean. For a component with much smaller atmospheric life-time like CO and HCHO this approach is less satisfying, better would be to include data from a global CTM like GEOS-CHEM.

P11402, line 23: Eq. 4 is not very clear. Better would be to give the 3 terms as separate equations followed by the overall summation of the terms

P11404: The authors seem to disregard the antropogenic emission of NMVOC's other than HCHO completely, as well as natural CH4 emissions

P11413, line 11 and further: Sensitivity of the uncertainty in the modeled concentration to the number of particles released is only tested for the aircraft measurements and only for 100 resp. 500 particles. The results seem to indicate that for high concen-

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trations and lower observations (tall towers!) the number of particles does matter.

P11414, line 3: in Fig 8 a cut-off of the footprint at 1% of the maximum (corresponding to ln(f) about -10) seems more appropriate then the current ln(f)=-20 corresponding to 3e-5%

P11414, line 14: It would still be interesting to see how the inversion system would handle the EDGAR emission inventory data. From the graphs we cannot conclude whether the correlation of measurements and model is better when using the EPA or the EDGAR prior inventory. A table with statistics on correlation coefficient, rmse and explained variance would be welcome here.

P11417, line 11: European antropogenic CO emissions are also mainly from mobile sources (diesel engines), where large seasonal effects are not expected. Heating and power generation sources on average have very low CO/CO2 ratios in Europe, but still can explain the higher CO emissions in winter.

P11417 Section 3.3 needs a rewrite. It is not clear from the current text why certain events work out well and other don’t as we cannot see from the text or pictures which event was affected by pyro-convection or not. Sites closer to the fires would not help in resolving emissions from pyro-convective events as long as this phenomenon is not included in the model parametrisations.

P11419, section 3.5.1. It seems more logical that in the BRAMS meteorological driver the boundary layer height is less well represented for coastal areas as the transport field itself. Also the STILT lateral boundary condition at 145 W will work less well for east coast sites.

P11421, line 8. The model result that methane decomposition is the main source of CO in summer is somewhat surprising and not entirely consistent with recent literature as is stated in line 12 unless one considers CH4 to be a biogenic VOC or one considers a factor of two as margin. The question arises how these numbers depend on the

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uncertainty given to the prior emissions of the different sources and uncertainties in the chemistry.

P11422, line 7. Reasonable results for one tower do not allow for this strong conclusion; the part "providing strong tests"; could well be but are not supported by this paper.

Technical corrections:

P11395, line 8: high fidelity->satisfying line 9: from tall towers and point measurements->for one tall tower and

P11397, line 18: Oliver->Olivier (and reference at P11428) (no, that's not me ;-))

p11404, Line 23: EDGAR 32FT2000 should be cited as: Olivier, J.G.J., Van Aardenne, J.A., Dentener, F., Ganzeveld, L. and J.A.H.W. Peters (2005). Recent trends in global greenhouse gas emissions: regional trends and spatial distribution of key sources. In: "Non-CO2 Greenhouse Gases (NCGG-4)";, A. van Amstel (coord.), page 325-330. Millpress, Rotterdam, ISBN 90 5966 043 9.

p11405, Line 15: EDGAR V3.2 should be cited as: Olivier, J.G.J. and J.J.M. Berdowski (2001). Global emissions sources and sinks. In: Berdowski, J., Guicherit, R. and B.J. Heij (eds.) The Climate System, pp. 33-78. A.A. Balkema Publishers/Swets & Zeitlinger Publishers, Lisse, The Netherlands. ISBN 905809 255 0.

P11409, line 11: (Palmer et al., 2006)-> Palmer et al. (2006)

P11410, line 2+4 : estimates->estimate

P11421, line 22: compounts->compounds

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