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Atmospheric Chemistry and Physics Discussions

Interactive comment on "North American acetone sources determined from tall tower measurements and inverse modelling" *by* L. Hu et al.

Anonymous Referee #1

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This paper describes the constraint and inference of acetone sources over North America based on a GEOS-chem inverse model simulation and 1 year of high time-resolution acetone observations from a tall tower observatory from Minnesota, USA. The study derives new information concerning the seasonality of contributions from different natural and anthropogenic acetone sources, and investigates the atmospheric chemistry impacts of acetone from the optimised simulation. The results will be of strong interest to the atmospheric chemistry community, particularly with regard to the importance of biogenic acetone sources in light of future projected changes to anthropogenic VOC emissions and the possible effects of climate change on terrestrial biogenic emissions. The paper is very well written, and both the observational and modelling methodologies are well described and thorough. I recommend the paper for publication in ACP, with a number of minor corrections / clarifications, detailed below.

Page 30878, Line 25: Doesn't methanol have a very large photochemical source? i.e. methane oxidation?

Page 30881, Lines 5-6: Why should the fractional error in BL height be directly equal to the forward model error in the inversion? Is there a justification for assuming that this is the dominant forward model error?

Section 3.3 - discussion of reasons for opposing signs of a priori errors in the (broadleaf + shrub + herbaceous) and (needleleaf + secondary biogenic) sources. Is another possible explanation the modelled acetone yields from the biogenic precursors? i.e. either as a result of model chemistry errors or OH bias.

Section 3.6 - Impact on Atmospheric Chemistry. I find this the weakest section in the paper in terms of its purpose, and its design. While I understand that it is useful to quantify how the optimised source estimates and even how future changes in anthropogenic sources, change the model prediction of atmospheric chemistry, the projections made for "future climatic conditions" require some further explanation or caveats. It is not clear what purpose these projections serve as they are not constrained by temperatures and meteorology consistent with projected climate change. While I realise that such temperature changes have been accounted for when discussing possible future changes in biogenic sources, I feel that temperature change impacts on atmospheric chemistry cannot simply be ignored, especially since PAN is a major focus of the discussion. Is it possible to account for how the temperature changes would affect the PAN and NOy partitioning discussed? At least some statement recognising this omission from the calculations and an estimate of the likely impact on e.g. PAN lifetime should be added.

Figure 1: I suggest making the outline of the filled circle in each panel showing the tower measurements thicker/bolder, as it is difficult to make it out in the present format.

Figure 6: Is it surprising that there is little seasonality in the long-range transport absolute acetone source? Making a simple assumption that transport to the site displays

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little seasonality, this means that there is little seasonality in the atmospheric acetone abundance upwind of the site. This might be surprising given the expected seasonality in both biogenic and photochemical sources, which might be expected to affect the 'background' acetone abundance. The secondary anthropogenic source also shows little seasonality. Does this indicate a compensation between increased photochemical production and increased photochemical loss during spring/summer compared with winter? Can the authors shed any more light on this from their simulations?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 30869, 2012.