

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

Anonymous Referee #2

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I think that this is a significant body of work, adding to the acetone measurements required to obtain more information on the global sources, particularly biogenic of acetone. It adds to the body of information tabulated in Jacob et al 2002. However, I find the paper confusing. The main thing is the organization of the paper. I have some questions/concerns. They are as follows:

How high is the top of the model? How is the boundary at the top of the model treated? Is the figure 1 the full domain? On the map where is the sub domain?

It looks like there are results from the model in the methodology section (30877 top). It may make more sense to structure the paper closer to Jacob et al 2002. I think that it would make more sense to put the “Inverse analysis (3.2)” and GEOS-Chem ...called section 3. Model Description section. Followed by a section on “Observations

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and Comparison to Inverse Model results" or something like that. The details of the sensitivity tests need to be lumped with the model. It jumps back and forth to the model and observation and becomes confusing.

Pg 30877 "...NA biogenic acetone source is 4.8 Tg or approx 13% of the global terrestrial biogenic acetone source." I am assuming this is primary and secondary production. This scales up to a global terrestrial source of 36.9 Tg. Who's estimate of 36.9 Tg did you use? (Jabob et al 2002 had a terrestrial estimate of ~43 Tg)

Jacob et al 2002 found that the only way that their inverse modeling could account for the differences low acetone in Georgia and the high values in Alabama and Tennessee was for there to be large local vegetation sources near the site. You have stated that in the summer biogenic primary production dominates. With the biogenics dominating won't the concentrations at the site be largely influenced by local emissions? How does the local vegetation affect your assessment that broadleaf trees, shrubs and herbaceous plants are underestimated and that your needle leaf trees are over estimated?

Did you test for losses of Acetone when drying to -25C? There can be a quasi-liquid layer at -25C which can result in the loss of acetone.

Line 25 on Pg 30879. The priori primary and secondary sources are not described in section 2.2 (GC-MS/FID measurements). They are described throughout 3.2.

Line 1 on page 30880. What are the sources being optimized? State explicitly at the start what the primary and secondary sources are. Later on you talk about 8 categories but also talk about the ocean. It is confusing trying to follow what you were doing. As you perturb the model source do you rerun the GEOS-Chem model?

I can see why you would lump the broadleaf plants and shrubs together. It makes sense. I can even see why you would lump together secondary biogenic production and primary emissions from needle leaf trees together since the majority of the sec-

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ondary biogenic production is through the oxidation of monoterpenes which have a short lifetime. However, you combine primary and secondary anthropogenic sources, which does not make sense to me. The anthropogenic secondary production of acetone can be very different from its primary emission source since this is dominated by propane and isobutene. Can these terms not be separated?

In some places you refer to the acetone outside the domain as long range transport and in others as your boundary acetone conditions. You need to pick one way to address it. I like regional background.

Line 18 pg 30883 - You state that you infer a 40% underestimate of the acetone boundary conditions. This should depend on the boundary conditions which are not clearly stated.

In the source apportionment section you now are back to the total 8 or 9 source categories rather than the 4 above. It seems that you are jumping between a Priori and A Posteriori results without explaining which are which

Maybe you need a table of your results since you are stating a lot of numbers for various factors in various seasons. It can be confusing.

It should be impact on PAN not on atmospheric chemistry as the heading for the last section. I don't feel that the last section on PAN adds all that much to the paper and it leaves the paper on a bit of a low note.

Figure 7 Not really all that clear since almost the whole of NA is the same color >10.5 for acetone. Maybe you need a different scale (color coding) for the Acetone and the PAN. There is no detail in the maps.

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