We thank referee # 2 for their comments. Responses and changes made to the manuscript are outlined below. Referee's comments are in **bold** *italics*.

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?

The nested version of GEOS-Chem has the same vertical extent as the full global model (up to 0.01 hPa, or ~ 80km), so no vertical boundary condition is needed. The lateral boundary conditions of all species for each vertical layer are taken from year-long global simulations carried out at 4° × 5° resolution, at a frequency of every 3 hours. We have clarified these points in the revision. Figure 1 shows the full domain of the nested simulations performed in this study. The sub-domain mentioned in the caption is slightly smaller than the full domain, and is only used for computing North American source magnitudes.

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 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.

Thanks for the suggestion. We prefer to keep the structure of the paper as-is.

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)

Yes, the 4.8 Tg includes primary (76%) and secondary production (24%) of acetone, as stated in the manuscript. The global terrestrial source of 36.9 Tg is from the 4x5 GEOS-Chem simulation which provided the boundary condition for the nested model (0.5x0.6667) used in this paper. The simulation reflects the updated global budget estimate of Fischer, Jacob, et al. (GRL, 2012). We clarified this point in the revision.

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?

The observation presented here are from a 244 m tall tower. The elevated sampling height, combined with the 32 day acetone lifetime, helps to integrate over this type of fine-scale heterogeneity in emissions. Our source optimization does implicitly assume that the prior spatial distribution of emissions within each source category (derived based on plant functional type distributions) is accurate. The strong correlation

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between our simulated and observed acetone concentrations (R = 0.83) indicates that this assumption is reasonable.

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.

We have used the same approach previously (Millet et al., 2005) and observed no detectable 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.

Thanks for catching this. We corrected this point in the revised version.

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?

This information is provided on p. 30880: "These sensitivities were derived with respect to eight distinct acetone sources within the North American domain of Fig. 1: biogenic emissions from broadleaf trees, needleleaf trees, shrubs, and herbaceous plants; secondary production from biogenic precursors; primary anthropogenic emissions; secondary production from anthropogenic precursors; and biomass burning. We also derive the sensitivity with respect to sources outside North America, manifested as the boundary conditions for the nested model domain." On the same page (Line 7) we state that gross ocean emissions are not included in the state vector. As we discuss in Section 3.2, only 4 of those source categories can be resolved by our observations, and this set is what we optimize. These 4 source categories being optimized are listed at the end of Section 3.2. We now list them again at the start of Section 3.3 for clarity.

Yes, we rerun GEOS-Chem model for each perturbation, and we have clarified this point in the revised version.

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 secondary 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?

No, the anthropogenic secondary production of acetone and its primary emission cannot be separated in our analysis based on the fact that these two terms are highly correlated. This point is made in the paper on page 30883. This multicollinearity would lead to non-robust scale factors if the terms were 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.

Thanks for the suggestion. We have made this change in the revised version.

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.

We added a description of the boundary condition to the Section 2.3.

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 expaining which are which .

We have clarified this point in the revision.

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.

Thanks for the suggestion. We believe Figure 6 is sufficient for this purpose and prefer not to add an extra table.

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.

See response to related comment by reviewer #1.

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.

Thanks for the suggestion. We changed the scale bar for acetone.

References

Millet, D.B., Donahue, N.M., Pandis, S.N., Polidori, A., Stanier, C.O., Turpin, B.J., and Goldstein, A.H.: Atmospheric volatile organic compound measurements during the Pittsburgh Air Quality Study: Results, interpretation and quantification of primary and secondary contributions, J. Geophys. Res., 110, D07S07, doi:10.1029/2004JD004601, 2005.