

We thank referee # 1 for their comments. Responses and changes made to the manuscript are outlined below. Referee's comments are in **bold italics**.

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

Yes, methanol has a photochemical source mainly from oxidation of methane. The magnitude of this secondary source is estimated at 18 – 38 Tg/y globally, or approximately 15% of the total global methanol source (Jacob et al., 2005; Millet et al., 2008; Stavrakou et al 2011). The photochemical production of methanol is a diffuse source and highest in the tropics (Jacob et al., 2005) and is not likely to appreciably impact the variability observed at our tall tower. However, in the revision, we changed the wording on this point to avoid any confusion.

***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?***

We have found in the past (as have others) that errors in the model BL height can be a key source of error in simulating tracer concentrations near the surface. As a starting point for the inversion, we therefore made the assumption that BL height uncertainty is the dominant source of forward model error, and can result in a corresponding error in the simulated surface concentration. However, the reviewer is correct that there can certainly be other errors in the forward model, which is why we carried out two sensitivity tests to assess how our findings depend on the model error estimate used in the inversion (doubling and halving it). As we see from Figure 4, these sensitivity runs did not significantly change the result, showing that the inversion is not very sensitive to this assumption.

***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.***

Yes, this is possible and we mention it in the paper (p. 30883): “However, because of the way source categories are combined in the state vector, we cannot rule out the possibility of an underestimate of direct acetone emission across all PFTs, combined with an overestimate of secondary biogenic production.” The reviewer is correct that this type of overestimate for secondary production could arise from model errors in the acetone yields or in OH.

***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 optimized 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 realize 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 NO<sub>y</sub> 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.*

Thank you for the thoughtful comment. In the revised version, we have improved the language in this section to better communicate what is being done. We are not examining future scenarios or projections, but testing the sensitivity of atmospheric acetone and PAN with respect to biogenic and anthropogenic acetone emissions within North America. The reviewer is correct that climate changes would also affect other parameters such as the PAN lifetime. We now make this point in the text and emphasize that we are examining partial derivatives of atmospheric acetone and PAN with acetone source changes in North America.

*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.*

Thanks for the suggestion. We have made the change in the revision.

*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 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?*

As the reviewer points out, the secondary anthropogenic contribution to acetone at the tower displays little seasonality. This likely reflects a compensation between an increased photochemical production rate in the summer and an increased acetone lifetime in the winter. A similar compensation effect is probably the reason for the weak seasonality in the background term. We have added a mention of this point to the manuscript.

## References

- Jacob, D. J., Field, B. D., Li, Q. B., Blake, D. R., de Gouw, J., Warneke, C., Hansel, A., Wisthaler, A., Singh, H. B., and Guenther, A.: Global budget of methanol: Constraints from atmospheric observations, *J. Geophys. Res.*, 110, D08303, doi:10.1029/2004JD005172, 2005
- Millet, D. B., Jacob, D. J., Custer, T. G., de Gouw, J. A., Goldstein, A. H., Karl, T., Singh, H. B., Sive, B. C., Talbot, R.W., Warneke, C., and Williams, J.: New constraints on terrestrial and oceanic sources of atmospheric methanol, *Atmos. Chem. Phys.*, 8, 6887–6905, doi:10.5194/acp-8-6887-2008, 2008.
- Stavrakou, T., Guenther, A., Razavi, A., Clarisse, L., Clerbaux, C., Coheur, P.-F., Hurtmans, D., Karagulian, F., Mazière, M.D., Vigouroux, C., Amelynck, C., Schoon, N., Laffineur, Q., Heinesch, B.,

Aubinet, M., and Müller, J.-F.: First space-based derivation of the global atmospheric methanol emission fluxes, *Atmos. Chem. Phys.*, 11, 4873-4898, doi: 10.5194/acp-11-4873-2011, 2011.