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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observational constraints on the global atmospheric budget of ethanol" *by* V. Naik et al.

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We thank the reviewer for reviewing our manuscript and providing us constructive comments. Their comments with our responses are given below.

Response to Anonymous Referee #2

The paper describes a modeling experiment with a global chemical transport model which goal is to better constrain the global budget of ethanol by comparing model results with currently available observations. The paper is well written and provides useful and original information regarding the possible sources and sinks of ethanol. I only have minor comments.

1. Section "Model and experiments": Why do the Author need to update the POET

emissions inventory? Is there no geographical distribution provided? How do they estimate the fraction of ethanol which is emitted from ethanol production (10%)? How are their results sensitive to this fraction? What are the uncertainties associated with these anthropogenic emissions?

Thank you for drawing our attention to ethanol emissions used in this study. Before we proceed to answer your question we would like to address a mistake in the text. We did not scale the POET emissions for each region with respect to the regional production assuming that 10% of the ethanol produced is emitted into the atmosphere. Instead, we redistributed the POET emissions on the basis of the regional production statistics keeping the same global total as in the POET emissions. Coincidentally the redistributed emissions in each region are approximately 10% of the regional ethanol production. We have corrected the explanation of emissions in the manuscript. The POET database includes gridded emissions from industrial and biofuel sources for ethanol. Our initial assessment of the geographical distribution of these ethanol emissions indicated that it did not match the regional ethanol production statistics provided in the Renewable fuels Association Ethanol Industry Outlook (2006). For lack of detailed information on how ethanol emissions were compiled by Olivier et al. (2003), we updated the spatial distribution as described above. Detailed analysis of the uncertainties associated with these anthropogenic emissions is beyond the scope of this study.

2. Section "Results and discussion": I find the statement "The model underestimate of aircraft measurements over NA is not necessarily inconsistent with the overestimate of ship observations along the tracks. . . " a bit weak. The Authors base (to some extent) this statement on the relative contributions of different tagged tracers while the overall agreement between model and observations is rather low. This makes it difficult to actually trust these tagged contributions. Could they rephrase and possibly extend this discussion?

We have rephrased the discussion in the paper as follows: One could interpret the

model underestimate of aircraft measurements over North America and an overestimate of ship observations along the northeastern U.S. coast (described above) as a disparity between the two sets of observations. However, it is possible to reconcile these comparisons given the different regional coverage of measurements particularly if different sources contribute to the observed ethanol concentrations. The aircraft flight tracks cover a much larger area (60° to 130° West and 24° to 52° North) than the ship tracks (67° to 75° West and 41° to 44° North). The tagged tracers in Fig. 5 indicate that industrial and biogenic sources contribute equally to the ethanol concentrations sampled by the aircraft in the lower troposphere (below 2 km) over North America, while industrial sources are the primary source of ethanol measured off the northeastern U.S. coast in agreement with the findings of de Gouw et al. (2005).

3. Section "Results and discussion", Figure 4: The additional diffuse source used in the SYNEOH simulation enhances ethanol mixing ratios by about 0.1 ppbv throughout the column over the TRACE-P region for example. In contrast, mixing ratios over continental regions seem to be only marginally affected by this additional source (at least not to the same extent). Could the Authors explain this a bit better?

Seasonal and regional variability in loss processes (OH-loss, dry deposition and wet scavenging) drive the enhancement in ethanol concentrations from the additional diffuse source in the SYNEOH simulation over each aircraft campaign region shown in Figure 4 (now Figure 6). Losses of ethanol are smaller over the TRACE-P region in February-April as compared to other regions for different months resulting in the build-up of ethanol, therefore, the additional diffuse source results in enhanced ethanol concentration over the TRACE-P regions as compared to other regions. This is demonstrated in the monthly distribution of SYNEOH concentrations in the model at 500 mb in Figure A.

4. Section “Results and discussion”: Could the Authors elaborate a bit on the potential role of aqueous chemistry and if possible, provide an estimate of this potential source?

For lack of evidence in the scientific literature on the impact of aqueous-phase chemistry on the atmospheric abundance of ethanol, we have removed this sentence from the manuscript.

References:

de Gouw, J. A., Middlebrook, A. M., Warneke, C., Goldan, P. D., Kuster, W. C., Roberts, J. M., Fehsenfeld, F. C., Worsnop, D. R., Canagaratna, M. R., Pszenny, A. A. P., Keene, W. C., Marchewka, M., Bertman, S. B., and Bates, T. S.: Budget of organic carbon in a polluted atmosphere: Results from the New England Air Quality Study in 2002, J. Geophys. Res., 110, D16305, doi:10.1029/2004JD005623, 2005.

Olivier J., Peters, J., Granier, C., Petron, G., Müller, J. F., and Wallens, S.: Present and future surface emissions of atmospheric compounds. POET report #2, EU project EVK2-1999-00011, 2003.

Renewable fuels Association Ethanol Industry Outlook, 2006, http://www.ethanolrfa.org/objects/pdf/outlook/outlook_2006.pdf.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 925, 2010.

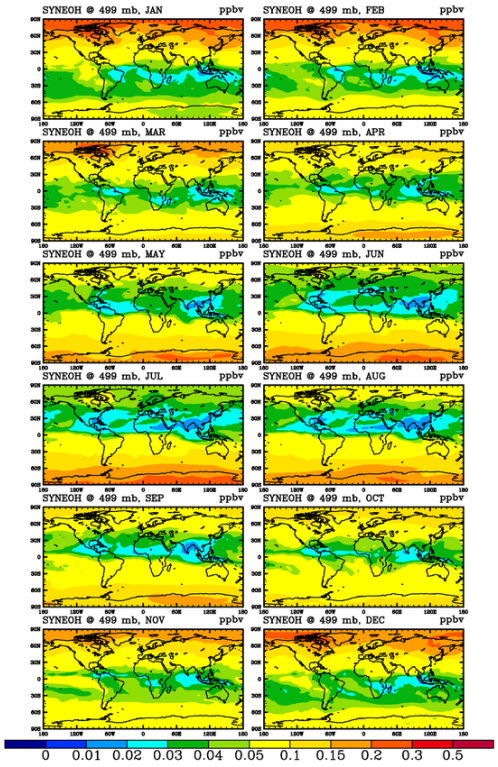

Fig. 1. Figure A. Monthly mean concentration of tracer SYNEOH at 500 mb