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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Are biogenic emissions a significant source of summertime atmospheric toluene in rural Northeastern United States?" *by* M. L. White et al.

Anonymous Referee #1

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White et al., open an interesting discussion in assessing the significance of biogenic toluene emissions. The sources of toluene are studied with field measurement using several methods as well as comparing with anthropogenic emission factors. Interpretation and scaling of local measurements to atmospheric scale always leaves open questions of how representative they are and what is the effect of annual variation. White et al. use their limited measurement sets, done with different methods and measured at different sites and different years, in a good and transparent way. The different measurements are bravely combined for determining an important, as well as challenging, question of possible biogenic toluene sources in continental scale. However, parts of the manuscript are misleading and a few major weaknesses weaken or hinder

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the conclusion of importance of biogenic emissions on local toluene concentrations. Even the main conclusion that that part of summer (2006) increases are from biogenic emissions is weakened, since seasonality in fuel evaporation (what authors suggest as main sources for summer 2004 and 2005 increases) and seasonality in air mass sources are not fully addressed. The title is misleading, the authors state that seasonality in fuel grade and its evaporation is responsible for increases in summers 2004 and 2005. The authors suggest that only when crops was changed from corn to alfa alfa, in summer 2006, biogenic emissions made a significant contribution. As the authors state, missing of corn emission measurement data is problematic. Information on the surroundings, locations of anthropogenic sources and biogenic sources is missing (e.g. map). In addition, seasonal enhancement of transport from year-round emissions from local industry can not be excluded based on local wind direction analysis is not same as the direction of coming air mass. Below are detailed points to be addressed. The study presents important insight in possible biogenic toluene sources that are not taken into account in traditional inventories and is in the scope of ACP audience.

1. Change the title to better reflect the outcome

2. In introduction, page 12286, line 4, authors state that gasoline is summer grade 1 June to 15 September. Please clearly state this time also in the results and discussion when summer influence of fuel evaporation is discussed. Evaluate is this in fact the same as the period when toluene is higher, taking into account a time lag for the change fuel grade in vehicles.

3. Page 12287, line 11, "In particular, we identify and quantify the contribution of seasonal changes in gasoline formulation to evaporative toluene emissions." Please add this in the results, at the moment it is not clear that it is done. On the contrary is seems to be forgotten, summer evaporations are summed in table 3 on basis of spring time results. In Section 3, 3rd paragraph it is not clear for the reader if this effect of change in grade is taken into account, the authors point that the summer time toluene/benzene and C2H2/CO ratios are different than any other anthropogenic VOC

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relationships, but do not present results for the reader, refer to the unpublished results by Russo et al. Please clarify trough out the text how the change in fuel grade is taken into account.

4. Page 12288, line 3. Please check that there is no seasonality in the sampling times and include the result in the description. Concentrations have diurnal patterns due to sources, sinks and mixing. Even a change of few hours in sampling time can result in systematically different concentrations.

5. Section 3, first paragraphs. The effect of seasonality of boundary layer height and mixing is not taken into account, please add it and according references in the discussion. Clarify: could winter time heating and local burning increase during winter increasing anthropogenic winter emissions? Include an analysis on the seasonality of source of air mass done with trajectory analysis. Local wind direction (Page 12295, line 13) can be strongly effected by local geography and is not a sufficient indicator of air mass source even on 20 km scale.

6. Section 4. Please include a map of the surroundings with descriptions of different sources and their seasonal contribution to the measurement site from trajectory analysis. In Page 12295, starting at line 7 the need for a map is further increased. A 20 km radius is small for such a long lived compound as toluene. Would the results be different if a larger radius was considered?

7. Page 12293, lines 13-15, authors state that in 2004 and 2005 fuel evaporation was the major factor on the summer increase of toluene. They also state that change from corn to alfa alfa in summer 2006 increased the biogenic emissions and imply that local biogenic emissions were increased by the change of crops. a) this is in contradictory with table 3, were crop emissions are same for each summer. Missing of corn emission data is one of the major weaknesses of this paper. b) these results are in contradictory with fuel evaporation results summed in table 3, where the highest influence of fuel emissions is in 2006 Please clarify the outcome of the study and be consistent in

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interpretation of results.

8. Page 12295, lines 11-13, seasonality of air mass source can not be studied by local wind direction. Further more, without a trajectory analysis of the seasonality of air mass source the conclusion on Page 12295, line 22-26, can not be made. Reconsider and rephrase the sentence on lines 22-24 and remove that "All this evidence together rules out local industry as the major source responsible for the summertime toluene enhancements." Please include trajectory analysis and reconsider effect of industry based on the 20 km and a longer radius on the basis of it.

9. In 12298, second paragraph, the authors point out a weakness, the biogenic emissions are based on autumn fluxes of alfa alfa, but forget the change from corn. Please add that the crop in 2004 and 2005 was corn and that there is no estimate of its emissions (even that it would emit toluene) or present results of corn emission.

10. A major weakness is that description of the coverage of different sources, crops, forest and local industry are not presented. Please include them e.g. in a map mentioned in earlier comments or in a separate figure.

11. Table 3. In the text the authors suggest that crop emissions were increased in 2006 when crop was changed, however here they are same. Please clarify.

12. Figure 5. How does the chamber temperature and light change during the measurement? Is there a possibility that toluene is emitted from the soil?

13. Figure 6 b. Scale for a-pinene emission is confusing. This similar behaviour alone is hardly sufficient evidence of temperature dependent emission (that authors suggest). Please consider plotting a-pinene on another panel in the figure.

Technical comments

1. Page 12285, line 3, add brackets: "ozone O3 formation" to "ozone (O3) formation"

2. Footnote reference 1 Russo et al., is not yet in ACPD, correct the reference

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3. Page 12287, lines 18-19, correct "04:00-UTC" and "05:00-UTC" to "UTC-04:00" and "UTC-05:00"

4. Page 12288, line 8, "NMHC" to "non methane hydro carbons (NMHC)"

5. Page 12289, line 5, correct spelling "...no VOC emission artifacts are associated..." to "...no VOC emission artifacts associated..."

6. Page 12289, line 6, correct spelling "...sampled was placed..." to "sample was placed..."

7. Page 12290, line 2, remove "sweet-gum (Liquidambar styraciflua)", the data is not presented in the current paper.

8. Page 12292, line 12, change "can" into "canister"

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 12283, 2008.