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Interactive comment on “Contrasting organic aerosol particles from boreal and tropical forests during HUMPPA-COPEC-2010 and AMAZE-08 using coherent vibrational spectroscopy” by C. J. Ebben et al.

Anonymous Referee #1

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This paper describes novel measurements of optical properties of secondary organic aerosol (SOA) from laboratory-generated aerosol and two regions in which biogenic VOC reactivity dominates and infers chemical information mainly from comparison of the laboratory studies with the field samples.

The method used in this work represents a very interesting way to analyze ambient aerosol without extraction etc. or use of vacuum and should be of interest to the community. The paper is well written and I think this method and the results represent an exciting new analytical tool for aerosol. I believe this method has great potential and

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Discussion Paper



Interactive
Comment

I look forward to seeing more results employing it! However, before the manuscript is suitable to ACP, the authors should address the following issues in a revised submission for publication in ACP.

My main comment is that the authors state that the method used is a way to characterize the chemical composition, which is vague and could be interpreted very broadly that the overall (i.e. all aspects) chemical composition can be deduced from this method. I believe it has to be very carefully phrased what is meant by this claim and certainly their method needs to be compared/contrasted with existing methods for characterization of chemical composition such as AMS measurements of O/C ratios, FTIR analysis or filter extraction methods. As the results are from two major field campaign such results must exist, particularly AMS data.

I) The method description is excellent. However, a statement that simply and clearly states whether this method is a surface or bulk aerosol measurement, or under which conditions, which type of measurement dominates is necessary. I believe this aspect otherwise is hard to understand for non-experts and I highly recommend such a statement early on in the manuscript otherwise readers might get confused. For the implications and for comparison with other data this aspect is also very important. If the measurements are pure surface measurements the question of coatings or contaminations becomes important and should be addressed (see also a later comment).

II) The isoprene/a-pinene ratio is a large and compelling difference between AMAZE and HUMPPA. However an equally important difference is the O₃/OH ratio and potentially the NO concentrations. From the manuscript as it stands it is not clear to me how the authors can clearly argue that the differences observed cannot be from the change in chemical regime from O₃ to OH and/or medium to low NO and/or relative humidity. I don't disagree with the authors' conclusions but it would be very helpful if their argument could be strengthened, as it is a very important conclusion of the paper. Specifically, by stating (p. 16397 lines 25-end) that AMAZE is an important contrast because of low anthropogenic impact they clearly imply that HUMPPA had high

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anthropogenic impact and hence a difference in NO regime is important.

III) P. 16498 26-27 "... dominated by a-pinene derived SOA particles": I think it can only be said that it is consistent with the lab samples. I recommend making the existing statement weaker or finding additional proof. Have the authors studied laboratory generated b-pinene or limonene SOA, especially as they make such a statement. In particular in figure 1 a-pinene and b-pinene look very similar to each other hence the SOA could look similar unless the authors have information not available to the reader or the reasoning has to be stated more clearly. The authors should add results (if possible) on lab-synthesized b-pinene and limonene SOA to the manuscript - it could (potentially) greatly strengthen the implications.

IV) Table 1) The paper centrally hinges on comparison between lab and ambient samples. Conditions can substantially influence oxidative chemistry and potentially the resulting SOA composition/properties. Thus, a more explicit list of conditions needs to be listed in Table 1. The chamber samples/conditions should be added or a separate table added for these. The tables for both lab and field should include: representative O/C ratio of aerosol (from AMS) mixing ratios of OH, O₃, NO, and the VOCs already listed in the table T, relative humidity, organic and total aerosol load (e.g., PM₁). This values should be for the specific days the filters were sampled if possible.

V) Related to the above point. The table would allow better evaluation of a comparison of laboratory ozone/a-pinene SOA and isoprene+OH SOA between the chamber and the field. I could imagine that NO (and HO₂/RO₂) levels were quite different between the chamber studies and the field studies. This could change the product distribution of isoprene substantially. This aspect of chemical regime and in how far the field and lab samples can be compared should be discussed. For example, how different do the authors expect a-pinene SOA would be under the AMAZE oxidant conditions compared to ozone/a-pinene lab-aerosol.

VI) The SOA community has become accustomed to often think about SOA in terms

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of O/C ratios. The authors should add a short section briefly presenting what is known about the SOA from the field studies in particular O/C ratio, FTIR and filter extraction analysis and how these compares with the lab samples used in this analysis. For example if the O/C ratios are different the chemical composition has to be different and a broad claim that this method can identify chemical composition is not valid. I think adding this is critical in order to have the SOA community fully appreciate this paper and be able to put it in context.

VII) Have the authors reinvestigated the lab samples after storage time, in case aging of aerosol samples occurred in storage, e.g. via ammonia (Bones et al. 2010 and references therein). I don't expect that this is a big effect, BUT if the method used is surface sensitive it could substantially impact it as the aging/modification will be largest on the surface. This affect could also be different for different types of aerosol.

Specific comments:

1. P. 16395 line 9: "similar in chemical composition" This statement should be modified. I would say something along the lines of that it is "consistent with respect to property X" to synthetic material. The spectroscopic information retrieved clearly cannot be equated with a full chemical compositional analysis. Also the abstract should make clear whether this statement is derived from bulk-aerosol or aerosol-surface measurements.

2. P.16395 line 11-21. The phrasing in this section also should be modified to be more specific about what the results state given the comments above with respect to what is being compared as it cannot be stated with certainty that the ambient conditions and lab conditions are comparable (NO mixing ratio and RO₂/HO₂/NO ratio).

3. P. 16937 line 10-11. The statement on future climate seems out of place. Unless this is addressed later it should be eliminated.

4. P. 16397 line 18. How big was the total sampled air volume so one can deduce the

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inferred aerosol loads.

5. P. 16939 line 24-end. Can these mass loadings be compared with those of HUMPPA

6. P. 16940 line 4: Did the authors prepare “model compounds”, which sounds like specific chemicals, or “model SOA,” which is a mixture?

7. P. 16943 end of first paragraph. Here it would also be useful to state clearly and simply whether aerosol surface or aerosol bulk properties are measured.

8. P16946 line 7-8. Please add a statement whether the spectra of -a-pinene, -b-pinene or +limonene look different from their enantiomers.

9. P. 16947 line 20. “The . . . spectra . . . shows” should be “show”

10. P. 16948 line 24. “absent” do the authors mean “present”. Otherwise the statement is confusing. . .

11. P.16498 line 21. The lack of variability between clean and more polluted air (supposedly) for July 17 and July 21 is very interesting and it would be interesting to see more discussion. Also, can NO and NO_x be compared for the two days. Surely there are measurements.

12. P.16950 lines 3-7 confused me. I think the contrast between the filter samples needs to be made clearer (which one was sampled how long and when. . .)

References: Bones, DL ; Henricksen, DK ; Mang, SA ; Gonsior, M ; Bateman, AP ; Nguyen, TB ; Cooper, WJ; Nizkorodov, SA Appearance of strong absorbers and fluorophores in limonene-O(3) secondary organic aerosol due to NH₄(+)-mediated chemical aging over long time scales, Journal of Geophysical Research 115, D05203, 10.1029/2009JD012864 (2010).

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