

Interactive comment on “Ubiquitous Influence of Wildfire Emissions and Secondary Organic Aerosol on Summertime Atmospheric Aerosol in the Forested Great Lakes Region” by Matthew J. Gunsch et al.

Anonymous Referee #1

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Summary:

Aerosol composition measurements from an Aerodyne AMS and single-particle ATOFMS are presented for approximately 10 days at a rural ground site in Northern Michigan. Observations and HYSPLIT back trajectories suggest the site was influenced by a mixture of “background”, wildfire, and urban sources. Secondary organic aerosol was the dominant aerosol species by mass regardless of source region, and the majority of particles throughout the campaign were identified as having originated from biomass burning. While the ubiquitous nature of biomass burning in the region is

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interesting, the paper might benefit from additional analysis to overcome the shortfalls of having such a short time series. The paper is certainly appropriate for ACP and should be published after satisfying the following critiques.

Major Comments:

1. What metric is being used to separate the wildfire and “regional background” time periods? Figure 1 would suggest that ozone and PM_{2.5} mass/number concentrations are of similar magnitude between the two sources. Likewise, the relative compositional breakdown is identical in Figure 3. I believe it is a bit misleading to label one of these times as “regional background” when they both seem equally influenced by diffuse smoke. If the authors have used a quantitative metric for isolating these sources, please highlight this method in the revised text. The similarity in back trajectories does not seem sufficient from Figure S2. I suggest only presenting “urban” and “background” sources.
2. Ozone mixing ratios are quite low throughout the measurement period, as low as 10-20 ppb during background and wildfire periods. This seems anomalous for summertime North America. Please provide more context/validation for these measurements, especially with respect to your conclusion that the region was frequently influenced by transported smoke.
3. Your result of persistent biomass burning influence is somewhat in contrast to previous reports that biogenic emissions play a role in SOA production (and thus aerosol mass concentrations), e.g., Sheesley et al. (2004). Has something fundamentally changed in the transport pathways to the region, or can these differences be attributed to the VERY short duration of observations presented here?
4. The AMS and ATOFMS have very different sampling capabilities with respect to particle size. This is evident in the vastly different mass concentrations presented in Figure 3 and Figure 5. Can you comment on comparisons between the two measurements, in the context of measured size distributions shown in Figure S4. Do you think

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the number-based prevalence of biomass burning is consistent with AMS results given that you are only assessing the tail of the mass distribution?

5. Rationale for this work is attributed to model under-prediction of OC in the Great Lakes region (i.e., in the abstract line-4), but the source of SOA is not entirely clear from the paper as written. The real question that I don't think is answered with this study is whether the precursor organic material contributing to the significant SOA mass coating biomass burning cores originated from the fires or from more-local biogenic sources encountered during transport. Can AMS (or ATOFMS) mass spectra help assess the SOA source? AMS source apportionment often utilizes the positive matrix factorization (PMF) technique to answer this sort of question and might provide very useful insight into SOA formation processes that could inform the model/measurement disconnect.

6. The buildup of urban emissions culminating in, presumably, a frontal passage at night on 7/22 is an interesting aspect of the sampling period. During this time, it seems that sulfate aerosol is potentially being produced during transport and, as you state, aerosol properties like hygroscopicity and CCN activity are likely significantly altered. Can the authors comment on the meteorological conditions conducive to this sulfate production, and whether situations like this might be an important moderator of aerosol chemistry in the region?

Minor Comments:

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8/2: It is not clear at all from the N and M size distributions that this statement is true. Figure S4 shows the smallest $dM/d\log D$ peak for urban cases, opposite of the conclusion in the text. Please revise and be more specific.

8/25: The presence of Na and Ca attributed to Great Lakes or seawater sources is interesting. I understand that the additional manuscript will likely cover these details, but it might be useful to state here whether ratios are consistent with sea-salt or con-

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tinental dust. Further, soil dust is often lofted into the atmosphere in conjunction with biomass burning, is there any evidence for this during smoke transport? Whether this dust was coated with SOA or sulfates would also be a nice result, based on ATOFMS measurements.

10/3: Does the AMS mass spectrum support your hypothesis that these background particles are mostly SOA coated on a biomass burning core? $m/z=60$ has typically been used to quantify organic aerosols from biomass burning, can a lack of $m/z=60$ contribution be used to support the fact that most of the organics are not from combustion?

13/5: Given the source region for the biomass burning emissions, fires were more likely from agricultural burning. Were there any notable differences that could be highlighted in the mass spectrum or chemical properties of these particles compared to wildfires?

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