

Review of “Seasonal differences in formation processes of oxidized organic aerosol near Houston, TX” by Q. Dai et al.

General Comments:

This manuscript presents measurements of aerosol composition made with an Aerodyne HR-AMS in Houston, TX. The measurements were carried out in two different seasons, and the focus of the analysis is on differences in the OA composition and sources during these different times. Overall, this is a novel data set and the topic is certainly appropriate for *ACP*. The writing is generally good and the manuscript is well-organized. I do have a number of major issues with the manuscript – some addressed here, some in the section below – that prevent me from endorsing it for publication at this time. It may be suitable for publication after a major revision. My greatest concern deals with the analyses and discussion related to Figure 9 (lines 482 - 494) and Figure 11 (lines 550 - 560). This seems like the definition of “cherry picking” data to support one’s view, when the entire data set does not. There is no rationale for excluding such large amounts of data until one achieves a good linear fit. It supports the authors’ narratives, but I think the conclusions involving these Figures (which are central to the entire manuscript) need substantial revision since they are not consistent with the data.

My additional issues with the manuscript are detailed below:

Specific Comments:

- I think that the quantification of the aerosol organic nitrates (ON) have a large uncertainty that needs to be discussed. Equations 3, 4, and 5 indicate the derived ON concentrations are very sensitive to the R_{ON} value. Although the authors have used an R_{ON} value from a very well cited source, there is major uncertainty because the source they cite is based upon a study of SOA from β -pinene oxidation by the nitrate radical. Clearly, the ON formation in this study will be more complex, which adds significant uncertainty to the R_{ON} value and thus to the derived ON concentrations. Much more discussion of this point, including bounds on the ON concentration is warranted.
- The COA factor seems quite problematic given that 1) it is present in winter but absent in summer (cooking is presumably still occurring in the city during this period?), and 2) the diurnal profile of COA (Fig. 4) is inconsistent with both cooking activity and results from many other urban areas.
- The brief discussion on aerosol acidity (Lines 318 - 320) is completely wrong: briefly, the thermodynamic modeling did not include gas-phase ammonia or nitric acid, which significantly limits the ability to characterize acidity. See the extensive body of work from R. Weber and A. Nenes on this topic.
- I had a lot of difficulty with Figure 1 and the associated discussion (lines 349-360). I realize many published papers (including many in *ACP*) follow this standard formula for a paper reporting the results from a ground-based field study. However, I find it almost impossible to actually get anything useful out of Figure 1 – there is simply too much data

presented in too small a space. This is especially true for the discussions about pollutants and wind direction, which cannot be distinguished in Figure 1. If this discussion is central to the manuscript, then additional figures in the Supplemental are likely necessary. If not, then I'd suggest removing (or at least greatly modifying) Figure 1.

- Perhaps this is just a miscalculation, mis-labeled figure or a typo in the manuscript, but the ON concentration estimates (12% and 37% of OA) do not seem consistent with the results in Figure 2, Figure 6, and Table 1. For example, Fig. 2 lists ON contributions to NR-PM1 as 3.4% and 1.5% in winter and summer, respectively. Based on the reported averages (NR-PM1 concentrations of 6 and 3.6 $\mu\text{g}/\text{m}^3$ in winter and summer, respectively), this would give ON concentrations of 0.204 and 0.054 $\mu\text{g}/\text{m}^3$. These levels do not seem consistent with Figure 6, nor with the reported contributions to OA.
- All of the discussion about wet removal is misguided (lines 448-452, 468). The authors seem to imply here that the highest levels of aerosol LWC correspond to periods of precipitation. I seriously doubt that is the case, as precipitation events will greatly reduce all of the aerosol species, as well. Either way, the authors should have access to accurate precipitation data, so this point should be backed by evidence rather than speculated upon.
- The discussion linking MSA with aqueous processing is confusing (lines 507 – 520). It is entirely possible for aqueous processing to produce OOA and at the same time for the OOA factors to exhibit weak (or no) correlations with MSA (e.g., if the air mass had a continental origin).
- This is a relatively minor point, but I question the label of “summer” applied to the May measurements. Can the authors use comparison to prior measurement campaigns in Houston to show that May is representative of summertime conditions in terms of source influences, emissions, chemistry, etc.? Further, because of the short duration of the winter measurement period (2 weeks), the limitation that this campaign may not have fully characterized the winter season in Houston should be discussed.
- The paragraph in lines 64-70 seems contradictory with the current results: the reported measurements seem to indicate that Houston is well below the current (and future) standard.
- I understand that it is common to sample an AMS downstream of a Nafion drier (lines 150 - 152), but can the authors comment on potential artifacts from this measurement setup? E.g., the potential loss of semi-volatile organics (see El-Sayed et al., 2016, <https://pubs.acs.org/doi/10.1021/acs.est.5b06002>).

Technical Corrections:

The above issues are substantial enough that any technical corrections can be addressed on review of the revised manuscript.