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Reply on Anonymous Referee #1

Alexandra J. Boris et al.

Author comment on "Quantifying organic matter and functional groups in particulate matter filter samples from the southeastern United States – Part 2: Spatiotemporal trends" by Alexandra J. Boris et al., Atmos. Meas. Tech. Discuss.,
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Interactive comment from Anonymous Referee #1

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Please see responses in italics

This manuscript reports an original analysis of 6 years of filters collected from 4 different monitoring sites in the SE US. The application of the method is novel and the analysis is original. The results provide interesting insights on trends in PM_{2.5} during this period, which could fit equally well in ACP rather than AMT. Authors provide substantive comparisons of their results to relevant literature, and generally this is well done and sufficient for publication. In sum, this is a very high-quality set of original measurements by a very experienced group and merits publication.

We appreciate your positive assessment of our work.

My only misgiving is the extent to which the technique is different from the cited work. I ask the authors to revise the manuscript to be specific about this to merit publication in AMT rather than ACP.

We have adjusted our manuscript to emphasize the differences between our work and other cited work in the following ways:

- *Added to the abstract: "To the best of our knowledge, this is the longest time period over which this type of analysis has been applied, and this work also demonstrates the application of a more chemically complete and less destructive method than in prior work using alternate techniques."*
- *In Section 1.3, the "Research Statement", we added a clause to distinguish the role of our first paper within this series: "FG calibration models described by Boris et al. (2019; part 1 of this study, which describes the method development) were applied to SEARCH spectra in the current study to accomplish the following goals"*
- *In Section 1.3, the "Research Statement", added a sentence to highlight the novel measurements being made in this paper: "We herein present the application of FT-IR spectrometry to eight years of routinely collected SEARCH network PTFE ambient aerosol filter samples to examine trends in OM concentrations, OM/OC and their FG*

composition from 2009 to 2016. These quantities have not been directly measured in SEARCH network data prior to this work."

- *In the Conclusions, added a sentence at the beginning to summarize the distinct work discussed in this manuscript: "Multi-annual, seasonal, and daily trends in OM concentrations and composition were examined in PM₅ from the SE U.S. using FT-IR spectrometry and PLS regression."*

We thank the reviewer for suggesting that our work might merit publication in ACP; however, we would like to publish in AMT as this paper would be the second in a series, and we selected this journal for this pair of papers recognizing that neither AMT nor ACP would be a perfect fit for both works.

As a note, I find the most interesting part of the discussion to be about the contribution of fires to particular functional groups, but that seems a bit buried in the Discussion and is omitted from the Abstract, Title, and Conclusions.

We agree that the apparent correspondence of observed fires and variability in the daily (2016) OM concentrations is an interesting observation. Unfortunately, a smoke "fingerprint" within FG composition was not possible to discern in this work: as demonstrated in figure S1, while one composition of fire-impacted samples (as identified using satellite images) was observed in daily 2016 samples, another was observed in 2011-2016 samples collected every three days. We believe that this satellite imagery-based method of categorizing "fire" vs. "no fire" samples, as well as other factors affecting smoke composition and dilution arriving at the sampling sites, were responsible for this lack of observable smoke FG "fingerprint". We have updated our text to be clearer about this in the manuscript (Section 3.6, page 20).

We have also included clarification on the discussion of fire contributions in the following ways:

- *Added the ending clause to this statement in the abstract: "Daily samples from 2016 further elucidate the consistent impact of meteorology and biomass burning events on shorter term OM variability, including prescribed burning in the winter/spring and wildfires in the autumn, although these sources did not appear to be strong contributors to long-term OM or composition trends in the SE U.S."*
- *Added this sentence in the conclusions: "A fire "fingerprint" of FG composition was not, however, apparent in the present work, despite the clear relationship with overall OM concentrations, perhaps due to dilution or other obfuscating factors."*

To that point the authors note the potential usefulness of PMF, but apparently did not have the time/resources to do this, which seems a shame. Perhaps future work on this is planned?

Unfortunately, PMF was outside the scope of this particular paper, but we agree that it could be quite useful, and hope to perform PMF analysis using these data and similar datasets in the future.

Other questions:

1. to volume or are volumes same?

All samples in this work were collected over 24 hours at 16.7 liters per minute (24.0 m³). Section 2.5 (page 7) states that, "when available, if the observed flow rate, duration, or total volume was not 100±5 % of the expected value (e.g., <23.75 of 24.0 expected hours), the sample was excluded from the dataset; samples with three or more null flow observations were assumed to be inaccurately sampled and were also excluded."

The following text was added to Section 2.1 (page 5) to clarify this: "All samples were collected over 24 hours at 16.7 liters per minute (24.0 m³; see Section 2.5 for outlier handling). Filters were shipped and stored at < 4 °C (from Aerosol Research and Analysis, Inc., ARA, in Morrisville, NC) to minimize loss of volatile species."

2. Fig4 annual median spectrum is an interesting concept; can you provide more details about how this was determined?

We kindly refer the reviewer to our Supplement, Section S5, which contains a description of the method of median spectrum calculation (briefly, baseline correction and blank filter feature subtraction followed by median calculation at each wavelength). Figure S6 also shows the median spectra of other sites and years.

3. Table 2 – trends are not consistent; can you provide explanations for lack of consistency?

The trends in this table are expressed in terms of the changes in functional group (FG) concentrations over time, and the changes in the ratios of FG concentration to the total organic matter (OM) concentrations (FG/OM; Figure 3 trends are also as FG/OM). The normalization to OM in the latter category causes some trends to differ substantially in magnitude, and even direction, in some cases.

In response to this and another related reviewer comment, we have moved this table to the Supplement. Important trend values have been added within the text of the manuscript. We hope that the manuscript is more succinct without this complex table.