Atmospheric Measurement Techniques Discussions

Atmos. Meas. Tech. Discuss., referee comment RC2
https://doi.org/10.5194/amt-2020-401-RC2, 2021

Comment on amt-2020-401

Anonymous Referee #2

Referee 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., https://doi.org/10.5194/amt-2020-401-RC2, 2021

Review of "Quantifying organic matter and functional groups in particulate matter filter samples from the southeastern United States - Part 2: Spatiotemporal Trends" by Boris et al.

This manuscript describes FTIR analysis coupled with multivariate calibration of specific functional groups (aliphatic C-H, carboxylic COOH, oxalate O=C-O-, non-acid and carbonyl C=O and alcohol O-H) in filter-based fine PM samples collected at the surface over 8 years at 4 sites in the southeastern United States as part of the former SEARCH air quality network. The authors find that there is a decline in organic mass that is driven primarily by carboxylic acid and oxalate functional groups. They attribute this to reductions in anthropogenic emissions of SO2 and/or VOCs. There is a lot of analysis and the supplemental information is extensive.

Control experiments and quality assurance are discussed in the supplemental information (SI). It is difficult to understand the impact of not storing samples frozen over several years, and the SI indicates there is chemical change. Looking at Figure 4, my first instinct was the annual trend is related to sample degradation - I actually think a more thorough discussion in the main text would help the authors' case. Further, the authors attempt to make links to temporal trends among NOx or O3 with specific FTIR-derived functional groups almost exclusively with literature concerning ambient data. It's my opinion that links to laboratory literature with reference to specific spectra wavenumbers help make their arguments stronger.

I found Table 2 very confusing. It appears the aCH trend is driven by 'just' BHM summer, COOH 'just' in the summer, but many other values are presented. I understand how this would be value for careful readers. However there are more compelling plots in the SI that I think would make better use of the main text space.

Page 4, Line 16: The authors state fossil fuel combustion may contribute substantially to OM in the SE U.S. …. I think the authors here are referring to the carbon component specifically. It is well established that fossil fuel combustion aids OM formation, e.g., impacts on the NOx/oxidants and POA are known for over a decade...work by Lane,

Griffin, Carlton.

Page 11, Line 6/7: I do not follow the logic behind the statement that OM trends are unrelated to changing PM2.5 concentrations due to lack of trend in Si and K. These species