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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantifying primary and secondary humic-like substances in urban aerosol based on emission source characterization and a source-oriented air quality model" *by* Xinghua Li et al.

Anonymous Referee #1

Received and published: 29 October 2018

The manuscript is a comprehensive and well-structured study on the potential sources of HULIS, a ubiquitous and abundant atmospheric aerosol constituent. Besides the fact that it is based on a surprisingly extensive experimental setup covering different source measurements and long-term field sampling and observations, it also has a touch of novelty in that identifies residential coal burning as a potentially important yet previously ignored source of primary HULIS. The methodology applied in the manuscript is widely accepted by the aerosol community and its use makes the comparison with the results of other publications feasible. Although the study involves only the analyses of key

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aerosol constituents such as OC, WSOC, HULIS and inorganics, its conclusions are largely well-founded by the results of the measurements. There are just a few issues that raise some questions in the reviewer.

1) Except for the summer, HULIS are much better correlated with PM2.5 than with OC (Page 8, Line 220 and 224). This is surprising in the light of the fact that HULIS is actually part of OC whereas PM2.5 contains all sorts of other constituents. Not surprisingly, the correlations are the best for WSOC, the closest relative of HULIS. Is there any possible explanation for these observations? Perhaps the effects of vehicular exhaust contributing to OC (and EC) but less to PM2.5 mass concentrations?

2) In sub chapter 3.2 the differences in HULIS-to-OC ratios of biomass combustion emissions between this study and many other studies around the world are stunning. There are differences by factors of 3–5. The manuscript actually claims that nearly half of the OC are HULIS. Since these are emission measurements on biomass that should not be fundamentally different in different regions (albeit significant differences are seen between various species), there should be something in the experimental setup that causes these unusually high readings. Differences in combustion conditions, dryness of fuel, dilution ratios and excessive cooling may explain these high values. A comparative and critical assessment of the results with those of similar studies would be useful. This is critical since the source apportionment of primary HULIS is based on these emission values.

3) I would strongly discourage the application of simple correlations for secondary formation processes (sub chapter 3.4.2). These mechanisms are too complex to be captured by simple regressions: emission fluxes of precursors, rates of transformations, volatilities and water-solubilities of the reaction products, cloud-processing mechanisms, are all different and the processes are strongly non-linear. If, for example, HULIS is not correlated with sulfate, it may also mean that though they are both of secondary origin, the sources and emission fluxes of their precursors are very much different. Therefore lack of correlation does not indicate anything, neither does some

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moderate virtual correlation. Just think of the examples of sulfate and nitrate, both being secondary aerosol constituents, yet they exhibit completely different formation mechanisms relative to the emissions of their precursors.

Minor comments:

Typography throughout the manuscript: the improper use of hyphen instead of En dash and Minus characters.

Page 5 Line 122 'systemis' . . . space missing

Page 5 Line 122 'induced' . . . introduced?

Page 5 Line 124 'at ambient temperature' . . .below ambient temperature?

Page 5 Line 142 'measurements was' . . .were

Page 6 Line 154 'determination' . . . determined

Page 7 Line 198 'General of ambient'

Page 9 Line 239 'HULIC'

Page 9 Line 245 Please define 'WSOM'

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