

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 #2

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This work integrates ambient, source sample measurements and modeling investigation to quantify HULIS sources in Beijing. This integrative approach provides quantitative insights into HULIS sources that otherwise are not easily extracted from source and ambient measurements alone. The paper is well-written and easy to follow. I have one main concern regarding the estimation of secondary HULIS. It is estimated to be the difference of measured HULIS and modelled primary HULIS. The difference method is inherently associated with large uncertainty and it appears less reliable (see more details in the specific comments). Any overestimate in primary HULIS would translate to

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underestimate in secondary HULIS. It is desirable that the authors conduct a receptor model source apportionment (such as positive matrix factorization) using the measured chemical composition to estimate the secondary HULIS contribution and inter-compare with the results obtained from the CMAQ model.

Specific comments:

1. Model evaluation of HULIS. Fig. 4 compares predicted primary HULIS_c and observed HULIS_c on days with relative good primary PM_{2.5} model performance. In the main text it is reported fractional error of less than 0.6 was used to select the good model performance data. What is the percentage of data in this work's dataset fall outside this criterion of good modeling performance? Are there any patterns in the sub-group of data with poor agreement?
2. In this work, contributions of HULIS_c from secondary processes were determined by subtracting predicted primary HULIS_c from observed HULIS_c. The percentage contribution of secondary process was 40.2% in summer, 52.7% in fall, 14.3% in winter and 13.1% in spring. The secondary HULIS contribution was surprisingly low, considering the strong correlations of HULIS with secondary PM components such as sulfate and estimated SOC, especially for winter samples (Figure 3)
3. Related to the previous comment, and also the fact that on some days the predicted primary HULIS_c concentrations are greater than the observed HULIS_c, I have the concern whether certain assumptions made in the model have led to positive bias for primary HULIS_c (therefore negative bias for secondary HULIS_c) (e.g., assumption of foc values, see the next comment). How many samples were predicted by the model to have negative secondary HULIS_c? Are there any common characteristics in these samples that might shed some insights for the potential bias?
4. Table S3 lists the values of foc for primary sources considered in the model. “Residential” source has the largest foc at 62.80%. It appears this residential source is residential coal combustion (#91028) (Ying et al, 2018). Was this Residential source

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foc also applied to residential biofuel burning? If yes, is there supporting evidence for this assumption? The apportionment of primary HULIS sources by the CMAQ model in this work suggested that residential biofuel burning was the largest HULIS source year around (34-70%), and especially dominant during winter and spring (70%). The foc in open biomass burning (arguably a burning activity bearing similarity to residential biofuel burning) is 29.40%, only $\sim 1/2$ of the foc for residential coal combustion. Apparently, the foc value assumed has a large impact on the modeled source contribution. The authors need to clarify what foc value is adopted for residential biofuel combustion and the rationales behind.

5. Please comment on other potential primary HULIS source, such as cooking, which might make a contribution, but are not considered in the current model.

6. Table 2: provide a table footnote to briefly explain the abbreviations for the different residential coals.

7. Table S5: add a note to indicate the comparative relationship of this table with Table 3 in the main text.

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