Responses to Reviewer #2

General comments.

Although lots of data have been analyzed and discussed in the manuscript, however, some additional MS/MS experiments and Kendrick mass defect analysis are encouraged. Please refer specific comments in the attached PDF.

Re: Thanks for the comments. All your suggestions are of great guiding significance to us. We revised the manuscript according to your suggestions as follows.

1. The Basically, authors have discussed a lot regarding the homologous series. I think the Kendrick mass defect should be more suitable to this purpose, particularly for two-dimensional Kendrick mass defect plots such as KMD_CH2 versus KMD_SO3.

Re: Thanks for the good suggestion. We have added the KMD plots (Figure S6) (page 12, supplemental information). The *x*-axis direction indicates hydroxylation of the compounds, with different amounts of hydroxyl; The *y*-axis direction represents methylation, which varies in quantity and length of carbon chains.

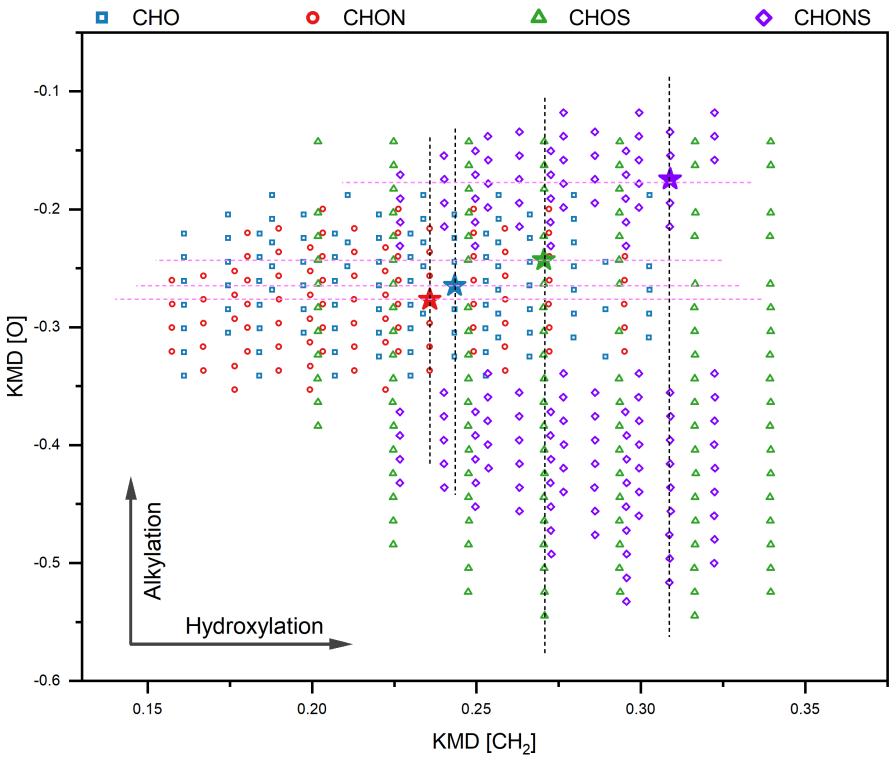

Figure S6. Two-dimensional Kendrick mass defect (KMD) matrix plot for CHO, CHON, CHOS and CHONS compounds of High SOA-loading-D sample. The KMD [O] denotes the

Kendrick mass defect of hydroxyl functional group (-OH). The KMD [CH_2] denotes the Kendrick mass defect of methylene group (CH_2). The magnified star symbols represent $C_{16}H_{24}O_8$, $C_{15}H_{25}NO_8$, $C_{15}H_{24}O_7S$ and $C_{10}H_{17}NO_9S$ with relative high abundance, respectively.

2. Authors should confirm this statement using MS/MS.

Re: According to previous studies, the ionization capability of nitro (-NO_2) and organonirate (-ONO_2) is strong in the ESI(-) mode (Song et al., 2018). Some CHON compounds with relative high abundance, such as $C_7H_7O_4N$ and $C_7H_5O_5N$, have been identified as typical markers of biomass burning with nitro (-NO_2) and organonirate (-ONO_2) structure in previous studies (Laskin et al., 2015). Thus, it is inferred that the CHON compounds in this study contain nitro (-NO_2) and organonirate (-ONO_2) structure. Of course, further confirmation with MS/MS does provide more evidence. This part of experiment will be made up in further research.

3. what does 'ture' mean? (line15 page8)

Re: Thanks for the good suggestion. We have modified this sentence (lines 14-16, page 8).

"The intensity-contribution of daytime samples is 23.9-25.4% higher than that of corresponding nighttime samples at moderate ones (Figure 2a-d), while it was opposite under the low SOA-loading, with a 3% higher intensity-contribution at night (Figure 2e, f)."

4. Authors should confirme this argument with MS/MS.

Re: According to previous studies, the ionization capability of sulfate group (-OSO_3H) is strong in the ESI(-) mode (Xie et al., 2020). CHOS compounds with O/S ≥ 4 are the peaks with relative high abundance in each mass spectrum. It can be inferred that these S-containing compounds contain sulfate group (-OSO_3H). Moreover, previous studies have shown that sulfur-containing organic matter formed by oxidation of biogenic VOCs are mainly organosulfates, and MS/MS results also show that it contains sulfate functional group (Surratt et al., 2008).

Of course, further confirmation with MS/MS does provide more evidence. This part of experiment will be made up in further research.

5. ESI(-) (line 15 page 8).

Re: Thanks for the good suggestion. We have modified this word (line 23, page 8).

"ESI(-)"

References:

Laskin, A., Laskin, J., and Nizkorodov, S. A.: Chemistry of atmospheric brown carbon, Chem Rev, 115, 4335-4382, 10.1021/cr5006167, 2015.

Song, J., Li, M., Jiang, B., Wei, S., Fan, X., and Peng, P.: Molecular Characterization of Water-Soluble Humic like Substances in Smoke Particles Emitted from Combustion of Biomass Materials and Coal Using Ultrahigh-Resolution Electrospray Ionization Fourier Transform Ion Cyclotron Resonance Mass Spectrometry, Environ Sci Technol, 52, 2575-2585, 10.1021/acs.est.7b06126, 2018.

Surratt, J. D., Gómez-González, Y., Chan, A. W. H., Vermeylen, R., Shahgholi, M., Kleindienst, T. E., Edney, E. O., Offenberg, J. H., Lewandowski, M., Jaoui, M., Maenhaut, W., Claeys, M., Flagan, R. C., and Seinfeld, J. H.: Organosulfate Formation in Biogenic Secondary Organic Aerosol, The Journal of Physical Chemistry A, 112, 8345-8378, 10.1021/jp802310p, 2008.

Xie, Q., Li, Y., Yue, S., Su, S., Cao, D., Xu, Y., Chen, J., Tong, H., Su, H., Cheng, Y., Zhao, W., Hu, W., Wang, Z., Yang, T., Pan, X., Sun, Y., Wang, Z., Liu, C.-Q., Kawamura, K., and Fu, P.: Increase of High Molecular Weight Organosulfate with Intensifying Urban Air Pollution in the Megacity Beijing, Journal of Geophysical Research Atmospheres, 125, 10.1029/2019JD032200, 2020.