Anonymous Referee #1

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 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.

Response: We thank the reviewer #1 for providing helpful comments and suggestions to improve our manuscript. Below are our responses to reviewer comments including descriptions how we have modified the manuscript.

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?

Response: Both HULIS and OC are strongly correlated with $PM_{2.5}$, indicating that they have similar sources such as biomass and coal burning, secondary processes. Perhaps the effects of vehicular exhaust contributing to OC (and EC) but less to PM2.5 mass concentrations, which need to explore in the future.

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.

Response: We summarized the HULISc/OC and HULIS/WSOC values from biomass burning (see the following Table). We think the combustion condition have much influence on the HULIS-to-OC ratios. For biomass open burning, HULIS-to-OC ratios varied less (from 0.14-0.35), while for biomass burned in the stove, ratios varied a lot (from 0.01-0.50). For advanced stove used in European (with secondary air), combustion is relatively complete, thus HULIS produce less (0.01-0.12), while for stove used in Chinese rural household, combustion is relatively inadequate, thus HULIS produce more (0.41-0.50). Dilution ratio (DR) and residence time (RT) could affect gas-particle partitioning, and thus also have effect on the results (Lipsky et al., 2006; May et al., 2013). Dryness content of fuels was found to be not correlated with HULIS-to-OC ratios.

We added a comparative and critical assessment of the results with those of similar studies in the revised manuscript and the following table was added in the supplement.

Summary of HULISc/OC and HULIS/WSOC values from biomass burning

Biomass	Combustion condition	Sampling condition	HULIS$_C$/OC	HULIS$_C$/WSOC	Reference
Wood (M=9.3%)	Improve stove	Chamber/hood DR≈40, RT≈80s	0.41±0.07	0.62±0.06	This study
Wheat straw (M=9.8%)	Improve stove	Chamber/hood DR≈40, RT≈80s	0.50±0.04	0.65±0.05	This study
Maize stover (M=8.0%)	Improve stove	Chamber/hood DR≈40, RT≈80s	0.42±0.04	0.62±0.04	This study

Wood (M=7~14.8%)	Chimney type logwood stove with primary/secondary air	Dilution source sampler with DR=10, RT long enough (no specified)	0.04-0.11		Goncalves et al., 2010
Wood (M=10~16%)	Domestic tile stove	Dilution sampler with DR=3, RT=0.2s	0.01-0.12		Schmidl et al., 2008a
Leaves (M=25%)	Open burning	Smoke plume	0.33-0.35		Schmidl et al., 2008b
Rice straw (M=5.8%)	Open burning	Chamber		0.66±0.02	Fan et al., 2016
Corn straw (M=7.4%)	Open burning	Chamber		0.59±0.02	Fan et al., 2016
Pine branch (M=7.6%)	Open burning	Chamber		0.57±0.03	Fan et al., 2016
Rice straw	Open burning and chamber	Chamber/hood or downwind	0.34±0.05		Lin et al., 2010a
Sugarcane leaves	Open burning	Chamber/hood	0.28±0.03		Lin et al., 2010a
Charcoal	Open burning	Downwind	0.32		Lin et al., 2010a
Rice straw	Open burning	Downwind	0.14	0.33±0.02	Lin et al., 2010b
Sugarcane	Open burning	Downwind	0.15	0.30±0.01	Lin et al., 2010b
Rice straw (M=7.8%)	Open burning	Chamber/hood	0.26±0.03	0.63±0.05	Park and Yu, 2016
Pine needles (M=9.9%)	Open burning	Chamber/hood	0.15±0.04	0.36±0.08	Park and Yu, 2016
Sesame stems (M=10.3%)	Open burning	Chamber/hood	0.29±0.08	0.51±0.08	Park and Yu, 2016

Note: M, DR and RT are the abbreviations of Moisture, Dilution Ratio and Residence Time, respectively.

Reference:

Fan, X., Wei, S., Zhu, M., Song, J., and Peng, P.: Comprehensive characterization of humic-like substances in smoke $PM_{2.5}$ emitted from the combustion of biomass materials and fossil fuels, Atmos. Chem. Phys., 16, 13321–13340, 2016.

Goncalves, C., Alves, C., Evtyugina, M., Mirante, F., Pio, C., Caseiro, A., Schmidl, C., Bauer, H., and Carvalho F.: Characterisation of PM_{10} emissions from woodstove combustion of common woods grown in Portugal, Atmos. Environ., 44(35): 4474-4480, 2010.

Lin, P., Engling, G., and Yu, J.Z.: Humic-like substances in fresh emissions of rice straw burning and in ambient aerosols in the Pearl River Delta Region, China. Atmos. Chem. Phys., 10, 6487-6500, 2010a.

Lin, P., Huang, X.F., He, L.Y., and Yu, J.Z.: Abundance and size distribution of HULIS in ambient aerosols at a rural site in South China, J. Aerosol Sci., 41, 74–87, 2010b.

Park, S. S. and Yu, J.: Chemical and light absorption properties of humic-like substances from biomass burning emissions under controlled combustion experiments, Atmos. Environ., 136, 114-122, 2016.

Schmidl, C., Marr, L. L., Caseiro, A., Kotianova, P., Berner, A., Bauer, H., Kasper-Giebl, A., and Puxbaum, H. Chemical characterisation of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions, Atmos. Environ., 42, 126–141, 2008a.

Schmidl, C., Bauer, H., Dattler, A., Hitzenberger, R., Weissenboeck, G., Marr, I. L., and Puxbaum, H.: Chemical characterisation of particle emissions from burning leaves, Atmos. Environ., 42, 9070-9079, 2008b.

Lipsky, E. M., and Robinson, A. L.: Effects of dilution on fine particle mass and partitioning of semivolatile organics in diesel exhaust and wood smoke, Environ. Sci. Technol., 40(1), 155-162, 2006.

May, A. A., Levin, E. J. T., Hennigan, C. J., Riipinen, I., Lee, T., Collett, J. L., Jimenez, J. L., Kreidenweis, S. M., Robinson, A. L.: Gas-particle partitioning of primary organic aerosol emissions: 3. Biomass burning, Journal of Geophysical Research-Atmospheres, 118(19): 11327-11338, 2013.

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 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.

Response: we have deleted the sub chapter 3.4.2.

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’

Response: We revised these grammatical errors accordingly.