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Comment on acp-2022-464

Anonymous Referee #1

Referee comment on "Measurement report: Changes in light absorption and molecular composition of water-soluble humic-like substances during a winter haze bloom-decay process in Guangzhou, China" by Chunlin Zou et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-464-RC1>, 2022

This manuscript describes measurements of light-absorption properties and chemical composition of ambient water-soluble HULIS samples collected during haze episodes and clean periods. The measurements involve comprehensive chemical analyses including carbon analysis (OC/EC and TOC), ion chromatography for inorganic ions, and ESI FTICR MS for organics. The light absorption properties were quantified using UV-vis spectrometry.

General comments:

- The data set produced in this study, especially the ESI FTICR MS data, is extensive and informative. However, there are various instances where assertions are made that are not supported by the data, and are at some points contradictory with other assertions in different parts of the manuscript. Please see specific examples under 'Specific comments' below.
- The term HULIS as used in this manuscript needs to be better defined. HULIS is a vague term – much like brown carbon – that has been used to refer to different things in different studies. Here, HULIS is obtained based on an extraction procedure that isolates the less polar fraction (~50%) of WSOC. It would be helpful for the reader to explicitly indicate in the methods section that this definition is operational, and also contrast the definition of HULIS in this study with other studies. This is important because the results are compared to multiple previous studies on HULIS, and it should be noted that not all HULIS are defined the same way.

Specific comments:

- Section 2.4 and 2.5 should be combined: ESI-MS is also chemical analysis.
- Line 165: The manuscript presents results of PM_{2.5} concentrations, but there is not description of how the PM_{2.5} concentrations are measured in the methods section.
- Figure 1: There are inconsistencies in the x-axis values: the distance between the major ticks changes between 1 day (e.g. 1/24 – 1/25) and 2 days (e.g. 1/10 – 1/12).
- Figure 1f: how come the Lev/OC value are larger than 1? Lev is one of many OC species.
- Line 218-219: The statement that Lev/OC increased in haze-II is not accurate. There are 2 data point for Lev/OC in haze-II (Figure 1f): one is higher than haze-I and one is lower than haze-I.
- Line 232-233: This is not valid. AAE is a measure of the wavelength dependence of light absorption, not the magnitude of light absorption.
- Line 248-260: The statement on line 251 that MAE of HULIS is generally higher than WSOC is not valid. The values for HULIS (1.1 +/- 0.27) and WSOC (1 +/- 0.21) are virtually the same. In Lines 254-256, MAE of HULIS (1.1 +/- 0.27) is said to be “comparable” to other values ranging between 0.91 and 1.84. Then in line 257-259, MAE of 0.91 is said to be “much lower” than MAE of 1.3. These statements are subjective and inconsistent.
- Line 263-266: The argument that stagnant conditions lead to prolonged oxidation thus lower MAE for haze versus clean days is not convincing. It is not clear that the PM sampled during the haze days had longer atmospheric lifetime / OH exposure. What if the PM in the clean days had more contribution from long-range transported PM?
- Line 285-286: It is not clear how the presence of these 3 molecules suggests contribution from biomass burning and vehicular emissions.
- Line 308-318: This paragraph mentions that HULIS in haze days had higher MW than in clean days, and makes the point that high MW HULIS is more resistant to chemical transformation. This is in contrast with the assertions in section 3.2 that MAE on haze days were smaller than on clean days because HULIS on haze days underwent more chemical transformation.
- Line 319-330: This paragraph makes the point that lower AI_{mod} for haze days can be due to photooxidation during haze days and explain that lower MAE for haze days. How is this assertion reconciled with the larger MW and resistance to oxidation mentioned in the previous paragraph?
- Line 374-378: It does not look like the statement “relatively low BBOA content” is supported by the data in Figure 3. Most of the molecules are clustered in the region identified as BBOA. In any case, previous parts of the manuscript mention BBOA as being an important contributor to HULIS measured in this study, but this paragraph mentions that traffic sources are more important.

Minor comments:

- Line 80-83: The statement talks about 'recent years' but is supported by a reference from 2014. A newer reference is needed.
- Line 91: What is meant by 'exact'?
- Line 99-102: This sentence is not comprehensible.
- Line 180-182: This statement is not valid. Wind speed alone does not dictate stability (See stability classifications by Turner 1970). In fact, for an unstable atmosphere, increasing wind speed makes the atmosphere less unstable.
- Line 277-279: Vague statement. In what sense are the peaks "comparable" with peaks from other studies?
- Line 337-339: I assume you mean biomass burning aerosol (not biomass burning mixture). In any case, what does "comparable" mean here?