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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sources of humic-like substances in the Pearl River Delta, China: positive matrix factorization analysis of $PM_{2.5}$ major components and source markers" *by* B. Y. Kuang et al.

Anonymous Referee #1

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Review of "Sources of humic-like substances in the Pearl River Delta, China: positive matrix factorization analysis of PMÂň2.5 major components and source markers"

General comments: This manuscript presents novel insight to the sources of HULIS in the atmosphere through a thoughtful analysis of HULIS, carbon fractions, elements, and organic species and positive matrix factorization (PMF). The methods used are of high scientific quality and are clearly described.This study provides a thorough and meaningful quantitative analysis of the sources of HULIS, which has not previously

been accomplished with the level of detail and rigor shown here. The results are significant, in that they provide quantitative source apportionment of HULIS-C and other carbon fractions to major source categories in the PRD region. The presentation quality could be improved, and clarifications and further discussion of certain points are needed as outlined below.

Specific comments:

1. The observation of HULIS in the ship emission factor is surprising and a novel finding of this paper. The authors need to address the possibility that this HULIS could be associated with secondary processing of this source rather than primary emissions. The PMF model will include both primary and secondary sources in this factor if they sufficiently co-vary. The presence of secondary inorganic ions (e.g. sulfate and ammonium) suggests that some of the primary emissions have been processed.

2. The ship contribution to Hulis C, WSOC-H, WSOC, and WISOC does not appear to vary with wind direction. Why? It would expectedly be greater in marine air masses, so it is surprising that it is not.

3. Alkanes and PAH were measured, but ambient concentrations are neither discussed nor included in the source apportionment model. Perhaps it is best to

4. Please clarify what PM2.5 component was set as the master variable in PMF modeling. I could reasonably assume PM2.5 or HULIS-C, but cannot discern from the text.

5. Page 23918, line 5: Provide the rationale for including HULIS-C in the PMF model rather than HULIS mass.

6. Section 3.1.1. should briefly discuss the PM mass loadings observed in this study, their seasonal variation, and comparison to prior studies in the region.

7. Further explanation is needed on page 23920, line 23-24 – how was the WSOC proportion consistent with their suburban and urban location characteristics?

8. At line 23920, line 21 – a surprisingly large fraction of the OC was water soluble at 96%. Are these results consistent with measurements of PAH and alkanes? What conditions gave rise to such high WSOC levels? Related, and because the goal is to show that WSOC was a significant fraction of OC, perhaps the averages are a better statistic to use than the maxima.

9. Page 23921, lines 1-3 – While the absolute concentrations of WSOC were higher in autumn and winter, but the relative contributions to OC were lower. Why? The absolute levels are likely affected by seasonal patterns and meteorology, which will affect PM mass loadings and other components. Discussion of the relative levels of WSOC across seasons reflects different sources, and ties in with the other objectives of this manuscript.

10. Page 23921, lines 13-15: I am confused by "HULIS and WSOC may differ in their major contributing sources." Isn't HULIS a part of WSOC, so they have to have in part some of the same sources? Or maybe this says there are different sources of WSOC_h? Or maybe the sentence is not clear and should this be qualified to say: "The difference in absolute concentrations of HULIS and WSOC across the two study sites suggests that each site is impacted to different extents by WSOC sources." ?

11. Page 23921, lines 15-22 does an excellent job of comparing results of this study to prior studies. It is this level of discussion that is encouraged to be expected to PM2.5 and other key components in this manuscript.

12. Page 23921 line 27: The correlation between HULIS and levoglucosan does not "indicate that biomass burning was a source of HULIS in the winter", rather it indicates that they covary in time and suggests that they may be derived from the same source.

13. The paragraph beginning on page 23921, line 23 should be combined with the discussion of Figure 4 and section 3.2.1.

14. Because of the seasonal differences, correlations between sulfate and levoglu-

cosan with HULIS may be more demonstrative if broken into "summer" and "winter" in Figure 4.

15. Is there evidence that the two sites are impacted by different types of biomass burning? This could be assessed by comparison of levoglucosan/mannosan, levoglucosan/galactosan ratios or levoglucosan/K ratios across the two sites. Since different types of biomass (i.e. agricultural burning or wood burning) can produce emissions of different chemical composition, this should be ruled out as a potential source of the spatial differences in levoglucosan concentrations discussed on page 23922 line 6. Moreover, these ratios (mentioned at line 23923, line 9-11) can be used to gain insight to the types of biomass impacting the site during the various seasons.

16. Are the two sites impacted to different extents by agricultural burning? This is an important consideration because of its contribution to HULIS.

17. Ensure the reference to HYSPLIT is properly cited.

18. Page 23923, line 22: Provide a numerical statement of the "low correlation" coefficient observed for vehicle, dust, and ship emission tracers.

19. Table 1 contains substantial valuable information, but is rather difficult to follow. The following suggestions are intended to improve the readability and utility of the table: a. The authors should consider significant figures in making this table. The standard deviation should have 2 digits and the mean should be adjusted to match. For example, 12.39 $\pm$ 6.79 for sulfate, should be 12.4 $\pm$ 6.8. b. The mean $\pm$ standard deviation should be in one column and the range in another. Together, the numbers are too close and difficult to follow. c. "C2O4=" should be "oxalate" to match the text. d. It is suggested to break the table into addition sub-sections, "PM2.5 and carbon fractions" "Ions" "Trace elements" and "Organic Tracers" Appropriate units can be given in the sub-title. e. Ti, Ni should not have a minimum value or standard deviation of 0

20. Figure 2 – Can a brighter circle be used to mark the study sites instead of pin? It

is difficult to see, especially in black and white. The red text is also difficult to see.

21. Ensure that consistent units for levoglucosan are used throughout the text and figures – Figure 3 and 4 show ng and ug per cubic metre.

22. Figure 4 – January 26 can be removed from this figure and caption, since its differences due to New Year is noted.

23. The x-axis in Figure 5 should be reordered to follow table 1. In addition, the chemical names should match what is in the text (i.e. oxalate, OC, EC).

24. Page 23920 – line 3: “summary statistics” 25. Page 23929 – line 1: “residual oil”

26. Page 23929 – line 8-10 should be omitted, as it has not been peer reviewed.

27. Page 23930 – For consistency, use NS and GZ abbreviations in conclusion.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 23913, 2014.