

Overview – Peng et al. examine the HULIS content in aerosols emitted directly from rice straw burning. They also investigate the HULIS contained in atmospheric aerosols collected from the Pearl River Delta Region (PRD), China. Generally speaking, HULIS in aerosols is a timely research topic relevant to the ACP journal. Furthermore, the HULIS measurements Peng et al. are providing are unavailable currently for the PRD; thus, these results are potentially useful to the atmospheric aerosol research community. In this reviewer's opinion, the major drawback of the study is that the vehicular and biomass burning source assignments are arbitrary and uncertain. Although K is sometimes used as a biomass combustion marker, it is known to be somewhat unreliable. The source testing for this study seems to also confirm this conclusion. It would be very interesting to compare levoglucosan and HULIS concentration values for the source and atmospheric aerosols. Such a comparison may allow the authors to be less anecdotal about the biomass burning source contributions to HULIS. As it stands currently there is limited direct evidence in the present study for linking biomass burning aerosols to HULIS in the PRD atmosphere, although this link is strongly implied throughout (especially for rice straw burning). At least a statement should be made about specifically when the agricultural or biomass fires typically occur in this region, and the data should be examined for concomitant increases in HULIS. So, a revised paper should include less speculative discussion about what the combustion sources of HULIS are without more evidence. More specific comments for revision are provided below.

Introduction – Although the objectives of the study are clearly stated in the Introduction, information about the main hypothesis being tested is missing. In other words, the authors may want to provide more about what they set out to do. It seems that the attempt was to show that primary and secondary aerosols from biomass burning were the main sources of HULIS in the PRD atmosphere. This should be explicit at the outset.

P 7187, line 5: "theirs" should *its* and "compositions" should *composition*

Experimental – Are there other means to measure HULIS in aerosols? Isn't the definition of HULIS somewhat operational at the moment? If so, this should be briefly discussed in the text.

A coarse and fine ambient aerosol fraction was collected. Yet only data for the fine fraction are presented. The different size fractions can sometimes help with the apportionment exercise. For example, the marine aerosol may show more NaCl in the coarse fraction. Should these data be presented?

P 7189, line 17: insert *area* between "suburban" and "site" and delete "in the suburban area"

Results and Discussion

P 7191, line 24: sentence should read "The emission factors of.....vary with combustion conditions (Andrea and Merlet, 2001).

P 7192, lines 1-7: Check the boiling points of the potassium salts because smoldering fire burns at lower temperatures than flaming fire. This may explain why the K doesn't volatilize during smoldering fire. This observation can be checked for validity by burning rice straw from the same growing location under varying fire conditions. In addition, the discussion here is focused on an enrichment factor, which is not necessarily an emission factor.

P 7192, lines 24-25: please remove "in the presence of oxygen and"

P 7193, lines 14-16: please clearly explain why these samples are considered to be BB-influenced samples. What about the time of their collection with respect to local fire activity?

P 7195, lines 1-8: Many of the organic species being given here (e.g., benzoic acids, alkanes, -oic acids etc.) are emitted from anthropogenic sources including fossil fuel combustion. Why wouldn't these also be combined to form HULIS in this case? It may be fundamentally incorrect to isolate this discussion to biomass burning since identical pollutants emitted from different sources can just as easily be present and interact in the atmosphere. As suggested, the same is true for the NO_x, CO, and VOC compounds being described, many of these are not unique to biomass burning. Furthermore, is there need for such speculative discussion? Why not mention that many of these compounds that potentially react to form HULIS are potentially contributed from many combustion sources. The bottom line is that with

so many unknowns with regard to HULIS formation, the discussion is over-focused on BB as a source of atmospheric HULIS.

P 7196, line 11: The figures should be introduced in the appropriate order.

P 7196, lines 20-22: How do we know that HULIS comprises unresolved POM? Isn't the definition for HULIS in this study operational and couldn't some compounds resolved by certain GC-MS or LC-MS methods elute as HULIS in the current study? Please explain.

P 7196, lines 23-28: Do these values properly correspond to dates biomass burning activity in the PRD region?

P7197, lines 21-23: Please clarify this statement bearing in mind that EC is not specific to any individual combustion source and that EC emissions can vary substantially with combustion conditions.

P7197 and 7198, lines 24-end and lines 1-2: Again, it is not possible to ascribe pyrogenic EC to a single source, and the similar concentrations observed for the continental, marine, and transient atmospheric aerosols is more likely due to a regional effect as opposed to assuming "local" emissions sources (i.e., how is local really being defined here?). It's hard to fathom a marine aerosol that contains 20% EC without some contribution from ship emissions or other pollution source. Clearly, this is a polluted marine aerosol not a pristine one as the term "marine" otherwise suggests.

P7198, lines 3-15: How does $r = 0.36$ indicate "significant correlation". Although the NO and EC are positively correlated, I would argue just the opposite. Also, how are the ozone and NO_x species being measured? There is nothing mentioned in the Experimental section.

P7199, lines 3-5: This is a correct statement that should be reflected throughout the entire study.