

**Overview:** He et al. examine the gas- and particle-phase organic compounds in Singapore air from August to November 2006. There is an attempt to ascribe the biomass burning source to a 3-week event in October 2006 when high TSP concentrations are observed. Select organic markers are presented as evidence for the biomass burning. The subject matter is appropriate for the journal, and the experimental design and approach to chemical analysis appear sound. However, this reviewer has some concerns about the data interpretation given. For one, the interpretation could be improved by providing clearer definitions of some of the metrics and terms being used; specifics will follow. Next, hard physical evidence for the Indonesian fire activity supposedly influencing the October air masses in Singapore is quite scant in the paper (it's really not in the paper at all). So, this manuscript would also benefit from the addition of satellite data or a clearer discussion of the type of fire activity that most likely occurred in Indonesia to cause such a signal in Singapore. (The discussion may need to be more mindful of the fact that the geography of "Indonesia" is complex with respect to Singapore's location [as the National territory of Indonesia almost surrounds Singapore]). Such information should also support the thinking regarding the chemical data provided but more on that below. Lastly, the analytical methodology and how it was applied to determine concentrations needs to be better described. For example, the reader will benefit from knowing if the concentration data presented were background- or field blank subtracted, and how these calculations were performed. This information is important to state regardless of whether it appeared somewhere else before. Again, more specific comments are given below:

**Introduction:** The authors have done a commendable job introducing the problem and describing the lack of organic chemical data available for PM in Singapore. However, the main hypothesis of the research seems to be more that biomass burning in Indonesia strongly influences air quality in Singapore at certain times of the year possibly due to Monsoonal flow and out-of-state regional fire activity. It may be a good idea to introduce this hypothesis in the Intro, while also explaining the agricultural or forest burning activity that typically occurs in Indonesia at this time of year. Or, if this is just a one-time, random fire event that was opportunistically captured....this needs to be explained at the outset with potential implications for the study and beyond.

#### **Experimental procedures:**

p 8419, lines 12-13: please use metric units, convert inch to cm. This is an EGU journal.

PUF plugs are notoriously dirty, please describe what was done to ensure that the PUF plugs were adequately cleaned?

p 8420, lines 3-4: please provide the volume of solvent used for the ASE experiments along with the pressure and temperature conditions within the cell.

p 8420, line 10: explain why internal standards are being added at this stage and for a second time. Is the bottom line that the polar compound internal standards were added following the extraction and concentration steps? This needs to be made clear.

As discussed, please describe how the concentrations were calculated, including any background correction. If there was no background correction, how can the authors ensure the reader that contamination was not an issue for these experiments? In other words, please explain the quality control steps taken for reporting concentrations.

#### **Results and Discussion:**

p 8422, line 14: please define what total suspended particles (TSP) are.

p 8423, line 8: please clarify the sentence "The relative distributions of...mainly ranged from..."

p 8423, line 17: Is this "biogenic" input due to, say, the distillation of waxes during fire? In which case, the input is better described as pyrogenic perhaps. Or is it due to plant abrasion or some other natural wax removal process? The suggestion at this point that C<sub>max</sub> shift "could be" ascribed to the transport of biomass burning aerosol from Indonesia is premature without more evidence. Considering all the evidence for biomass burning being presented in this manuscript, the sum of the parts is unquestionably greater than the parts themselves. In other words, the authors may want to seriously consider presenting the results first and separate from the discussion. In the results focus on defining

the terms in use (more on that below) and presenting the observed values. In the discussion, accumulate the all the evidence for biomass burning in the air masses and then compare concentration, CPI, and PAH ratio values to other relevant work. This type of organization will make for a more compelling and coherent story.

p 8424, line 7: in eq format, please define CPI1, CPI2, and CPI3. It is very difficult to follow this discussion without knowing these eqs.

p 8424, line 19: please define WNA using an eq.

p 8424, lines 19-27: please explain why it is necessary to compare CP1 and WNA. What is underlying this comparison? Why wouldn't these two metrics be related? For the most part, they use almost the same values. No? Also, please clarify the last sentence in this paragraph. How do these *R* values indicate that WNA and CPI (which are interrelated) can be used for source assignments?

p 8425, line 14: please remove "...such as biomass combustion, vehicular and industrial emissions...."

p 8425, lines 15-16: please remove "...such as peat and forest fires."

p 8425, lines 21-22: what does the symbol  $\sim$  mean in this context?

p 8425, lines 23-25: Is it possible to be more specific and quantitative about the conifer burning in Indonesia? In other words, are the tropical pine forests in Indonesia burning due to large-scale deforestation? How much of this occurs when and why? This is a good time to introduce more about what types of fires are common in Indonesia.

p 8425, line 24: please remove "...of organic compounds..." and changes "distant" to distance. Although, it's hard to see where the evidence of "long-distance transport" is. Back trajectories themselves don't help without more on the fires.

p 8426, lines 1-30: First, it is important that the authors mention more about what Yunker was trying to accomplish in his 2002 article. He was attempting to use PAH and their ratios to separate natural petroleum sources (e.g., due to seepage) from combustion and environmental combustion sources in water and submerged soil samples. With that said, there is no reason for this article to present the PAH ratios for liquid petroleum in water and soil. It is irrelevant to the discussion at hand because PAH from these sources are unlikely to become airborne. So, all references to PAH in liquid petroleum should be removed for clarity. It's fine to mention this tact in the article. On lines 9-10 and on line 13, please be specific about which ratios are being referred to. Finally, the last sentence 23-30 is quite confusing because it is too long, and its conclusion about biomass and "peat" combustion stands unsupported.

p 8427, lines 8-10: It is good that the authors approached their work this way. However, Yunker 2002 compiles many different studies from different research groups. Thus, the question that really needs to be answered is: Is the combined phase method used here relevant to how Yunker 2002 treats the PAH measurements from combustion (since this is what the authors compare to)? Unless Yunker 2002 combines gas- and particle-phase PAH measurements to determine the ratios then the two analyses are potentially incompatible. Overall, this discussion may come a bit late and is much better suited to a separate discussion section, as mentioned.

p 8427, lines 10-20: This discussion is overly speculative and evidence contradictory to the point has been given. For example, see<sup>1-3</sup>, both either showed that higher molecular weight PAH may preferentially contribute to smaller particles or did not indicate preferential PAH segregation by particle size. Removal of this discussion is recommended.

p 8427, lines 20-30: Please describe whether the gas-, particle-phase, or both are being discussed. Here are for the *n*-alkanes as well.

p 8428, line 13: please clarify "unimodal GC chromatograph".

p 8428, line 24: please explain what is meant by "temporal trends"

p 8428, line 25: what "correlates" with what? Please clarify correlation between what two things?

p 8429, line 3: define mg/kg units. Please explain what type of wood combustion is expected from Indonesia.

p 8429, line 8: The exact emissions from Indonesian peat fires may be unknown, but there are other studies that may assist. For example, see <sup>4</sup> and references therein.

p 8429, lines 9-12: Again, this comes across as selective speculation. Please remove.

Figure 2: Consider adding reference lines for the PSI index. Please add the TSP axis on the right side and label.

Figure 3: x-axis is illegible, change the y-axis to represent order of magnitude differences (maxes of 10 and 1000 or something similar). As it is, they are difficult to compare.

- (1) Hays, M. D.; Smith, N. D.; Kinsey, J.; Dong, Y.; Kariher, P. H. *J. Aerosol Sci.* 2003, 34, 1061.
- (2) Venkataraman, C.; Friedlander, S., K. *Environ. Sci. Technol.* 1994, 28, 563.
- (3) Venkataraman, C.; Negi, G.; Sardar, S. B.; Rastogi, R. *J. Aerosol Sci.* 2002, 33, 503.
- (4) Sheesley, R. J.; Schauer, J. J.; Chowdhury, Z.; Cass, G. R.; Simoneit, B. R. T. *Journal of Geophysical Research-Atmospheres* 2003, 108, 4285.