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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical, physical, and optical evolution of biomass burning aerosols: a case study" *by* G. Adler et al.

Anonymous Referee #2

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The topic of this paper is of interest, but a number of issues should be clarified. Significant assumptions are made interpreting ambient data to investigate the chemical evolution of biomass burning smoke. Also, over half of the paper involves detailed analysis of AMS spectra that mimics then compares results to other AMS studies. Although this may be of interest to the AMS community, this approach does little toward advancing new insights on smoke evolution and, in my opinion, makes the paper of less interest than it could be.

The analysis of smoke from a unique holiday that involves widespread burning is an appealing idea. However, because measurements and interpretations are being based

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on data from one fixed sampling site, whereas the fires are widely distributed and possibly involve burning a range of fuels under varying conditions (small smoldering vs hot flaming, etc), it is not clear that meaningful interpretations can be made on the smoke aerosol chemical and physical evolution using this data set without first providing evidence that the approach is sound. Presumably, the analysis is based on the assumption that the smoke from many fires is well mixed throughout the night and persists throughout the following day so that the daytime evolution represents a regional average of all the emissions? This may be reasonable, but is there evidence to support this, for example, similar mass loadings throughout the region from network-type monitors, satellite data (fire maps or smoke), etc. At the very least, it seems all data should be presented as ratios.

Assuming that the smoke is regional and well mixed, the premise that the authors were able to record the evolution of the smoke plume from nighttime emissions to later daytime measurements, and that temporal changes later in the day were all due to smoke chemical and physical evolution should be verified. Can the nighttime emissions be more directly linked to the daytime measurements by a back-trajectory analysis, or if the assumption is that the region is uniformly impacted by the smoke, can this be justified by showing that species emitted exclusively by the fires, and largely unreactive on these time scales, remain invariant throughout the time period of interest. It is not clear that the argument made by the authors, that the PAHs and the AMS mass spectra measured in both periods (night vs day) are sufficiently unique to identify that the emissions measured at night are from the same source as those measured next day. It is curious that the idea of a uniformly impacted region does not appear to be consistent with the decrease in smoke in the morning and than increase again around noon. General consistency with expectations (ie, smoke becomes more oxidized, etc) is not sufficient to justify the analysis approach. Could it be shown that ratios of primary smoke components are constant throughout the period (eg, ratios of PAHs ? etc).

Specific Comments

ACPD

10, C11593–C11596, 2010

Pg 24373 line 1: nitrate and ammonium are generally more significant products of biomass burning than sulfate. Not clear why sulfate is mentioned over other more prevalent species.

Pg. 24373 line 17, “quantitative data”. What comparison substantiates this statement? In the past, the AMS was not considered quantitative as a stand alone instrument. For example, consider the assumptions made on page 24377. Give an estimated measurement uncertainty and what it is based on?

Pg 24374, The first paragraph is very specific and is likely of little value to researchers not familiar with AMS data analysis. Can it be clarified?

Pg 24376. Considerable detail is given to the sampling efficiency of the inlet/tubing as a function of particle size, and the size ranges sampled by various instruments, except for the AMS. Details are needed.

How are the organic mass errors, stated in Fig 1 caption, determined?

It seems somewhat questionable that time period B is really associated with the fires (discussed above)? Why did the concentrations from the fires drop (eg, a minimum at 9:00), then increase? Is there any CO or other tracer data available? This would suggest that there is the potential for mixing with other air not impacted by smoke during the sampling period. Or is the assumption that the whole region is uniform and the changes in concentration are just due to changes in fire emissions.

Pg 24380, what is the uncertainty relating to the PAH masses stated. Comparisons are made to other AMS data, but this provides no insight on actual masses. If statements are to be made on the health effects of these species, the accuracy of the masses stated must be known, otherwise it is speculation that these masses are of any consequence.

Pg 24382, (discussion relating to figure 6). One might wish to comment why the intercepts are vastly different and positive (m/z 44 is greater than zero when organic mass

ACPD

10, C11593–C11596, 2010

is zero)?

Pg 24383 Line 8. State here again what the significance of m/s 57 is.

Pg 24384: temporal changes in concentrations during period B are taken as chemical evolution of the aerosol, however, this assumes there are no other processes that could influence the temporal trend, such as transport (even if wind speed is low), change in BL height, all of which can change concentrations by mixing in air masses of differing chemical characteristics (discussed above). It is never shown that the aerosol is regionally uniform. Comparisons involving ratios, ie relative to some conservative co-emitted species, or OA (as is done in some cases) are likely more reliable. This concern applies to all of section 7.1.

Fig 7. More discussion is needed on why the data of period B falls out of the triangular region thought to represent most atmospheric observations.

Fig 10 The trace for the organic mass is incomplete. No data during event B is shown making the discussion on pg 24385 difficult to follow.

Section 7.1 It is not clear how temporal changes in number concentration relative to OA mass indicates new particle formation. Could it not be related to growth of particles into or out of each instruments measurement size range?

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 24371, 2010.