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Interactive comment on “Biomass burning impact on PM_{2.5} over the southeastern US during 2007: integrating chemically speciated FRM filter measurements, MODIS fire counts and PMF analysis” by X. Zhang et al.

Anonymous Referee #2

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General comments

This manuscript presents chemical speciation measurements of water-soluble organic and inorganic species in fine ambient particles in the southeastern US in combination with satellite-derived fire counts. In addition to showing spatial and temporal patterns in the individual chemical species levels, the authors infer relative and absolute contributions from biomass burning to PM_{2.5} mass concentrations using molecular source tracers and Positive Matrix Factorization (PMF). A clear seasonality was observed in the levoglucosan levels and associated biomass burning activities. Interestingly, the

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correlation between the various biomass burning indicators (i.e., the two tracers, levoglucosan and soluble potassium, and the fire counts) showed seasonal dependence as well, revealing the limitations of soluble potassium as quantitative biomass smoke tracer.

This paper is of high quality, both in terms of its scientific content as well as presentation of the methods and findings. The paper address relevant scientific questions and fits well within the scope of ACP. The scientific approach is sound, using valid assumptions for the most part, and the experimental design includes an extensive array of quality control measures. The results and discussion are presented in a clear and well-structured manner. It was a delight to read this paper, as the subject is interesting and the paper is very well written, and thus I highly recommend publication of the paper in ACP. Below are only some suggestions for minor changes along with some remarks.

Specific comments

The analytical method utilized for the quantification of levoglucosan (HPAEC-PAD) provides high sensitivity and selectivity for various carbohydrates, which is somewhat limited by the choice of the PA1 column, as discussed in the paper. The approach taken to correct levoglucosan concentrations due to co-elution with arabitol (using a conversion factor based on mannitol concentrations) is a good first approximation, but it certainly is associated with some uncertainty. Several recent studies have shown mannitol concentrations to be variable (relative to arabitol) and even smaller in some cases (e.g., Kundu et al., 2010). However, most previous studies have found arabitol (and mannitol) to be enriched in the coarse PM fraction. Thus, the uncertainties associated with the method used here (for fine PM) are likely not substantial, yet need to be noted, especially for measurements during warmer seasons and in wet environments (which enhances fungal activity and thus the release of these fungal tracers). The authors actually show the suitability of this method by the good agreement between measurements of levoglucosan using two independent methods (HPAEC-PAD and GC-MS). Nevertheless, an independent analytical method should also be used (in

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a future study) to determine an average arabitol/mannitol ratio for the specific region under investigation (and as a function of season), as both arabitol and mannitol show large spatial and seasonal differences in their ambient levels. In fact, could the poor correlation between levoglucosan and K⁺ during the summer months (as discussed below) be partially due to a larger uncertainty in the levoglucosan measurements during that time of year? One more comment on this issue: as the authors are probably aware of, an alternative anion exchange column (MA1) provides excellent separation of arabitol and levoglucosan.

The authors discuss the lacking correlation between the two biomass burning tracers (levoglucosan and K⁺), specifically during summer, and give a reasonable explanation for this observation, i.e., the contribution of additional sources to ambient K⁺, even when correcting for sea salt and soil dust. Vegetation and meat cooking have been reported as such additional sources (Lawson and Winchester, 1979, Morales et al., 1996, Schauer et al., 1999), which are both expected to be abundant in the southeastern US.

However, it should also be noted that additional factors may be responsible for the large variation in LG/K⁺ ratios and the poor correlation between the two tracers: the relative emission factors of these species are likely also influenced by the combustion conditions, such as fire regime (flaming versus smoldering) and thus fire temperature and combustion efficiency, which can vary significantly between fires even of the same type of biomass (e.g., due to differences in moisture content). Furthermore, several studies have shown catalytic effects of alkali and alkali-earth metal ions (including K⁺) on the formation of levoglucosan during biomass combustion, i.e., inhibited formation of levoglucosan in the presence of K⁺ salts (e.g., Nowakowski et al., 2008). Thus, a number of factors (which are difficult to identify and quantify) affect the relative concentrations of the two tracers in both source emissions and ambient air.

The authors state that levoglucosan likely provides reasonable estimates of biomass burning contributions to PM_{2.5} during winter. Considering short transport distances of the smoke aerosol (as the main type of biomass burning during that time of year is

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residential wood combustion) and reduced photochemical activity, as well as high ambient tracer concentrations, this is a fairly safe assumption, which is also shown in the good agreement with estimates from other studies. Nevertheless, the use of ambient levoglucosan concentrations for quantitative assessments of biomass burning source contributions needs to be done with caution, in particular in case the smoke particles encounter water (e.g., fog or clouds). Certainly, more research efforts are needed in this area.

Technical corrections

1. p. 7041, lines 15 and 17: Change "ml" to "mL"
2. p. 7043, line 19: Add "a" before "wavelength"
3. p. 7044, line 16: Add "concentrations" after "NH₄⁺".
4. p. 7045, line 22: Insert "the" after "limit of".

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