Review of "Fast two-dimensional GC-MS with thermal extraction for anhydro-sugars in fine areosols " by Ma et al.

Overview of the manuscript:

This manuscript presents a very interesting and potentially robust method for fast analysis of important molecular marker compounds for biomass combustion emissions. The authors have done an excellent job of describing the method details and present preliminary data that the method serves its purpose. Although method details were given in detail in a previous paper by some of these authors (Ma and Hays, J. Chrom. A, 2008), thus this paper is limited in its method application. There are only three analytes presented here with a maximum of 8 samples analyzed. My primary concern with this manuscript is that the number of samples that have been analyzed by this "fast" method is very low. Thus the statistical evaluation of the method performance is not convincing. Next, an implicit concern related to this work, is the method's potential for application with regard to widespread use.

Detailed comments:

Robustness? Its unclear in the presented tables and figures which samples were used and how many were used in each. Table 1 has the note n=5 and we assume these are the spiked standards. Table 2 it appears there are 4 sample types with triplicate analysis (both TE and SE). Are additional data available for SE to TE comparison? Table 3 has all 8 samples with 3 replicate analyses. I suppose the main difference between the selected compounds for Table 2 and 3 is the availability of SE-GC/MS data. It seems to me that these two data tables should be combined, so that it's more clear which samples of what types of data.

Why is the error for the loblolly pine so much higher than the other 3 sample types? Is there an interfering compound?

In Figure 1, the authors present calibration curves for the three anhydrosugars and report 2 significant figures, however in the text the linear range is reported with 3 significant figures and the calibration is reported to be consistent over 3 orders of magnitude. This statement does not seem consistent with Figure 1.

I understand that an instrument schematic was previously published; it would be really nice to see this figure in this paper as well.

Several years ago, method development for TE was being done by another group. In their approach, they used a silylation step. Have the authors tried this approach? Is data available from the previous work for comparison?

One of the very attractive features of SE-GC/MS methods is that they are widely applicable. The method is fairly easy and uses standard instrumentation, an obvious advantage. In this paper, TE-GC/MS is presented and is apparently very fast without the need to derivatize the polar functional groups. Can this method be applied more widely? Is the instrumentation standard? How many people have the capability to conduct 2-d separations? Can the instrument be operated remotely?

What are the authors implying with their comment about levoglucosan stability for almost 10 years in an ultralow freezer? Have the authors investigated storage of levoglucosan at lower temperatures for the same time periods? Are the samples collected and especially those stored over time ambient samples? We might expect ambient samples to have microbial activity and thus, the ultralow temperatures are necessary.

Related to the previous statement, does the Frasier and Lakshmanan, 2000 paper still have validity? If storage at ultralow temperature are required for levoglucosan stability, doesn't this imply that the ambient levoglucosan is not really stable?

On page 157, the thesis statement for the paper states that the focus of this paper is to present accurate and reproducible quantification of levoglucosan…with proficiency testing and comparison to silylation methods.

On page 165, the range of observed levoglucosan concentrations in ambient samples is given. In figure 4 it is suggested that since the measured values are within the range, which is quite wide 0.004 to 7.6 ug/m3, then the values presented here without consideration to season or location are acceptable.

In this reviewer's opinion, a more direct sample comparison is need to compare TE-GC/MS to SE-GC/MS or HPLC-PAD. While I agree that the four samples presented are promising in this regard, it appears to be contradictory to the title statement that this method is fast and easy because the analysis is of underivatized levoglucosan.

Perhaps more a question of style, but in my mind still worth mentioning, is that there are very many acronyms used in this paper. Quite a few of these acronyms are not used more than a few times, making their occasional use quite distracting. Along these lines, since the paper is about three anhydro-sugars, why not just use their names throughout the manuscript.

Minor comments:

On page 159, what is MSP? Is this a manufacturer or sampling site location?

What is PMT?