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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Airborne characterization of smoke marker ratios from prescribed burning" *by* A. P. Sullivan et al.

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Referee #1

The study is certainly well done and it merits publication, but the manuscript would benefit from an explicit statement or two that clearly describe the wider advantages of these measurements. It can better emphasize the value of these data and the reported relationships. In other words, what are the next steps? How will these data be used to improve which dispersion, air quality, or climate models? What is the benefit of knowing that marker/chemical property ratios are strongly influenced by fuel type? In the final analysis, the study does a superb job of presenting the data and of digging through certain interesting relationships among widely-measured chemical markers, but could

further clarify the intrinsic value of what was demonstrated.

-We have tried to better state the value of the measurements and address the above questions by adding the following text to the summary section (lines 415-423): These results should help to better constrain apportionments and models trying to determine the impact of biomass burning on air quality. For example, it has been shown that source smoke marker ratios for levoglucosan and galactosan collected from controlled laboratory burns can be applied to obtain accurate estimates of the impacts of prescribed burning on fine particle concentrations. This is not the case for mannosan and potassium. Ratios for these species cannot accurately be drawn in all cases from controlled laboratory burns and should be site and burn specific. Knowledge of fuel type specific smoke marker profiles can improve both chemical transport model and receptor model estimates of prescribed burning impacts on fine particle concentrations and haze.

(i)Abstract – Use of "RF" notation is confusing here. It should be defined.

-RF stands for research flight and the text has been updated to reflect this. Lines 52-55 now read as "Each fire location appeared to have a unique ïĄĎlevoglucosan/ïĄĎwater-soluble organic carbon (WSOC) ratio (RF01/RF02/RF03/RF05 = 0.163 $\pm$ 0.007 ïĄ■g C/ïĄ■g C, RF08 = 0.115 $\pm$ 0.011 ïĄ■g C/ïĄ■g C, RF09A = 0.072 $\pm$ 0.028 ïĄ■g C/ïĄ■g C, RF09B = 0.042 $\pm$ 0.008 ïĄ■g C/ïĄ■g C where RF means research flight)."

(ii)P11717, lines 15-20: This is a vaild point about GC-MS. Although, levoglucosan is normally so abundant in biomass burning plumes, the sampling time requirements are strongly reduced. For example, the TAG GC-MS is capable of measuring many of these markers in 30 min or less in the atmosphere without the benefit of being in a biomass burning plume. It was nice to see all the WSOC and LG measurements match up so well, but from the perspective or air quality (for example) the real benefit of measuring LG so quickly (the off-line analysis time is reported as 59 min) is not perfectly clear.

-As mentioned above, to try to better indicate the benefit of the measurements from an air quality perspective we have added the following text to the summary section (lines 415-423): These results should help to better constrain apportionments and models trying to determine the impact of biomass burning on air quality. For example, it has been shown that source smoke marker ratios for levoglucosan and galactosan collected from controlled laboratory burns can be applied to obtain accurate estimates of the impacts of prescribed burning on fine particle concentrations. This is not the case for mannosan and potassium. Ratios for these species cannot accurately be drawn in all cases from controlled laboratory burns and should be site and burn specific. Knowledge of fuel type specific smoke marker profiles can improve both chemical transport model and receptor model estimates of prescribed burning impacts on fine particle concentrations and haze. In addition, we added some text about the benefit of quick sampling to the summary section. Lines 395-399 now read as "Comparisons with other measurements aboard the Twin Otter show that the 2 min time resolution was adequate to characterize smoke markers in the smoke plumes. The ability to collect quick samples with the PILS followed by later off-line analysis provided advantages where rapid time resolution (minutes) is beneficial (i.e., plume sampling and/or aircraft measurements)."

(iii)Section 2.3 should come first so that the order of activity is clear.

-The first part of section 2.3 has been moved to the beginning of section 2 so that the methods section begins with a discussion about the airborne mission.

(iv)In Fig. 2 the legend symbol for LG does not match what's in the figure.

-The reason this appears to be the case is that 1 s data is being plotted. So the levoglucosan, which is a two minute time-integrated measurement, appears as a bar rather than individual points. To try and fix this, the symbol for levoglucosan has been changed from a cross to a square.

(v)Fig. 3a is for one flight. What was the correlation coefficient for all flights for LG/WSOC?

-The R2 correlation coefficient for all flights for levoglucosan/WSOC is 0.93. The text has been updated to include this and lines 244-246 now read as "In addition, the ratio between levoglucosan and WSOC appears to be fairly constant (R2 = 0.93 for all data), which will be discussed in more detail in the next section."

(vi)P11723, lines 1-2: "...with a peak in the CO concentrations are considered." Is not clear.

-This sentence has been reworded and lines 255-256 now read as "Only fraction collector samples that directly overlapped with a CO plume penetration are considered."

(vii)P11725, line 27: Why is a ratio being used to check this? Doesn't that just complicate the situation? How does one know the changes and rates of reaction for WSOC and LG?

-A ratio is being used to check the role of aging on smoke markers since it is a source smoke marker ratio that is used to apportion the contribution of biomass burning. Therefore, it is important to understand how this ratio changes with time. The rate of reaction for WSOC and levoglucosan are unknown and the reaction rate could play a role in aging. The text has been updated to indicate this and lines 331-336 now read as "Changes in plume composition occur with plume aging, due both to plume dilution (which can influence gas-to-particle partitioning) and photochemical reactions, but very little data quantitatively examines the impact (if any) of these processes on smoke marker ratios. Since a smoke marker ratio is needed to apportion the contribution of biomass burning this is important to investigate. But it is also important to note this impact would depend on the rates of reaction of levoglucosan and WSOC, which are unknown and could be similar."

(viii)P11726, lines 1-5: The ΔLG/ΔWSOC ratios can vary by nearly a factor of 2. What is the criteria for this ratio being "stable". Is this measurement error?

-It is true that when considering all the data, the Δlevoglucosan/ΔWSOC ratio ranges across a factor of 2. However, within a particular fuel type or fire location this ratio is constant or stable with a standard deviation generally less than ± 0.04. To better indicate this lines 338-340 now read as "Over the range of smoke plume ages (up to approximately 1.5 h), the observations give no clear indication that the ratio changes across a fuel type or fire location in a consistent manner as the plume ages."

(ix)P11726, lines 18-20: Again, not sure that I agree Δm/z60/ΔOA is a model of stability as implied here. What happened with RF08?

-We agree that our original statement about Δm/z60/ΔOA was a bit strong and these lines have been removed.

(x)Table 2 should be added to the Supporting Information

-Table 2 has been moved and now appears as Table S1 in the Supporting Information.

Referee #2

Methods It would be useful to have additional information on the types of fuels burned (type of vegetation, amount of acreage combusted, etc.).

-The size of the fires sampled is now included in Table 1. The only fires that we have fuel information for are the burns that occurred at Fort Jackson since the other burns sampled were just fires of opportunity and we had no ground-based measurements. This is indicated in lines 152-155 as "The first flights (RF01/RF02/RF03/RF05 where RF means research flight) focused on prescribed burning at Fort Jackson, SC. Once these burns were completed, airborne sampling only of other prescribed burns taking place in SC began (RF08 and RF09)." and lines 288-290 as "Ground-based sampling of the Fort Jackson burns included fuel characterization [Yokelson et al., 2013], which indicated the interior environment was a longleaf pine/wiregrass system."

Also, how representative is this controlled burn compared to a "normal" forest fire?

-Prescribed burns are usually less intense that a "normal" forest fire. The text has

been updated to reflect this and lines 143-148 now read as "The research flights conducted were part of a combined ground-based and airborne-based study to examine the emissions from prescribed burning in the southeastern U.S. In the southeastern U.S., prescribed burning is often implemented every one to four years in wildlands to maintain or restore fire-adapted ecosystems. Burns are conducted so fuel consumption will only be in the understory and the forecast transport is such that smoke impacts will be minimal. Therefore, in general, prescribed burns are less intense than wildfires."

Referee #3

1.The manuscript would benefit from including more information regarding the prescribed burns samples from the airborne platform. For example, the authors say on p.11723 (lines 20-25) that the Δlevoglucosan/ΔWSOC ratio observed for flight RF09 suggests that "...the vegetation may have been different at the two ends of the fire being sampled during RF09. In South Carolina it is very common for marshy bays to be mixed in with a forested areas." This hypothesis could be easily verified, if more information regarding the fuel burned during this prescribed burn is presented. Also, the statement on p. 11724: "Similarities in smoke marker ratio values suggest that the Fort Jackson burns (RF01/RF02/RF03/RF05) were dominated by the burning of grasses, RF08 by leaves, RF09A by needles, and RF09B by marsh grasses" could be easily verified.

-The only fires that we have fuel information for are the burns that occurred at Fort Jackson since the other burns sampled were just fires of opportunity and we had no ground-based measurements. This is indicated in lines 152-155 as "The first flights (RF01/RF02/RF03/RF05 where RF means research flight) focused on prescribed burning at Fort Jackson, SC. Once these burns were completed, airborne sampling only of other prescribed burns taking place in SC began (RF08 and RF09)." and lines 288-290 as "Ground-based sampling of the Fort Jackson burns included fuel characterization [Yokelson et al., 2013], which indicated the interior environment was a longleaf pine/wiregrass system." However, we do have an independent analysis that calculated emission ratios for the fire sampled during flight RF09. They also found two groups of emission ratios for this fire further suggesting that the vegetation at the two ends of that fire were different. Text indicating this has been added and lines 275-281 now read as "The ïĄĎlevoglucosan/ïĄĎWSOC ratio was 0.042 ± 0.008 ïĄ■g C/ïĄ■g C for RF09B, which was lower than the ratio of 0.072 ± 0.028 ïĄ■g C/ïĄ■g C for RF09A, suggesting that the vegetation may have been different at the two ends of the fire being sampled during RF09. In South Carolina it is very common for marshy bays to be mixed in with a forested area (B. Manks, personal communication, 2011). In addition, an independent analysis to calculate the emission ratios for these same fires found two groups of emission ratios for the fire sampled during RF09 [McMeeking et al., 2014]." Information on the size of the fires sampled is now included in Table 1.

2.The authors say in the Introduction (p. 11717) that "...traditional methods used to measure smoke markers, such as gas-chromatography-mass spectrometry (GC-MS), require a large amount of mass for analysis." "Large" is a relative term – do authors mean micrograms, mg, grams? Modern GC/MS instruments are very sensitive and usually require no more than a few tens of micrograms for polar species analysis.

-We were trying to indicate that a large amount of aerosol mass is needed since generally a high concentration of a particular organic species is needed for analysis. In order to better indicate this, lines 102-108 now read as "One of the main reasons for this approach is that traditional methods used to measure smoke markers, such as gas chromatography-mass spectrometry (GC-MS), generally require a high concentration of a particular organic species for analysis. In order to reach this concentration a large amount of aerosol mass must be collected. Therefore, ground-based sampling would provide the best means to collect a large amount of aerosol mass as this is generally not feasible from an aircraft platform that can quickly fly through a plume."

3.The sentence on the bottom of p. 1171, starting with "Other measurements presented here..." is not clear, something is missing here.

-The sentence has been reworded and lines 223-230 now read as "Other measurements presented here include 3-D location and windspeed collected with a wing-mounted Aircraft Integrated Meteorological Measuring System probe (AIMMS-20, Aventech Research, Inc.) to estimate time since emission values, six second time-integrated organic aerosol (OA) concentrations determined by a High Resolution - Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) [DeCarlo et al., 2006], one Hz carbon monoxide (CO) determined by a Picarro cavity ring-down spectrometer, and AFTIR (Airborne Fourier transform infrared spectrometer) data analysis products including modified combustion efficiency (MCE) ratios [Yokelson et al., 1999; Burling et al., 2011; Akagi et al., 2013]."

4.Figure 2, the legend symbol for levoglucosan is not consistent with the graph.

-The reason this appears to be the case is that 1 s data is being plotted. So the levoglucosan, which is a two minute time-integrated measurement, appears as a bar rather than individual points. To try and fix this, the symbol for levoglucosan has been changed from a cross to a square.

5.Fig. 4a is too busy and difficult to read. Could different colors be used for different fire locations? The same comment for Fig. 5.

-Figures 4 through 9 have all been updated to have the various fire locations indicated by different colors.

6.Page 11726, Lines 5-12: the authors discuss a tight correlation between Δm/Δz 60 vs. ΔOA concentrations shown on Fig. 7. However, this correlation is mostly driven by flights RF03/RF05, which was dominated by the same type of fuel (grasses).

-Although RF03/RF05 did sample the highest concentrations, the correlation shown in Figure 7 is not driven by this data. To help indicate this, the text has been updated to include the R2 values for each fire location as well as all the data. Lines 348-351 now read as "Despite the differences in fuel type and ïĄĎlevoglucosan/ïĄĎWSOC ratios across burns described above, this AMS ratio shows a very tight relationship ($R2$ = 0.99 for RF03/RF05, $R2$ = 0.98 for RF08, $R2$ = 0.99 for RF09A, and $R2$ = 0.99 for all data)."

7.Fig. 6 and discussion on p. 11726. There is a large spread in the Δlevoglucosan/ΔWSOC initial ratios for flights RF03, 08, and 09A. Not clear why the authors concluded that this ratio "is stable". Similarly, it is not apparent from Fig 6b that the Δm/z 60/ΔOA concentration ratios are consistent "...across burns and fuel types..." and that this "...reinforces the quality of m/z 60 as a quantitative biomass burning source marker for use with AMS data sets."

-It is true that when considering all the data, the levoglucosan/WSOC ratio covers a large range. However, within a particular fuel type or fire location this ratio is constant or stable with a standard deviation generally less than $\pm$ 0.04. To better indicate this lines 338-340 now read as "Over the range of smoke plume ages (up to approximately 1.5 h), the observations give no clear indication that the ratio changes across a fuel type or fire location in a consistent manner as the plume ages." In addition, we agree that our original statement about the quality of m/z 60 as a quantitative biomass burning source marker was a bit strong and those lines have been removed.

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