Interactive comment on "ACE-FTS measurements of trace species in the characterization of biomass burning plumes" by K. A. Tereszchuk et al.

Anonymous Referee #2

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The authors present ACE-FTS measurements of a large number of chemical species in biomass burning plumes in the free troposphere. They are able to identify with reasonable accuracy the age and geographical origin of the biomass burning plumes and go on to present tracer-tracer correlations and normalized excess mixing ratios. This paper represents a good first step towards obtaining more detailed knowledge about biomass burning emissions in the free troposphere from space-based measurements in consideration of the age of the plume and type of material burnt, which may help in constraining atmospheric models. The following specific issues should be addressed:

- Section 2.2, p. 16616: Some more detail is needed about the processing of the ACE-FTS data. Which type of retrieval is used? Are retrievals performed using optimal estimation, for example, in which case prior constraints would need to be carefully considered? Also, the reasons behind filtering the data in the way described in the paper are not clear. What is it that means low values less than 10% of the median value should be rejected? What is the reason behind using the criterion for rejecting data with a measurement error (do you mean retrieval error here?) of greater than 100 times the mean absolute deviation of the dataset? Do these represent retrievals that have failed to converge? How do you screen for cloud contamination? It would be useful to include a brief error budget for the retrieved species to provide an indication of the accuracy of the measurements used in the subsequent analysis.

In this work, Version 3.0 of the ACE-FTS data was employed, specifically the dataset where the retrievals have been interpolated onto a 1-km altitude grid (http://www.ace.uwaterloo.ca). Specific details of the retrieval method, error budget and interpolation are outlined in the following publication which will be added to the references.

Boone, C.D., Nassar, R., Walker, K.A., Rochon, Y., McLeod, S.D., Rinsland, C.P., and Bernath, P.F.: Retrievals for the atmospheric chemistry experiment Fourier-transform spectrometer. Applied Optics, 44, 7218-7231, 2005

The data filtering was conducted to eliminate any potentially erroneous measurements from the ACE-FTS dataset. Typically, cloud contamination is the biggest factor. ACE-FTS measurements of the free troposphere which contain cloud in the FOV of the instrument causes the retrieval to fail at the altitudes containing clouds and the outcome of the retrieval is a skewing of the vertical profile to unrealistically low and even negative values for the VMR of a given molecular species. The median filtering seeks to eliminate all occultations which contain said skewing to assure that the measurements being used are only those where the troposphere is cloudless during the time of the measurements. So occultations that possess molecular VMRs at a given altitude that are less than 10% of the median value at that same altitude are rejected. The measurement error referred to is indeed the retrieval error from the ACE-FTS measurement. Occultations with unrealistically large retrieval errors were also filtered out as they often do indicate failed convergence. I will add further details to the manuscript.

- Section 2.3 p16616: In figure 1, the back trajectory appears about 200 km away from the nearest fires, and so in this respect this figure is not entirely convincing. Presumably the tangent point of the occultation is used as the starting point for the back trajectory calculations and so perhaps the authors could acknowledge that the horizontal resolution of ACE-FTS is poor, in common with all limb sounding instruments, on the order of hundreds of km, and that this could introduce substantial errors into the back trajectory calculations depending on how the plume is sampled. A visual check of the geolocation of the ACE occultation against IASI total column CO measurements could be helpful to check the sampling of the plumes. It is stated in the paper that the IASI CO measurements are used to 'ensure that measurements are from a singular origin and not a mixture of biomass burning outflows from different locations'. However, it's not clear to me how, by inspecting a single scene, as is implied by the inclusion of Fig 2, it can be ascertained for certain that the plume is not a mixture of biomass burning emissions from different locations. Did you use further back trajectory calculations and the corresponding IASI data from previous days to check for the convergence of air from different sources?

The HYSPLIT trajectory model is exactly that, a model. It is used to help give a general indication of the direction of the movement of air masses at a given time to assist in determining the source of biomass burning emissions. HYPLIT backtrajectories have been used with great success in the BORTAS-A and BORTAS-B campaigns (http://www.geos.ed.ac.uk/research/eochem/bortas/) to confirm the locations of Boreal fire emissions and was used to corroborate IASI CO near real time data and and GEOS-5 CO model forecasts that were employed during the campaign for flight planning purposes. In this work, IASI CO data is used as the principle indicator of the sources of biomass burning and to confirm the results of the HYSPLIT backtrajectories. Of course, a single "scene" is not used to determine if the measurements of biomass burning outflows are from a singular source. Animations are create using the morning and evening passes of IASI to observe plume movements over the period of time leading up to the time of the ACE measurement to determine where the plume outflows are originating from and if the the is convergence from different sources. Since I cannot put an animation in the paper and can only potentially include it as supplementary material which can be accessed online, I am forced to use a snapshot of the IASI data in figure 2 to simply confirm that the HYSPLIT backtrajectory in figure 1 demonstrates that the plume source is indeed from the Boreal fires indicated from the AATSR data. I will expand upon this in the manuscript.

- Section 3: It is necessary to inform the reader of how many occultations were used. Also the units of the Std Dev in terms of the mixing ratio are not easily interpreted. The standard deviations need to be normalised according to the mean mixing ratio of each species to aid comparison of the quality of the fit.

It would appear from anonymous referee #1 that not only are the number of occultations necessary, but the total number "n" of data points from all the occultations used in the correlation calculations should be reported. This will be included in the final manuscript. The std. dev. will be normalized accordingly.

- Section 3.1 p. 16620: It would be helpful to extend the discussion of the chemistry of the plumes.

I will extend upon the discussion of the chemistry of the plumes, but remember that the goal of the work is to present preliminary finding on a more qualitative level to determine the potential and possibilities of using ACE in the characterization of biomass burning emissions. Now that the BORTAS flight campaign (July 2011, http://www.geos.ed.ac.uk/research/eochem/bortas/) is concluded, we can now validate the ACE data with aircraft and ground measurements of Boreal biomass burning and incorporate these finding into chemical models. From there, I will be able to elaborate more in the actual chemistry that is occurring in these plumes.   Furthermore, as mentioned in the manuscript, additional VOC and OVOC molecules are to be retrieved by ACE to assist in having a more complete understanding of plume chemistry and aging over time.

- There is no mention of whether the occultations used are for sunrise or sunset or a mixture of these. On p. 16621, I am concerned that this could be a hidden factor for NO2 , which is photolyzed on a short time scale in the mid-upper troposphere. Was this taken into account for the comparisons of the correlations for NO2 between Boreal and savannah fires and Amazon fires?

This is an excellent point. The occultations used were a mixture of sunrise and sunset measurements. I will make a point of elaborating more on this in the manuscript. In future publications, once I have gone through the entire ACE-FTS dataset and have identified all measurements with instances of biomass burning, I will endeavor to investigate the potential differences in the sunrise and sunset measurements to differentiate day- and nighttime chemistry within the plumes.

-Section 3.2 p. 16624: The authors state that their values of the emission factors for HCN and OCS are an order of magnitude smaller than those reported by Akagi et al. (2010), but they only hint at an explanation. Could they be more specific about what could cause this discrepancy?

At the moment, I cannot account for this discrepancy other than that it should be kept in mind that the emission factors calculated by Akagi et al. are for fresh, young plumes measured at the instance that the biomass material is being combusted, while the ACE measurements are measurements of young plumes that have aged in the troposphere from 1-3 days. Comparison with other satellite data and the aircraft and ground measurements made during the BORTAS-B campaign with confirm if it is systematic error associated with the ACE-FTS retrieval or due to age dependence from the time of measurement.