Atmos. Chem. Phys. Discuss., 10, C8410–C8414, 2010
www.atmos-chem-phys-discuss.net/10/C8410/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nitrogen oxides and PAN in plumes from boreal fires during ARCTAS-B and their impact on ozone: an integrated analysis of aircraft and satellite observations" *by* M. J. Alvarado et al.

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Received and published: 7 October 2010

Here we reply to the comments of Anonymous Referee 2 (C6825). The referee's comments are in bold followed by our responses. We thank both of the reviewers for their time and helpful comments.

Anonymous Referee 2 Received and published: 25 August 2010 General comments: This paper attempts to improve understanding of chemistry within fresh, aged and old biomass burning plumes using aircraft and satellite observations

made during the ARCTAS-B campaign. It provides the first observational confirmation of rapid PAN formation in a boreal smoke plume but there was little evidence for ozone formation within the boreal smoke plumes measured during the campaign period. I find the paper very clearly written and should be accepted by ACP once the authors address the specific points listed below.

Specific comments: Page 15330, line 17: How did the authors arrive at the assumptions made to identify the plumes? The choice of 20 ppb seems rather arbitrary to me. Is this a standard technique to identify boreal plumes? Also, why was r2>0.3 chosen as the filter for a significant correlation?

Our approach follows that of Mauzerall, et al, 1998, as stated on page 15331 of the original paper. There is some subjectivity in deciding what is a plume and what is not. Our choice of quantitative criteria (20 ppb CO enhancement and correlations of CO with HCN and/or CH3CN with r2>0.3 as discussed on page 15330) was an attempt to reduce that subjectivity. Our choice of 20 ppb was made to ensure that the changes in CO were large enough to be due to a plume rather than changes due to the changing altitude of the aircraft, etc. The r2 cutoff of 0.3 for CO and HCN/CH3CN correlations was similarly chosen to ensure that the enhanced CO was likely from a biomass burning source and to eliminate plumes dominated by anthropogenic emissions. Reducing the r2 cutoff to 0.2 would only add one plume to the study, while increasing it to 0.6 would only eliminate one aged plume, so changes to these somewhat arbitrary values would have little to no impact on our results.

Page 15332, line 1: In section 2.2, the authors discuss the use of TES observations to look at ozone and CO enhancement in boreal smoke plumes. Median vertical profiles from DC-8 aircraft data are shown in Figure 5. Could the authors please comment on the thickness of individual plume "layers"'. To what extent does the vertical thickness vary? This is a very important observation and has a direct bearing in section 3.3.3 which uses the TES satellite instrument to detect CO and ozone in plumes.

About 27 of our 34 identified biomass burning plumes observed by the NASA DC8 were sampled during ascents and descents, allowing us to get a lower-bound estimate of the plume thickness. The median vertical thickness was 1.5 km. Eight plumes were less than 1 km thick in the vertical, with 2 plumes less than 0.5 km thick (one fresh and one aged). Seven plumes were greater than 3 km thick, using our criteria of a CO concentration greater than 120 ppbv to define the plume. Occasionally within the thick plumes, the plume would consist of concentrated and dilute layers of biomass burning smoke, with the thickness of each layer being as thin as 100 m.

In identifying plume using TES, we only counted TES retrievals that showed CO concentrations of 150 ppb or greater at 510 hPa, which is much larger than the a priori value at this altitude (about 110 ppb). While this procedure will detect thick plumes in the free troposphere that are being transported between continents, it is true that this will not detect plumes that remain near the surface or are relatively thin or highly diluted. We have added a paragraph to Section 2.2 (lines 139-147) of the revised paper to emphasize this limitation.

Page 15334, line 1: The authors refer to the Val Martin et al., 2010, paper which derived that only 4-12% of North American fire plumes are injected above the boundary layer. It is my understanding that this study excluded pyro-cumulus clouds intermingled with smoke, so was possibly biased low. The authors comment on page 15337, line 14, that they find a value higher than this from the DC-8 measurements. One point is included (labelled "A") which was one such cloud. Were the authors sure that none of the fires labelled in section B and C were associated with pyro-cumulus cloud, leading to a higher bias?

We examined the data for cloud index provided in the ARCTAS data archive by Bruce Anderson for the plumes encountered by the DC8. Twelve of the 34 plumes (8 fresh, 3 aged, 1 old) showed evidence of liquid water or ice cloud mixed with the plume. One is the pyroconvection event labeled A in the figure. Of the remaining seven fresh plumes mixed with liquid water or ice, 2 are below the local BL while 5 are above. Of the five

above the BL, 3 are in group C at 4.0, 8.1 and 5.7 km while 2 are in group B at 3.2 and 10.3 km. Removing these plumes leaves 6 of 8 plumes in group B above the BL, reducing the discrepancy with Val Martin et al., 2010. We have added this discussion to our revised paper at lines 269-275, and thank the reviewer for his comment.

Page 15338, line 14: In the section on enhancement ratios the authors contrast their findings with aged plumes over the Azores. Could the authors please comment as to how significant the vegetation type is (i.e. the type of boreal biomass burned) to the difference in results?

We do not have sufficient information to address this question. ARCTAS sampled boreal fire plumes of various ages, and very aged boreal plumes were sampled over the Azores (as determined by back-trajectories). As we mentioned above in response to a similar question from Referee 1, we used back trajectories to determine the approximate origin of each intercepted plume, as discussed in page 15330, lines 21-23. However, we have not attempted to identify an exact origin for each smoke plume, or to identify the vegetation and ecosystem types present. Even if we did, we would not know how much of the fuel burned was duff, or the various above ground fuels. This can of course affect real-world emissions of NOx as biomass with a higher nitrogen content emits more NOx, all other things being equal.

Page 15339, line 1: As the error bars overlap, these two measurements are the same in my opinion.

We agree – we included this discussion to emphasize that point to casual readers who may only look at the mean values and be confused as to why the sums do not match.

Page 15345, line 25: In section 3.3.3 the authors compare GEOS-chem results with TES special observations. Given that TES is a nadir viewing satellite instrument, how sensitive is TES to tropospheric ozone and CO? It is mentioned that the TES averaging kernels are applied to the GEOS-CHEM data, which is a generally accepted approach when comparing atmospheric measurements to a

model, but no averaging kernels are shown in the paper. Can the authors please include a "representative" averaging kernel which provides the reader with more information on the vertical sensitivity of the measurement and comment on the tropospheric sensitivity of TES to CO and ozone. Can they be sure that the retrievals are not just reproducing the a priori information?

As we mentioned in response to Referee 1, we have added a new Figure 1 to our revised paper showing a typical TES averaging kernel for this period. The retrievals shown in the paper generally have 1 degree of freedom of signal (DOFS) below 250 hPa for both ozone and CO, suggesting that the retrievals include at least one piece of information on tropospheric ozone and CO concentrations. In identifying plume using TES, we only counted TES retrievals that showed CO concentrations of 150 ppb or greater at 510 hPa, which is much larger than the a priori value at this altitude (about 110 ppb).

Technical corrections: Figures 10, 11, 12, 13, 14, 15: what does the solid blue line represent?

The solid blue vertical lines represent the approximate horizontal extent of the plume. We have added text to the captions to make this clearer.

Figures 5, 6, 7, 8, 10, 12, 13: could the authors please replace "O3" with "O_3".

We have adjusted the figures in the revised paper.

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