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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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Here we reply to the comments of Anonymous Referee 1 (C6617). 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 1 Received and published: 19 August 2010**

**The paper interprets aircraft and satellite observations of trace gases during the ARCTAS-B campaign to improve understanding of chemistry in fresh, aged and**



**old biomass burning plumes, with a particular focus on nitrogen oxides and PAN. It's a well written paper and should be accepted by ACP. Below, I list some minor comments and questions, which I would like the authors to address.**

**The authors use the ratio of CO to a range of hydrocarbons to determine the age of the plume. This is an approach that has been used in many previous studies but given that the plume aerosol might slow down the photochemistry, I wonder how robust this approach is. Did the DC8 measure aerosol optical depth? Have the authors looked at the model and observed OH?**

Using the correlations of hydrocarbons with CO to represent plume age is of course an imprecise measure, which is why we only use it to classify plumes into the classes fresh, aged, and old, rather than to provide a quantitative estimate of plume age. The range of OH used in the calculation of species lifetimes given in page 15331 lines 7-11 of our original paper is based on the range of OH concentrations observed by the DC8. While we do not compare this with modeled OH in this paper, Mao et al., 2010 provides an extensive comparison of GEOS-Chem modeled OH and HO2 with observations from the DC8 during ARCTAS-A in the spring of 2008. They found that model OH was within 15% of measured OH between 1-6 km, with an underestimate of 40% below 1 km and an overestimate of 40% above 6 km. We have added a reference to this work in our revised paper at lines 102-104.

**My understanding of biomass burning plumes is that they can be remarkably thin in the vertical (100s m), making them difficult to intercept by plane and challenging to view them from space. Each plume is different, of course, and I'm wondering whether ARCTAS-B had a chance to assess the dimensions of the intercepted plumes. The vertical extent of the plume, in particular, will have implications for its detectability by satellite instruments that typically have sensitivity over relatively large altitude ranges. Can the authors comment on this issue regarding TES. It would be useful if the authors showed a ''typical'' TES averaging kernel for the observed scenes during ARCTAS-B.**

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.

We have added a figure showing a typical TES averaging kernel for this period to our revised paper (Figure 1 in the revised paper). 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 plumes 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). In addition, for many of the plumes observed the tropospheric ozone column retrieved by TES was significantly different from the a priori value within the plume. 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 added a paragraph to Section 2.2 (lines 139-147) of the revised paper to emphasize this limitation.

**The PAN chemistry will be dependent on the speed and height of the pyroconvection. The authors assume, based on Val Martin et al, 2010, that all fires are injected uniformly within the boundary layer. This is a reasonable prior assumption but I'm wondering how sensitive the results to injection 10% of the plumes above the boundary layer. I notice that none of the model calculations shown in this paper even capture the vertical distribution of CO (Figure 5) which is a relatively simple gas.**



One correction here – Val Martin et al., 2010 found that 4-12% of plumes observed by MISR over North America are in fact injected above the boundary layer. We looked at the heights of the fresh plumes encountered by the DC8 versus the 3 hour average boundary layer height from the GEOS5 meteorology fields, and have summarized our results in Section 3.2.1. We found that 4 of the 10 plumes encountered above the burning region were above the boundary layer, larger than the estimate of Val Martin et al., 2010, and attributed the difference to the later sampling time of the aircraft relative to the satellite. In response to the third comment of reviewer 2, we examined the cloud index data of Bruce Anderson in each plume, and found that two of the plumes above the burning region were mixed with liquid water cloud. Since Val Martin et al., 2010 excluded plumes mixed with clouds from their analysis, we find 6 of 8 non-cloud fresh plumes, or 25%, above the BL in Area B, versus 4-12% in their study, and we have adjusted the discussion at lines 269-275 in our revised paper to reflect this.

Injection of emissions in the boundary layer is the default setting for GEOS-Chem, and we didn't adjust it in this study. Leung et al., 2007 found that injection height was indeed significant for determining the PAN and ozone formation from biomass burning emissions, as discussed in our original paper at page 15343, lines 22-26. The disagreement of the model with observations of CO at higher altitudes could be related to a model underestimate of anthropogenic emissions of CO, an underestimate of biomass burning plume height, or an underestimate of emissions from Siberian biomass burning plumes, as discussed in page 15342, lines 27-29 of the original paper. While a full discussion of anthropogenic and biomass burning sources of CO is beyond the scope of this paper, which focuses on NOy species and ozone, Fisher et al., 2010 performed an inversion study of model CO emissions using DC8 data from the spring ARCTAS–A campaign, and some of their results are discussed on page 15334 of our original paper. We have expanded this discussion in our revised paper at lines 188-191 to include the anthropogenic results, and discuss their implications for our work at lines 409-412.

**I question the approach taken to reduce emissions by adjusting the emission fac-**

**tors. Our understanding of the interplay between chemistry and plume dynamics is incomplete so comparing model and observed ratios between NOy to CO is not necessarily a robust approach for determining emission factors. What about assumptions made about combustion completeness or the burned area? I am not suggesting that the emission factors are perfect but I am suggesting there are other variables that could be altered that would lead to the same emission reduction.**

Estimates of emission factors based on observed enhancement ratios within plumes, as in this paper, provide one of the best empirical methods we have to determine such factors, and this method has been used extensively in the literature. It is true that many variables affect the emissions, and there are many potential parameters to change that can give an equivalent amount of emissions for any given trace gas. However, we need to separate two issues here – the estimate of the total amount of biomass burned (kg DM/hour), and the species emission factors (g gas /kg DM) used to convert the rate of biomass consumption to the rate of emission of trace gases in g gas/hour. Estimates of biomass consumption (kg DM burned per unit area), sometimes computed using combustion completeness, and burned area are included in the estimates of total amount of biomass burned in the FLAMBE emission database. Changes to these parameters affect all trace gas species equally. The species-specific emission factors are applied within GEOS-Chem to estimate species-specific emission rates. Changes to a single emission factor only change the emission of that gas.

In our paper we adjust the model CO emissions to match the DC8 observations by adjusting the estimate of the amount of biomass burned, which implies a change to fuel consumption and/or burned area. This procedure implicitly assumes that the emission factor for CO is perfect, and changes emission rates of all gases by the same proportion, as discussed in page 15342 lines 1-5 of our original paper. We make no statements as to whether the errors in the biomass burned are related to combustion completeness or to burned area. We also change the initial emission factors for NOx,



and partition the NOx emissions between NOx, PAN, and HNO3 to account for the rapid in-plume chemistry that the global model cannot resolve, as discussed in page 15342, line 5-15 of our original paper. This is only one set of possible assumptions that could lead to the same emission rates, and reflects our attempt to make the minimum number of adjustments to the model required to better match the observations. We have expanded our discussion in the revised paper (line 393-397) to clarify this.

**Have the authors a clear idea of the origin of each intercepted plume? If yes, can they relate the plume to vegetation type? Soja et al, (2008) suggest that crown fire inject smoke (and presumably gases alike) to altitudes of 5-7 km. This appears to be inconsistent with the authors' prior assumption about injection in the boundary layer.**

We used back trajectories to determine the approximate origin of each intercepted plume, as discussed in page 15330, lines 21-23 of our original paper. The plumes encountered over the active burning regions (area B of Figure 4 in the original paper) were generally encountered at low altitudes (< 3 km) near their source, so back trajectories were unnecessary. We identified only one pyroconvection event in the DC8 dataset (Area A in Figure 4). It is not clear what data Soja et al., 2008 used to determine their estimate of injection heights, but using the cloud index data of Bruce Anderson we find 2 fresh plumes mixed with liquid water or ice clouds in Area B and 3 in Area C, consistent with their results (see our response to the third comment of Reviewer 2 below). However, we have not attempted to identify an exact origin for each smoke plume, or to identify the vegetation and ecosystem types present. The FLAMBE inventory uses 10 land cover classes, as described in Reid et al. (2009) which we cite in the paper. Maps showing fire counts during ARCTAS can be found in Jacob et al. (2010), the overview paper. The GEOS-Chem model allows for different emission factors for three broad types of vegetation, extratropical and tropical forests, and grasslands.

The injection of biomass burning emissions only within the boundary layer is an important limitation of our study, as we discuss on pages 15333-15334 and page 15343 of

our original paper. See our response to the third comment above as well.

**The authors also note the role of storm updrafts as a mechanism for lifting emissions above the boundary. Is there an approach the authors could use with the DC8 data to separate the plumes lifted by pyroconvection versus storm updrafts?**

In most cases, storm updrafts would occur without the fires, but the fires provide added buoyancy leading to stronger updrafts and perhaps a greater concentration of clouds. We are not aware of a diagnostic way to assess what component of the updrafts would occur without a fire, and what component is due to the fires using the DC8 data alone. One way to assess the role of fires would be to perform very high resolution modeling and compare the updrafts and vertical mass flux with and without fires included in the simulation (see, for example, Luderer et al., Atmos. Chem. Phys., 6, 5261-5277, 2006). However, this would be a major meteorological modeling effort, and is well outside the scope of this paper.

**Page 15341: Can the authors explain the approach taken to determine pseudoemission factors that account for sub-grid processes.**

The pseudo-emission factors are calculated using Eqn. 1 on page 15340 of the original paper, where the emission ratio ERi used for each species is replaced with the observed mean enhancement ratio within fresh plumes from Table 4. Thus the procedure is identical to the calculation of emission factors, with the exception that the true emission ratio is replaced by the observed enhancement ratio in fresh plumes. We have clarified this in the revised paper (lines 361-363).

**From section 3.3.1. it appears no one sensitivity calculation addresses the discrepancy between the model and the DC8 measurements. Any suggestions for other avenues of research?**

It is true that none of the sensitivity studies shown in this paper give an exact match



between the model and observations. However, the GC-SENS model simulation represents a substantial improvement of the model performance over the GC-ORIG model for the chemical species examined here. Remaining issues include the underestimate of high-altitude CO discussed on page 15342, the underestimate of ozone discussed on page 15343, and the erroneous partitioning between HNO3 and PAN discussed on page 15344. We suggest the following avenues of future research in our original paper: investigating the injection of biomass burning emissions above the boundary layer (p. 15342), investigating stratospheric-tropospheric exchange of ozone near boreal smoke plumes (p. 15343, 15347), investigating the sources and sinks of acetaldehyde to address the PAN discrepancies (page 15344), and developing a better understanding of the distributions of fuel nitrogen content and the effect of combustion phase on NOx emissions (page 15348). We have added these recommendations in the conclusions of the revised paper (lines 573-576) to ensure they are given proper emphasis.

**I remain unconvinced about the results shown in section 3.3.3. How much of the TES observed CO and TES, particularly in the lower troposphere, is due to the a priori used in the retrieval?**

See our discussion of TES averaging kernels, DOFS, and differences from a priori estimates in response to the second comment. There is sufficient signal in these profiles (1 DOFS below 250 hPa for both CO and O3) to ensure we are not merely retrieving the a priori, and the retrieved values differ from the a priori. We have added text to the revised paper at lines 139-147 to address these concerns.

**Figure 3: Can the authors provide some indication of the altitude for each intercepted plume? The colours could be used to denote the altitude and the symbols used to denote the two ratios.**

We have added the altitude information to the figure using text labels.

**Figure 5: This reader would find it useful if the plot also showed how many measurements were used at each altitude to determine the mean statistics. The**

**legend needs to be larger.**

We have added the requested statistics and increased the legend size in this figure.

**Figures 11, 14 and 18. Suggest it might be better to show the differences between the model and TES.**

We have decided to show the TES data, rather than model-TES differences, because we are using these figures to illustrate two points: (1) there is little correlation between TES ozone and CO in these retrievals and (2) there is little correlation between model ozone and CO for these plumes as well. Merely showing model-TES differences would not allow readers to evaluate for themselves whether there is a correlation between the ozone and CO in the plumes as retrieved by TES.

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