Atmos. Chem. Phys. Discuss., 14, C11916–C11924, 2015
www.atmos-chem-phys-discuss.net/14/C11916/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Properties and evolution of biomass burning organic aerosol from Canadian boreal forest fires" *by* M. D. Jolleys et al.

M. D. Jolleys et al.

matthew.jolleys@manchester.ac.uk

Received and published: 5 February 2015

Reviewer comment 1: I would follow the discussion of the near and far-field ratios with some-thing like "However, the changes in combustion conditions between the near- and far-field samples (smoldering for the near-field, flaming for the far-field) make it unclear whether the apparent decrease is due to aging or the change in combustion efficiency." Right now, it sounds like you are saying it is clearly due to aging, and that the changing combustion conditions are a minor effect, and I don't think you have proven that.

Author response 1: We agree that the contrasts in OA/CO between fresh and aged plumes are a consequence of both aging and combustion conditions, as is stated in this section (Manuscript L22-24). Regardless, a line has been added to clarify

Change 1: Text added L27 - 'creating an underlying contrast in emissions prior to any transformations associated with aging'

R2: I don't think I understand this sentence. F44 is lower in the fresh smoke than in the aged according to Figure 7, right? If so, I think this sentence is potentially misleading. Do you mean that you would have expected more formation in the far-field plumes than you observed?

A2: This sentence was simply intended to state that f44 is lower in the near-field plumes as a result of shorter aging times compared to more aged, transported plumes, and has been edited to clarify

C2: Text edited L32-34 – 'f44 is lower on average in near-field plumes than those sampled in the far-field, in accordance with longer aging times as plumes are transported a greater distance from source'

R3: Again, I don't think you can state that the effect you observed was primarily due to aging and not due to combustion phase given the evidence you present. You need to put both effects on more equal footing in your discussion.

A3: Text altered to provide greater balance

C3: L41-44 – 'The elevated levels of oxygenation in aged plumes, and their association with lower average ïĄšOA/ïĄšCO, are consistent with OA loss through evaporation during aging due to a combination of dilution and chemical processing, while differences in combustion conditions throughout the campaign also have a significant influence on BBOA production and composition'

R4: I think you want the word "emissions" after "(BBOA)"

C4: Added (L52)

R5: This sentence doesn't really say much that I relevant to the paper, so I'd suggest cutting it.

ACPD

14, C11916–C11924, 2015

A5: We think it's important to state how this work could ultimately contribute to reducing uncertainty through the use of aerosol measurements and to define the overriding objective for this research

R6: There are several places you use this "influence of" phrasing, but it sounds awkward to me. Here, you don't mean the influence of SOA is unclear, you mean the rate of formation and loss of OA is unclear, or the importance of SOA is unclear, right?

C6: Changed to importance (L67)

R7: I don't see why Figure S1 and S2 are not in the main paper rather than the supplement. They seem pretty important to understanding the conclusions of the paper, so I'd recommend putting them in the main text. More discussion of the source locations and transport pathways o the plumes, including any potential vertical motion during transport, would also be useful.

A7: Discussion of source locations and plume transportation for BORTAS is provided by O'Shea et al. (2013, ACP), while back trajectory analysis for individual plumes sampled in flight B626 is also provided by Taylor et al. (2014, ACP). Given the large total number of plumes included in this study, more detailed analysis of individual plume histories (as in Taylor et al.) would be a significant undertaking and is beyond the scope of this paper

C7: Figures moved to main manuscript, reference to analysis of back trajectories in O'Shea et al. and Taylor et al. added (L97-100)

R8: I'd say "this size range" rather than "the full size range" – the second makes me think you mean the full size range of the aerosol distribution, not just the part measured by the SMPS

C8: Changed (L146)

R9: Do you mean the excess particle number concentrations as well, or the absolute values?

ACPD

14, C11916–C11924, 2015

C9: 'Absolute' added (L157)

R10: I think these two sentences would fit better after L2, when the CO and number conc. thresholds have been introduced.

C10: Sentences moved (L157)

R11: This section was confusing to read. The beginning of this section suggested that your proposed indicators were already well established, but here you are presenting evidence of how they correlate with CH3CN and HCN. If this comparison is a key result o your paper, it should be in the results section, but if the indicators you are using have been used successfully in the past, it's not clear why you need this additional evaluation.

A11: This was included as without it it may be somewhat confusing as to why two different classification schemes have been applied to data from the same campaign, especially as the Le Breton et al. method was already published at this point and several references are made to the paper throughout this manuscript.

C11: L173-181 – 'A scheme using a HCN concentration threshold of six times the standard deviation (6σ) has been used during BORTAS in an analysis of high sensitivity 1Hz chemical ionisation mass spectrometer (CIMS) measurements and their consistency with CO and CH3CN concentrations (Le Breton et al., 2013). However, as many previous datasets do not include HCN measurementsa screening procedure using only OA, CO and number concentration data has been applied here, so that the approach can be used consistently across a broader range of data.'

R12: Consider changing to "at the source to up to 5 days after emission."

C12: Changed (L208)

R13: Concentrations of what in aged plumes? OA? CO? Both?

C13: Changed to clarify (L227-230) –'Concentrations of the majority of sampled

ACPD
14, C11916–C11924, 2015

species in aged plumes during flights B621-B624, including OA, CO, CO2 and BC, consistently exceeded those at source from B626, irrespective of the effect of dilution as plumes dispersed into the ambient atmosphere'

R14: I get 180/50, not a factor of four, from Figure 1a. Either the text is wrong or the figure is cut off and should be fixed.

C14: Changed to 3.6 (L231)

R15: CO is also associated with moldering combustion.

C15: Changed to 'both OA and CO' (L234)

R16: I'd suggest "analyzed" instead of "relevant" here.

C16: Changed (L246)

R17: Is this a typo? How can the campaign average be less than both the fresh and aged samples?

R17: Average had not been updated from an older iteration, now corrected

C17: Changed to 0.104 ± 0.003 (L249)

R18: I think you can cut this sentence.

C18: Removed (L267-268)

R19: I think you want to add "in the aged plumes" after progressively and end the sentence with ", suggestive of OA losses during aging in these plumes with predominantly flaming smoke."

C19: Changed (L272)

R20: Why don't you discuss the modified combustion efficiency (MCE) of the plumes when you discuss combustion conditions? That should give you a pretty god indication of the change in combustion phase.

ACPD
14, C11916–C11924, 2015

A20: We did attempt this, but it did not provide any meaningful results, largely due to the difficulty in calculating a background value for CO2 (hence why we only report absolute CO2, not ïĄšCO2) given the high variability both in and out of plume. We attempted several different methods of determining background levels, including averaging for different periods of time immediately before and after plume interceptions and using different thresholds for the standard deviation in designated out-of-plume periods, but none proved to be successful.

C20: Changed – 'Background concentrations for CO and other species were calculated for each flight according to minimum observed concentrations. CO2 was the only exception, with high variability both in and out of plume making it difficult to define an appropriate background concentration. As a result, only absolute concentrations are reported for CO2, as opposed to excess values.' (L158-162)

R21: You need to make clear here that f44 isn't exclusively produced during smoke aging, but that fresh some also has a significant amount of f44 as well.

C21: Changed – 'm/z 44 is also a constituent of fresh smoke and has been shown to be significantly elevated at source, dependent on combustion conditions' (L284)

R22: Again, why don't you use MCE here?

A22: See response to R20 above

R23: I think you switched your numbers here?

A23: Yes, they were the wrong way round here

C23: Changed – '0.128 $\pm$ 0.006 to 0.078 $\pm$ 0.003' (L371)

R24: low CO2 relative to what?

C24: Added – 'relative to levels throughout the rest of the aged plumes' (L382)

R25: I can't see any correlation with f44, but I can see it with f60.

ACPD

14, C11916–C11924, 2015

A25: This is described in more detail in line 381 onwards, addressing the varying correlations for each case. Regardless, OA and CO do show an overall decrease with f44 – when binning by f44, maximum average OA and CO both occur at minimum f44, and vice versa

C25: Added – 'Furthermore, when binning concentrations by f44, maximum bin-average ïĄšOA and ïĄšCO both coincide with minimum f44 and vice versa.'

R26: This is true, but almost trivially so. Aren't you just saying that aging is more important for the aged smoke?

C26: Removed R27: I'd add "during BORTAS" after "plumes" here.

C27: Added (L431)

R28: Missing delta before BC.

C28: Added (L444)

R29: Didn't you say on page 25110 that this boundary layer enhancement was due to biogenics? Doesn't your statement here conflict with that?

A29: This refers to the more general elevation in OA/CO and f43 in the boundary layer, where as the possible influence of biogenics (if it even is that) is a single, isolated case of particularly high OA/CO ($\sim$0.4), which we state in the text (L426) – 'Whilst the further properties of aged plumes discussed here would suggest this effect is isolated and limited in its overall impact. . .'

C29: Changed – '. . .contributing to the typically higher levels of ïĄšOA/ïĄšCO and f43 at low altitudes.' (L448)

R30: I don't think this sentence adds anything, so I'd cut it.

C30: Removed

R31: Could this be due to secondary production of aldehydes and ketones in the smoke

ACPD

14, C11916–C11924, 2015

plumes?

A31: This is possible, and is discussed in lines 275-282. The second reviewer also commented that this interpretation had been clearly presented

C31: Added – 'However, fragmentation of oxygenated aldehyde and ketone molecules has been shown to produce elevated levels of f43 relative to f44 in BB emissions (Schneider et al., 2006), suggesting possible contributions from secondary formation.' (L506-508)

R32: I'd ay a range of "ages and combustion phases" is more accurate

C32: Changed (L606)

R33: This is not a fair summary of the literature. You do need to point out the many studies that have found significant OA formation in biomass burning plumes here as well.

A33: Text altered to provide greater balance

C33: Changed – 'While contrasting aging behaviours and significant SOA formation have been identified in some studies, an absence of increasing ïĄšOA/ïĄšCO has been observed in several previous BB assessments. The trend of decreasing ïĄšOA/ïĄšCO with increasing distance from source in BORTAS further emphasises the importance of source conditions for aging plumes.' (L609-613)

R34: This last sentence is more of a generically true statement about fires than a conclusion of your paper, so I would cut it.

C34: Removed

R35: I'd say "near the source", not "at the source" here.

C35: Changed (L630)

R36: I'd add "near the source" at the end of this sentence on photochemical age.

ACPD

14, C11916–C11924, 2015

C36: Changed to ‘over short aging times’ (L633)

R37: I’d say “these aging BB plumes” to make clear again that other have gotten different results.

C37: Changed (L634)

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 25095, 2014.

ACPD

14, C11916–C11924, 2015