Atmos. Chem. Phys. Discuss., 14, C11925–C11928, 2015
www.atmos-chem-phys-discuss.net/14/C11925/2015/

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

R1: A cohesive paragraph in the methods section summarizing which flights and flight segments are used to make conclusions between fresh and aged plumes; near and far-field characteristics would be very helpful. This information is currently buried in the text, among further discussions of measurement analyses by which other flights that were excluded from the analysis.

C1: Added – 'Measurements from 5 BORTAS flights (B621-B624 and B626) were included in this analysis. Flight B626 provided the only measurements of fresh BB plumes throughout the campaign, with all other flights sampling air masses downwind of the source region at ages of several days. Data were screened in order to isolate

emissions with a biomass burning influence, resulting in a total number of 26 valid plume interceptions (3 fresh and 23 aged) across the 5 flights.' (L150-154)

R2: The number of plumes/flights used to make conclusions should also be reflected in the abstract and conclusions so that they do not seem more general than they are.

C2: Changed - 'Measurements at source comprised 3 plume interceptions during a single research flight and sampled largely smouldering fires. 23 interceptions were made across 4 flights in the far-field. . .' (L24-28) 'Average ïĄšOA/ïĄšCO in 3 plumes sampled close to source (0.190 $\pm$ 0.010) exceed ratios in the far-field from 23 interceptions. . .' (L606-607)

R3: The authors mention two flights (B622 and B624) as having captured a decrease in OA/CO over various segments of its flights (Section 3.1); can other conclusions in the manuscript regarding the contribution of atmospheric processing be strengthened by further examination of these two scenarios?

A3: This is a more general point about the overall decrease in average OA/CO with increasing distance from source, ie. the lowest average is observed for B624, which is furthest east over the Atlantic, while the highest is for B622, closest to the source region in the west. However, individual OA/CO ratios for each measurement point also decrease overall with increasing distance from source, based on co-located positional data, although there is significant variability throughout this trend.

R4: While "aging" and atmospheric "processing" is used very often in the community, the authors may find it useful to describe the processes embodied in this term (e.g., heterogeneous reaction, condensation/evaporation) such that the discussion regarding observed variations in f44, f43, and f60 can be tied to specific mechanisms.

A4: Clarification of specific processes now given where relevant

C4: 'The elevated levels of oxygenation in aged plumes, and their association with lower average ïĄšOA/ïĄšCO, are consistent with OA loss through evaporation during

ACPD

14, C11925–C11928, 2015

aging due to a combination of dilution and chemical processing' (L40-43) 'Several fundamental aspects of the BBOA lifecycle remain poorly characterised (Hallquist et al., 2009), including the conditions and processes controlling formation and the effects of transformations occurring during aging, such as gas-particle partitioning of low volatility organic compounds following photo-oxidation, heterogeneous reactions with existing OA and losses through dilution-based evaporation or volatilisation' (L60-64) 'f60 was also shown to decrease concurrently with increasing f44 during ARCTAS, as a result of the oxidation of primary levoglucosan-type species with aging.' (L303-305) 'However, these changes also coincide with a trend of decreasing ïĄšOA/ïĄšCO (Figure 8c, left panel), belying the expected addition of OA mass resulting from increasing oxygenation as semivolatile products condense to the particle phase.' (L473-475)

R5: Should not the CO and OA be defined with respect to altitude? As the authors point out, their background concentrations have different altitude-dependent profiles.

A5: Background OA concentrations remain below 1 ugm-3 throughout the vertical profile derived for BORTAS, while background CO only varies between 20 and 25 ppb up to a height of 6000m, above which there is more variation (up to 40 ppb). However given the absence of BB plumes above 6000m this does not affect this analysis, while the effects of the limited variations lower down the profile will be minimal

C5: Changed – 'Background concentrations for CO and other species were calculated for each flight according to minimum observed concentrations, which were applied to all measurements throughout the full vertical extent of sampling, given the limited variation in background concentrations with altitude.' (L158-161)

R6: The discussion of ER and NEMR and its use should appear sooner, e.g. in the Methods section, as ratioed values are used extensively throughout the manuscript. There should also be a caveat that the proposed interpretation applies along a Lagrangian trajectory, which corresponds only to a few contexts in this study (when a liberal interpretation of a Lagrangian trajectory is used)

ACPD
14, C11925–C11928, 2015

C6: Changed – ‘Throughout this study, extensive use is made of normalised measurements as a means of assessing the relative abundances of different species. Normalising to a co-emitted, non-reactive tracer such as ïĄšCO provides an emission ratio (ER) when calculated at source. Normalised excess mixing ratios (NEMR) are used to represent these values for any other point in a plume away from source along a Lagrangian trajectory, and account for the effects of dispersion as concentrations in plumes decrease through dilution. These ratios can also be used as a marker for potential SOA formation, as the longer atmospheric lifetime of CO ($\sim$1 month) relative to that of OA (on the order of several weeks) makes it likely that any enhancement of the ratio between the two species will be a result of the addition of OA, rather than increased removal of CO in isolation.’ (L198-206)

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