

We thank the anonymous reviewers for their useful comments and suggestions. We present the reviewers' comments in bold, our responses in standard font and the changes to the text in italics. Additionally, we have added Dr Mark Parrington to the author list to recognise the contribution he made during the flight planning, though this does not affect the scientific content of the paper.

Responses to anonymous reviewer #1

The following four comments have been addressed together

Abstract. "There are few qualitative studies of wet removal in ambient environments."

Yes, but there are additional relevant studies (e.g., Kreidenweis et al., 1997; Jacobson, 2003). Please discuss.

Introduction. "Wet deposition is the dominant mechanism for BC's removal" and "Such (vertical) profiles are often poorly represented." However, when wet deposition is treated physically, vertical profiles can be relatively consistent with data, supporting the contention that wet removal is the dominant mechanism of BC removal (Jacobson, 2012, Figure 9 and Table 3).

Introduction. "Alternatively, many models use parameterizations designed to emulate: : ." Please clarify that some global models treat explicit size-resolved cloud liquid, ice, and mixed-phase microphysics (e.g., Jacobson, 2012, 2003).

Introduction. "Below-cloud scavenging is calculated by multiplying the precipitation rate by a scavenging coefficient." Again, please clarify that some global models treat explicit size-resolved collision-coalescence and activation, accounting for composition (Jacobson, 2012, 2003).

The end of this paragraph now contains the sentence

"Some models treat explicit size- resolved, cloud liquid, ice and mixed-phase microphysics (e.g. Jacobson, 2003), and this can generate better agreement with measured vertical profiles. Observations (particularly of size distribution, hygroscopicity and mixing state) are needed to constrain both types of wet removal scheme (Koch et al., 2011). Explicit microphysical models are also used to investigate smaller scale phenomena (e.g. Kreidenweis et al.,1997), and ambient measurements are similarly required for constraint and comparison."

Abstract. The conclusion, " : : suggesting that nucleation scavenging was the likely dominant mechanism" should be clarified to state whether this is referring to mass or number. If mass, the result appears consistent with that of Jacobson (2003), who states in the abstract, "washout (aerosol- hydrometeor coagulation) may be a more important in-plus below-cloud removal mechanism of aerosol number than rainout (the opposite is true for aerosol mass)."

This sentence now reads

"This depleted the majority of the plume's BC mass, and the largest and most coated..."

This is consistent with Jacobson (2003)- we have referenced this work several times in the text but do not want to in the abstract as it distracts from our work.

Introduction. "For typical BC size distributions, : : , impaction scavenging therefore favours smaller BC, whereas nucleation scavenging favours larger." This conclusion

was drawn in Jacobson (2003). Please clarify.

We have added a reference to this paper at the end of the sentence

P. 19481. Please state up front whether the BC/CO, etc. ratios in the plumes are measured or modeled and for what time period they apply to.

Figures 4-7. It is not clear at what time during the plume the measurements are valid for.

The sentence now reads

“Figure 3 shows the BC/CO, OA/CO and BC/B_{Scat} ratios measured in for the three plumes described in Sect. 2.3”

Section 2.3 contains details of the in-plume criteria and references table S1 which contains the plume location and times.

With respect to the shell to core ratio, at what relative humidity was the coating determined at? As the RH approaches 100%, the particle swells to a large ratio of shell to core, increasing the MAC. If only the shell to core ratio of a dried particle is examined, this will underestimate the MAC (Jacobson, 2012). The authors may need to re-do calculations if they used the shell to core ratio of a dried particle.

We have added the sentence

“We are also unable to constrain any Hygroscopic growth the particles may undergo at ambient humidities, and therefore all our calculations are performed under the dry (< 40%) RH conditions of the sample line.”

This is a limitation in this work but the effects of hygroscopic growth on optical properties (and hygroscopic growth in general) are subject to much ongoing research and are not in the scope of this analysis.

Responses to anonymous reviewer #2

It seems that only one back trajectory was calculated for each of the three plumes.

Given the uncertainties of the model and the fact that all further analysis and argumentation is based on these results a sensitivity study is necessary. It should at least include multiple releases of back trajectories from slightly varied locations around the plume centers to test the robustness the results.

We refer the reviewers to Figure 6c in O’Shea et al (2013). They performed backtrajectories over the entire flight track from B622, almost all of which passed over the region of fires in question. We considered making a similar plot, with many trajectories, but it made it impossible to see the fire locations in Fig.1 and the satellite data in Fig. 2. The comparison with O’shea et al. shows our trajectory results are robust. The following sentence has been added to Section 2.4.

“A comparison with O’Shea et al. (2013, Fig. 6c), who performed back trajectories along the entire flight track of B622, shows that these trajectories are representative of the airmass history at these altitudes.”

The ratios presented in Section 3.2 are not emission ratios but ratios that are affected by chemical and microphysical processes in the atmosphere. Rename the section to “chemical characteristics”, “tracer ratios” or something similar and change the terminology for the chemical ratios that are not emission ratios in the text.

This section is now called Chemical Tracer Ratios and the text has been edited when referring to the ratios measured in this study

For better comparability with existing and future studies of biomass burning aerosol, more information on the organic aerosol should be included to elucidate the chemical age of the plumes you describe. Given the AMS data set you can include the fraction of the mass to charge ratio 44 of the whole OC mass (f_{44} , e.g. Ng et al. 2010) or convert this into the O:C ratio. Either parameter will give further ideas regarding the solubility of OA due to its degree of oxygenation which might play a role for the wet removal, and can further describe chemical similarities or differences between the plumes which again is crucial for your analysis. In addition, including more information on the OA concentrations in plume 3 in the text would be helpful, because values cannot be read from the graph due to the coarse resolution of the axis.

We have added the maximum OA concentration measured in plume 3 to the caption on figure 3. We have also added a paragraph to the end of section 3.2.1

“OA comprised over 85% of the aerosol measured in Plumes 1 and 2. The fraction of OA mass measured in the AMS at m/z 44 (f_{44}) provides a measure of the oxygenation of the OA fraction (Ng et al., 2010). The mean values of f_{44} measured in Plumes 1 and 2 were 0.085 and 0.120 respectively, which are indicative of a reasonable degree of oxidation. Previous studies have shown that increased f_{44} is qualitatively related to increased hygroscopicity (e.g. Duplissy et al., 2011), though this is to an extent system dependent. The f_{44} in these plumes indicates the OA was likely hygroscopic and may act as a CCN at sufficient supersaturations. The mass of OA measured at m/z 44 in Plume 3 was not sufficient to make a robust calculation of f_{44} , so it is not possible to make a comparison.”

Include information on how many data points you have from the SP2, AMS and SMPS measurements during plume interception.

The number of data points are now included in Figure 3

Specific comments:

Be more specific about the different types of diameters throughout the manuscript (e.g. mobility diameter on p. 19477, l. 24.)

This line now reads

“...measured distributions of particle mobility diameter (D_{mob}) divided into...”

The caption to Fig. 4 now says D_{mob} instead of D_p

We have also clarified in the definitions of CMD and MMD that they refer to D_c

Section 2.1: There is no information about the flight track and altitude. Insert the flight track in figure 1 and include a more detailed description in this section.

The flight track has been added to figure 1 and the following has been added to section 2.1

“The flight track for B622 is shown on Fig. 1. The ARA took off from Halifax Stanfield International Airport at 14:56 UTC and flew a series of sawtooth and straight and level runs on a generally East-West axis, before landing at Québec City Jean Lesage International Airport at 19:11 UTC.”

Section 2.3: Elaborate how you identified the regional background and why $R^2 \geq 0.55$ is considered as threshold for a good correlation between CO and CH₃CN.

The background is already stated in the text

“These thresholds were determined from the 99th percentile measured in background air on flight B625 on 24 July 2011, on which no biomass burning plumes were detected (Palmer et al., 2013)”
An R^2 of 0.55 does show a strong degree of correlation between CO and CH₃CN. We did not consider it a threshold and have not used it as such. To remove confusion, the 0.55 has been removed from the text, which now reads
“...good correlation between CO and CH₃CN, which are commonly-used biomass burning tracers.”

Section 3.1, until p. 19480, l. 5: These are not results, move these paragraphs to a new subsection in section 2.

This section has been moved and is now Section 2.4

p. 19480, l. 12: give numbers for the altitudes

This sentence now reads

“...a vertical emission profile which would be expected to join the trajectories at the altitudes shown in Fig. 1.”

p. 19480, l. 16: How many hours before sampling?

This now reads

“The back trajectory from plume 3 showed precipitation between 11:30–13:00 and 21:00–22:30 UTC on July 19 2011 (approximately 27 and 18 hours before sampling respectively), as indicated by the aqua-coloured regions in Fig. 1.

p. 19481, l. 14: give a reference that describes the decrease of the OA/CO ratio due to evaporation

We have added a reference to Donahue et al. (2011)

p. 19485, l. 2: Is this only because of the coating or also because of the size? Include a brief discussion on this.

This sentence now reads

“This suggests that the more coated particles were more effective CCN, and hence were removed more efficiently like due to both the increased D_p and greater soluble content.”

p. 19488, l. 11: Elaborate, from the given information it's not clear why the result would be 0.1.

This sentence now reads

“We have not quantified TE_{BC} in this study, but by dividing the BC/CO ratios listed in Table 1 it was ~ 0.1 – 0.2 ”

p. 19488, l. 13f: Has this been shown before? If yes, give references.

We are not sure what the referee is specifically referring to but we have made reference a number of times to *Moteki et al.* (2012) which is the only comparable study we are aware of.

Table 2: The nomenclature for plumes of the ARCTAS campaigns is confusing, because it's not clear that “ARCTAS Asia” means BB plumes from Asia measured during ARCTAS. Change the nomenclature so that this becomes clear. If there is information on the age of plumes that are

compared in this table and the type of fire (smoldering, crown fire etc.) include it, because it is important information regarding their comparability.

The nomenclature has been changed

ARCTAS ASIA → ARCTAS Asian BB

ARCTAS Canada → ARCTAS Canadian BB

ARCTAS-CARB California → ARCTAS-CARB Californian BB

TexAQS biomass burning → TexAQS BB

The table caption also now has the added sentence

“The ARCTAS values are averages of multiple fires sampled over North America.”

The ARCTAS measurements are campaign average and cover a range of fire types. For the additional information, Kondo et al. (2011) showed that CMD and σ_{geo} did not substantially change with differences in MCE, age or fire location. There was a weak relationship between CMD and σ_{geo} and MCE, but as we do not have robust MCE measurements from our data this contributes nothing to our analysis.

Figure 2: Include the back trajectories in this plot for better readability.

Done

Technical comments:

p. 19471, l. 13: single-scattering albedo

Done.

p. 19471, ll. 15-17: “: : , possibly due to the thick coatings: : :” does this refer to the particles you measured or to the Asian outflow aerosol?

This now specifies “Canadian biomass burning particles”

p. 19471, l. 17: “: : : provides important constraints: : :” give examples

This now reads

“This study provides measurements of BC size, mixing state and removal efficiency to constrain model parameterisations of BC wet removal...”

p. 19471, l. 26: insert “: : :as well as its chemical processing and lifetime in the atmosphere and optical properties. “

This has been changed

p. 19473, first sentence: BC aerosol that is coated with hydrophilic material wouldn't be considered fresh anymore. The sentence needs to be rewritten accordingly.

This now reads

“Fresh BC is generally considered hydrophobic, though it may act as a cloud condensation nucleus (CCN) in liquid cloud if subsequently coated with hydrophilic material”

p. 19474, l. 4: insert “BC” after “diesel-dominated”

Done

p. 19474, l. 8: insert “a” before “precipitating cloud”

Done

p. 19474, l. 8: delete the sentence “Franklin et al: :” There is no gain in information.

We have clarified that our study directly helps explain the results of Franklin et al, and the two studies are complementary

“Franklin et al. (2014) recently highlighted aerosol depletion in similar plumes using remote sensing measurements, but were unable to determine the mechanism or measure the properties of any particles remaining in the plume. We examine the aerosol size distributions and BC coating properties using in-situ measurements sampled in the three plumes to determine the most likely removal mechanism and consider the effect this has on optical properties.”

p. 19475, l. 9: split sentences here: “: : temperature. During BORTAS: :”

Done

p. 19477, l. 4-5: organic aerosol, no capital letters

Done

p. 19478, l. 23: delete “in the Supplement” before “in Table S1”

Done

p. 19479, l. 9: What do you mean by “contrasting”?

This word has been removed

p. 19479, l. 11: use the newest reference for the HYSPLIT model

The reference we have used is the recommended reference on the HYSPLIT website

p. 19480, l. 22: the correct time period is: 12:45 – 13:00 UTC

This is the satellite time, not the time of the HYSPLIT calculated precipitation. To clarify, the sentence now reads

“The back trajectory from plume 3 showed calculated precipitation between 11:30–13:00 and...”

p. 19480, l.29: give a reference

Added reference to (Lin & Rossow, 1997)

p.19482, l. 26: specify which ARCTAS campaign/s

We have specified ARCTAS-B

p. 19490, l. 14: BC removes environments? I think it’s “remote”.

Done

p. 19490, l. 18: “: : because the original size distribution was smaller than Moteki et al: :.”. I hope that the particles are smaller than Moteki et al: :. Please change to:

“: :was smaller than the one described by: :”

Done

Figure 5, 6, 7: Include a note regarding which instrument generated the data.

Done

Responses to anonymous reviewer #3

Page 19471, line 22. IPCC AR5 (fig. 8.17) suggests that other well-mixed GHGs (in particular CH₄) may also have a stronger RF than black carbon.

This sentence now reads

“Black carbon (BC) is the dominant absorbing aerosol in the atmosphere and is an important, ubiquitous climate warming agent (Ramanathan and Carmichael, 2008; Chung et al., 2012; Bond et al., 2013).”

Page 19472, line 8. Textor et al. shows that wet deposition dominates in models. The models are probably correct in this regard, but nevertheless this should be made clear, or reference made to a study validating this aspect of the models with observations.

This sentence now starts with

“Modelling studies suggest wet deposition...”

Page 19478, line 25. Please explain the significance of a correlation between CO and CH₃CN.

We have clarified that CO and CH₃CN are commonly used biomass burning tracers.

Page 19479, lines 17–23. While it is reasonable to expect HYSPLIT to resolve the largescale ascent associated with a front, and the associated vertical tracer transport due to the lifting of the warmer air mass, it is not clear from either the explanation given or the reference (Stohl et al.) that the model will capture the full extent of the transport due to unresolved deep convection triggered by the resolved frontal ascent (which is the type of precipitating cloud encountered by the plume, as described on page 19480, line 24). Please clarify the extent to which the model can be expected to capture this deep convection, as opposed to the frontal ascent with which it is associated, and the implications of this for the analysis presented.

The text is already clear that it will not capture any small-scale convective systems, but can capture large-scale ascent in the front in which the clouds are embedded. Perhaps more importantly, it also cannot capture the pyroconvection associated with the fire plume that lofted the smoke up to “join” the trajectory. We have also explained this in the next paragraph. What happened to the air before it entered the region of fires and clouds, and how much of the convection was due to the fires, frontal uplift or isolated convective cells is largely irrelevant to our analysis. We have used the back trajectories to track the plume back to the region of cloud/fires and used this as one of several parts of our argument that most of the aerosol acted as CCN and were rained out.

Page 19482, lines 6–8. The sample size (three plumes, one of which encountered precipitation) seems too small to justify the strength of conclusion (“It is therefore clear that ... were largely the result of the wet removal”) without demonstrating

that the result cannot be due to variability between plumes. Please clarify why the result is robust despite the small sample size, or qualify the statement.

The conclusion is not just from the HYPLSIT runs it is from the combination of HYSPLIT/GOES and the comparison to literature BC/CO and OA/CO. The literature values are from many plumes in many different environments and the only ratios that come close are the ones that were affected by precipitation. This sentence now reads

“By comparing to literature values and the other plumes in this study, it is clear that both BC/CO and OA/CO in Plume 3 were largely the result of the wet removal itself, rather than the initial combustion conditions.”

Also, regarding the small sample size, see the response to reviewer 2’s first comment and the comparison to O’Shea et al (2013).

Page 19483, line 4. Change “was likely have had” to either “would likely have had” or “was likely to have had”.

Done

Page 19488, line 20. Change “by (Schwarz et al., 2010b)” to “by Schwarz et al. (2010b)” (i.e. \citep to \citet if using LATEX).

Done

Page 19490, line 14. Change “remove” to “remote”.

Done

References

- Donahue, N. M., Epstein, S. A., Pandis, S. N., & Robinson, A. L. (2011). A two-dimensional volatility basis set: 1. organic-aerosol mixing thermodynamics. *Atmospheric Chemistry and Physics*, *11*(7), 3303–3318. doi:10.5194/acp-11-3303-2011
- Duplissy, J., DeCarlo, P. F., Dommen, J., Alfarra, M. R., Metzger, A., Barmpadimos, I., ... Baltensperger, U. (2011). Relating hygroscopicity and composition of organic aerosol particulate matter. *Atmospheric Chemistry and Physics*, *11*(3), 1155–1165. doi:10.5194/acp-11-1155-2011
- Franklin, J. E., Drummond, J. R., Griffin, D., Pierce, J. R., Waugh, D. L., Palmer, P. I., ... Saha, A. (2014). A case study of aerosol depletion in a biomass burning plume over Eastern Canada during the 2011 BORTAS field experiment. *Atmospheric Chemistry and Physics Discussions*, *14*(3), 3395–3426. doi:10.5194/acpd-14-3395-2014
- Jacobson, M. Z. (2003). Development of mixed-phase clouds from multiple aerosol size distributions and the effect of the clouds on aerosol removal. *Journal of Geophysical Research*, *108*(D8), 4245. doi:10.1029/2002JD002691

- Kondo, Y., Matsui, H., Moteki, N., Sahu, L., Takegawa, N., Kajino, M., ... Brune, W. H. (2011). Emissions of black carbon, organic, and inorganic aerosols from biomass burning in North America and Asia in 2008. *Journal of Geophysical Research*, *116*(D8), D08204. doi:10.1029/2010JD015152
- Lin, B., & Rossow, W. B. (1997). Precipitation water path and rainfall rate estimates over oceans using special sensor microwave imager and International Satellite Cloud Climatology Project data. *Journal of Geophysical Research*, *102*(D8), 9359. doi:10.1029/96JD03987
- Moteki, N., Kondo, Y., Oshima, N., Takegawa, N., Koike, M., Kita, K., ... Kajino, M. (2012). Size dependence of wet removal of black carbon aerosols during transport from the boundary layer to the free troposphere. *Geophysical Research Letters*, *39*(13), L13802. doi:10.1029/2012GL052034
- Ng, N. L., Canagaratna, M. R., Zhang, Q., Jimenez, J. L., Tian, J., Ulbrich, I. M., ... Worsnop, D. R. (2010). Organic aerosol components observed in Northern Hemispheric datasets from Aerosol Mass Spectrometry. *Atmospheric Chemistry and Physics*, *10*. doi:10.5194/acp-10-4625-2010
- O'Shea, S. J., Allen, G., Gallagher, M. W., Bauguitte, S. J.-B., Illingworth, S. M., Le Breton, M., ... Lewis, A. C. (2013). Airborne observations of trace gases over boreal Canada during BORTAS: campaign climatology, air mass analysis and enhancement ratios. *Atmospheric Chemistry and Physics*, *13*(24), 12451–12467. doi:10.5194/acp-13-12451-2013