**Anonymous Referee #1:**

General comments: The authors analyze datasets from a number of studies to examine the influence of biomass burning (BB) particles on Arctic clouds. It is a difficult undertaking, since there are not only many datasets but also many different instruments. The subject is important, for the reasons the authors discuss, and I think the authors have done a good job of estimating some potential effects of BB particles on Arctic clouds as the title describes.

*Thank you.*

That said, there are improvements needed before the paper is worthy of publication in ACP. The estimates of radiative forcing need to be clarified, as does the use of the term "background", and there is a lot of speculation made in Section 3.3 that is not substantiated by the observations and adds considerably to the length of the paper. Detailed comments follow.

*Please see our responses to the individual detailed comments below.*

Major comments:

1) The radiation forcing estimate given in the abstract, discussed on page 22844 and again in the conclusions needs clarification. On Page 22844, you say "Therefore, the -2 to -4 W $m^{-2}$ range is only applicable in the subarctic in some conditions. Nonetheless, this estimate at least provides a rough indication of how important these effects might be." Putting aside the surface albedo, is the -2 to -4 W $m^{-2}$ estimate for local effects by BB on clouds, or is it based on some anticipated coverage of the Arctic by clouds and BB plumes? Also, most of the observations were from studies conducted during springtime. Is your forcing calculated for the spring or does it include the summer too when the sun is higher and the albedo is lower? Please elaborate.

*Thanks for pointing out that this was unclear. We now add more detail and supporting information, as follows (with changes in bold):*

*Section 3.2*

Based on model output by McComiskey et al. (2008) (their Fig. 2a), we estimate that given the case study median ACI value of 0.05, the smoke-derived cloud albedo effect on **summertime local shortwave** radiative forcing could be between -2 to -4 W $m^{-2}$ for regions with surface albedo of ~0.15. **Typical shortwave spectrum broadband (0.3–5.0 μm) albedos over subarctic Canada range from ~0.09-0.17, compared to ~0.23-0.71 in the winter (Davidson and Wang, 2005); thus, any local forcing in other seasons from smoke ACI effects would likely be reduced, compared to the summer**. The McComiskey et al. (2008) output was **also** based on the assumption of homogeneous, unbroken clouds with CCN concentrations of 600 $cm^{-3}$, a LWP of 50 g $m^{-2}$, and a cloud base height of 500 m. Such surface albedo and cloud/aerosol conditions are similar to some of the summer terrestrial conditions sampled over Canada during ARCTAS-

B. The summer subarctic biomass burning clouds we describe from ARCTAS-B CCN and LWP levels bracket the model's assumptions, ranging between 1-94 g $m^{-2}$ and 68-6670 $cm^{-3}$, respectively. However, cloud base heights were typically higher than the model assumed-500 m, and although unbroken clouds are observed there, the ACI value we use was determined in a broken cloud system. Periodic broken cloud conditions, cloud heterogeneity (McComiskey et al., 2008), and the patchiness of smoke will all reduce the net cloud albedo radiative forcing over wider spaces and times. Therefore, the -2 to -4 W $m^{-2}$ range is only applicable in the subarctic in some **summertime** conditions. Nonetheless, this estimate at least provides a rough indication of how important these **local** effects might be **during the most relevant time periods (i.e., when burning is most likely to occur)**.

*Changes to abstract text are as follows:*

"Using our calculated ACI values, we estimate that the smoke-driven cloud albedo effect may decrease **local summertime** shortwave radiative flux by 2–4 W $m^{-2}$ or more under some low and homogeneous cloud cover conditions in the subarctic, although the changes should be smaller in high surface albedo regions of the Arctic."

*And changes to text in the conclusions are as follows:*

"Based on a previous model study by McComiskey et al. (2008), the ACI value of 0.05 from the case study suggests that smoke may reduce **local summertime** radiative flux via the cloud albedo effect by between 2-4 W $m^{-2}$ or more under **low and homogeneous cloud cover conditions** in the subarctic. At higher latitudes where surface albedo is already high, the impact on radiative flux is likely to be smaller."

2) Section 3.3 uses four pages and five figures to suggest that coagulation of particles associated with a clean environment might influence the hygroscopicity of BB particles by up to 10-20%. It relies on one reference (Lohmann and Leck, 2005) and later adds a second (in section 4; Lawler et al) to suggest the hygroscopicity of smaller particles in the Arctic may be relatively high. This process may be worthy of mention, but there are many things discussed in this section that are speculative without sufficient justification; I have made several specific comments about this below. The presentation related to this section needs work, and overall I feel it detracts from the main aspect of the paper already presented. This section really forms the basis for a different paper, and I think it should be treated that way or the presentation should be made much more concise.

*We have now made this section much more concise. We have reduced the text from 10 to 4 paragraphs, we have removed figures 11-13, and we have condensed figures 9 and 10 into one figure. A figure has also been removed from the associated Appendix (now Appendix A). To better substantiate our case, we have taken into account the references that the reviewer gave us in the specific*

*comments below (Leaitch et al., and Tunved et al), and have added various other references as well. To reduce speculation, we have better clarified the conditions in which the process in discussion could matter (e.g., summertime Arctic regions with dilute smoke, and subarctic areas where dilute smoke is mixing with oceanic air masses).*

*Furthermore, because the reviewer suggested making this section more cohesive with the rest of the paper, we have reframed the discussion as more of an uncertainty for the ACI value at dilute smoke concentrations. Framing the discussion in this way makes this section more seamless with the rest of the discussion. Simultaneously, it allows us to keep in the information we think is relevant to this work and it also allows us to address the portion of reviewer's comment #3 below where it was suggested that we add more discussion on diluted smoky air masses. Please see responses to the specific comments 21-25 below and the new section 3.3 and new Appendix A for more details.*

3) Use of the term "background". Page 22833, lines 17-21 – These concentrations are high, particularly the sulphate and BC values. They are not "background" values. The sulphate and BC values (<0.9 and <0.3) represent Arctic Haze. They are reference values for your BB assessment, but the use of the term 'background' is inappropriate. Page 22834, lines 1-7 – The CO levels mentioned here are clearly not background values. They too are simply reference values for BB. Values of 0.2 ppb of acetonitrile can be found over the ocean (e.g. de Gouw et al., J. Geophys. Res., 108, 2003). On page 22835, line 3, you put background in parentheses, whereas everywhere else it is not. Replacing background, everywhere used, with "reference" would suffice. Additional related comment - You appear to be mostly considering direct hits of the BB plume on the cloud. But BB plumes may disperse and dilute leaving lower concentrations of BB particles available to still influence cloud, and such influence could be relatively more significant in the long run (e.g. less impaired by competition for water vapour).

*Since there are multiple related points in this comment, we will address them individually, in a-d below:*

a) Page 22833, lines 17-21 – These concentrations are high, particularly the sulphate and BC values. They are not "background" values. The sulphate and BC values (<0.9 and <0.3) represent Arctic Haze. They are reference values for your BB assessment, but the use of the term 'background' is inappropriate.

*Thanks for pointing out that mistake - there was actually a typo here. The values for $SO_4^{2-}$ and BC should have been listed at 0.3 $\mu g\ m^{-3}$ and 0.12 $\mu g\ C\ m^{-3}$, respectively. Although the $SO_4^{2-}$ background cloud data weren't shown in the original ACPD paper, if one were to back-calculate from the ln(BC) data presented in Figures 6 and 7, one can see that that BC data never rose above this point in "background" clouds.*

b) Page 22834, lines 1-7 – The CO levels mentioned here are clearly not background values. They too are simply reference values for BB.

*Here we disagree. Before explaining why, we first just to provide a little more clarification on our methods. The CO, $CH_3CN$, BC, and $SO_4^{2-}$ data were only used for classifying "background" air masses in ARCTAS – as stated in the text, in all other campaigns a "background" classification was obtained by using a $CN_{PCASP}$ concentration of <127 particles $cm^{-3}$. We feel fairly confident that this non-ARCTAS particle concentration cutoff really does indicate background conditions, as based on the information in Table 6, which shows that ARCTAS pollutant levels in air masses that satisfy this criterion (based on $CN_{PCASP}$ equivalent data) are all well below literature reported "background" concentrations for the Arctic. Note also that the values of $SO_4$ < 0.3 μg $m^{-3}$ and BC < 0.12 μg C $m^{-3}$ we used for ARCTAS data are also well below literature reported concentrations – again, our apologies for the typo previously. Secondly, we also wanted to mention that within ARCTAS, "background" air was not based solely on $SO_4$ and BC values. To be classified as background, the air masses also at the same time had to have CO concentrations < 123 ppbv and $CH_3CN$ levels < 0.14 ppbv.*

*Regarding the references listed on Page 22834, lines 1-7 (Lathem et al. (2013), Moore et al. (2011), and Lance et al. (2011)), CO cutoffs of 160-170 ppbv were used along with $CH_3CN$ cutoffs of 0.1 ppbv. In these publications, these cutoff values were not used by the authors of those studies merely as reference values for comparison to polluted cases, but specifically as classification criteria for "background" or "clean" air masses. To clarify why the authors considered these values "background" and to provide more information on why we chose the 123 ppbv CO value for our threshold, we now add the following text:*

*"*For comparison, Lathem et al. (2013) and Moore et al. (2011) defined background air masses as having CO and $CH_3CN$ values at <170 ppbv and 0.1 ppbv, respectively, and Lance et al. (2011) used a criterion of ~160 ppbv CO. **Such high background CO values are observed periodically over springtime Alaska due to higher emissions from Asia during spring and reduced photochemical loss during winter months (Brock et al., 2011). In 2008 specifically (during a similar time period as ARCTAS-A), background CO was elevated further due to unusually early and frequent Asian wildfires that year (Moore et al., 2011). However, background Arctic CO levels can frequently be lower than these values. For example, during a separate summer campaign in 2011 over eastern Canada, Sakamoto et al. (2015) observed and used a lower background CO threshold of 120 ppbv. Our chosen CO threshold of 123 ppbv, was chosen in part because it enabled the use of a consistent value to characterize background conditions across the wide temporal and spatial region covered during ARCTAS.***"**

*To determine background conditions, we not only used a CO cutoff of 123 ppbv, but we also combined the gaseous tracer criteria (CO and $CH_3CN$) with a complementary combination of aerosol tracer criteria ($SO_4$ and BC), making our ARCTAS classification of "background" air as or more rigorous than any other*

*similar study for this region that we are aware of. For the various reasons listed above, we believe the term "background" is appropriate, and have kept it in.*

c) Values of 0.2 ppb of acetonitrile can be found over the ocean (e.g. de Gouw et al., J. Geophys. Res., 108, 2003).

*Values of $CH_3CN$ up to 0.2 ppbv have been observed over the ocean in some mid-latitude locations such as in the de Gouw et al. study mentioned above. However, in the Arctic, observations suggest that the range of background acetonitrile data is closer to ~0.050 ppb in the marine boundary layer to < 0.140 ppb in the free troposphere (e.g., Warneke et al. (2009); Kupiszewskiet al., (2013); A. Wisthaler, unpublished data). For the reader's reference, the Kupiszewskiet al. and Warneke et al. references have been added into the discussion of background $CH_3CN$ as follows:*

"Although for simplicity we define a single background Arctic $CH_3CN$ level here, background $CH_3CN$ can range from ~0.050 ppbv in the Arctic marine boundary layer to ~0.1**4** ppbv at altitudes of ~8 km (**Kupiszewski et al., 2013; Warneke et al., 2009;** A. Wisthaler, personal communication, 2015)."

*However, even in the unlikely case that Arctic background $CH_3CN$ levels ever did reach levels as high as 0.2 ppbv, we do not believe it would affect any of our results in a meaningful way. For a background classification, it only means that we are being more conservative in our cutoff when we say nothing with $CH_3CN$ values > 0.14 ppbv can be classified as background. For smoke cases, an ARCTAS $CH_3CN$ value of 0.2 ppbv alone was not enough for a smoke classification; CO, $SO_4$, and BC also had to be elevated (in ISDAC criteria were based on SPLAT II particle chemical composition). We now add a line to section 2.4 mentioning this to the reader, with new text in bold.*

"ARCTAS "biomass burning" influenced air masses were classified following the procedure of Lathem et al. (2013), where BB-influenced air masses have concentrations of >175 ppbv and 0.2 ppbv CO and $CH_3CN$, respectively. **A manual scan indicated that aerosol pollutant tracers BC and submicron $SO_4^{2-}$ were always elevated with respect to background concentrations under these conditions in this dataset**."

d) You appear to be mostly considering direct hits of the BB plume on the cloud. But BB plumes may disperse and dilute leaving lower concentrations of BB particles available to still influence cloud, and such influence could be relatively more significant in the long run (e.g. less impaired by competition for water vapour).

*That is a good point. In the multi-campaign analysis, it was unfortunately not possible to include any intermediate conditions between BB and background end points because we did not have a good way to ensure moderately low particle concentrations were due to smoke and not some other aerosol source. In the case study we only had 3 intermediate points between smoky conditions and*

*background conditions, and so we did not try to draw conclusions from those points alone.*

*This uncertainty from competition for water has already been mentioned in the last paragraph of methods section 2.5, and we touch on it again in new text in the discussion (please also see our response to comment #18). In addition to that discussion, we have now rephrased section 3.3 so that it touches closely upon uncertainties in diluted clouds caused by potential enhancements in volume/changes in hygroscopicity from background particles mixing with smoke. We also bring up this uncertainty in the last sentence of the concluding section 4:*

*"***Future remote sensing or ground-based analyses may be able to more completely address the different impacts of dilute vs. concentrated smoke aerosols in Arctic clouds**."

4) Page 22830-22831, first two paragraphs of section 2.2.2 – There is very little about the qualities of either the CAPS-CAS and the FSSP-100 droplet measurements. The FSSP-100 measurements are at least discussed relative to other independent measurements (LWC from hot-wire), but it seems that the CAPS-CAS observations are assumed to be of high quality without any demonstration of such. Based on the LWC and N(liq) in Table 8, the mean size of the volume weighted distribution varies between about 5 um diameter to 3.5 um diameter, which means that about half of the LWC and most of the droplet numbers are below those diameters. How accurate was the CAPS- CAS in 2001, when the measurements were made, at measuring droplets below 5 um diameter? What are the consequences if those measurements are of relatively poor accuracy?

*We now add more information on the quality of the ARCTAS LWC and size-distribution data in the new text below:*

"LWC was estimated from the CAPS-CAS probe based on integrated volume droplet size distributions between 0.75-50 μm. Throughout this size range, precision was estimated to be 20% within each size bin based on pre-calibrations with sized glass and polystyrene latex spheres. We expect accuracy to also be ~20%, since pre-campaign calibrations were performed with spheres of known size, and since post-campaign tests with latex spheres were consistent with the expected sizes. Unfortunately, we could not validate in situ accuracy because simultaneously collected hot-wire probe LWC data were unobtainable due to high noise in out-of-cloud samples. For this reason, in-cloud hot-wire LWC data are not reported here other than to note that they showed qualitatively consistent trends with the CAPS-CAS LWC data."

*We have three sidenotes that pertain to the ARCTAS LWC data as well:*

*1) Specifically with regards to the ARCTAS case study ACI values, accuracy is less important than precision because the measurement relies on differences between smoky vs. background clouds. Where accuracy (vs. precision) would be*

*most important for ARCTAS data is in their inclusion into the multi-campaign analysis, where we were comparing across different campaigns that used different instruments. It would also matter where absolute $r_e$ values were listed (although again here, the focus of the discussion was oriented towards differences between two groups, rather than on the absolute values of the groups). For now, we have not specifically mentioned any uncertainty in the accuracy of absolute $r_e$ sizes or in the multi-campaign ACI value from ARCTAS values because post-campaign analyses were consistent with expected values.*

*2) Just as a minor note to clarify the record, ARCTAS data were taken from 2008, not 2001.*

*Since the reviewer also mentioned the FSSP data, we would also like to bring their attention to some new text regarding the FSSP data. This information has been added in response to reviewer #2, who wanted more information on why the FSSP values were lower than hot-wire probe values in the FIRE.ACE campaigns. In response to that reviewer, we have now changed some of the data input data (now just focusing on the time periods relevant to this study, and not the whole campaign). Doing so allows the stronger relationship with coincident hot-wire probe values during the time periods relevant to this study to become apparent (see the new Table 5). For the NRC FIRE.ACE data, we also now use a different FSSP data source, which after some additional analysis (see response to reviewer 2's question 2), we believe to be a more reliable data source. The use of this new data source has improved the correlation with the hot-wire probe data. New text with more information on the FSSP measurements has been added into section 2.2.2, as follows:*

*"*During the **UW and NRC** FIRE.ACE campaigns, LWC was determined from droplet size spectra gathered from Forward Scattering Spectrometer Probe (FSSP-100) measurements for particles with diameters between 0.5-47 µm **and 5-47 µm**, respectively. These measurements are functionally very similar to the CAPS CAS measurements from ARCTAS. **During the sampling periods where air mass classification matched the criteria described in section 2.4, the** FSSP data had a close relationship to hot-wire probe measurements of LWC **for both campaigns (Table 5). For the NRC FIRE.ACE campaign, two FSSP probes were available (serial numbers 96 and 124, denoted hereafter as FSSP-96 and FSSP-124). The FSSP-96 is normally recommended for use by the data originators because the FSSP-124 had an intermittent hardware problem during the NRC FIRE.ACE campaign, and because it may have undersized particles >30 µm diameter. In this analysis, the hardware problem did not occur during our time periods of interest, and the FSSP-124 droplet distribution for droplets with diameters within 30-47 µm closely matched those of the FSSP-96. However, the FSSP-124 had higher droplet numbers in particles with diameters < 30 µm compared to the FSSP-96 during the relevant sampling periods used in this study. We believe this discrepancy to be due to a deficiency in the FSSP-96**

**data during this time period, because the FSSP-96 underestimated King and Nevzorov probe LWCs by ~23% and 26%, respectively, whereas the FSSP-124 data estimated King and Nevzorov probe data to within 8%, on average (Table 5). Therefore, the FSSP size distribution data reported here for the NRC FIRE.ACE campaign are based on FSSP-124 data between 5-47 µm.**"

*The figures and information in the text have been corrected accordingly, but the impact on the results is very small, because there were only 2 distinct cloud cases that matched our background criteria in the NRC FIRE.ACE study. For more information, please also see our response to reviewer #2 (their question #2).*

Minor comments:

5) Page 22831 - A comment on potential artifacts from droplet shattering on the probe tips (e.g. Korolev et al., B. Am. Meteorol. Soc., 92, 967–973, 2011). The reference is for ice crystals, but very large droplets may also shatter creating artifact droplets. It is likely a non-issue for the mostly smaller droplets you measure, but could be important for some of the reference measurements.

*Thanks for this comment and reference. We have added the following text in section 2.2.2:*

"Note that similarly to ice particles (e.g., Korolev et al. (2011)), very large droplets may shatter on any of the cloud droplet probe tips. This may introduce some potential artifacts when droplet sizes are very large (e.g., for some of the reference measurements available in FIRE.ACE and ISDAC)."

6) Page 22832, lines 19-22 - Understandable, but the horizontal extent of a cloud and the number of times it will be sampled by an aircraft may be related: it is a tendency in these studies to sample clouds of greater horizontal extent more than smaller clouds. Since larger clouds will have a greater radiative impact, should they not be considered more than smaller clouds? It might be different consideration if you were examining a process only, but you are considering an impact here. Does your approach potentially bias the impact lower?

*That is an interesting question. The reviewer refers to the following passage:*

"In some instances in the multiple-campaign analysis, the same cloud or very similar clouds were sampled more than once, often intentionally, either through an entire vertical cloud transect or through a portion of a cloud. In order to reduce the potential for pseudo-replication in the analysis, transects that were deemed to be from the same cloud or from very similar clouds were averaged to provide one aggregated profile or $r_e$ and $N_{liq}$ value for those instances."

*The reviewer is correct that clouds with more transects did tend to be horizontally larger clouds in this study.*

*However, there are two problems with weighing certain clouds more than others. First, we don't have a way to accurately quantify cloud horizontal extent, which makes weighting complicated (see (a) below for more detail and discussion). We could possibly remove cumuliform-type clouds from the analysis, and estimate the ACI in stratiform-only clouds only, but it would result in a significant loss in sample size.*

*The second problem is that each individual cloud is impacted by some unknown meteorological component; if an individual cloud point were weighted more, the confounding meteorological component would be as well. But if each individual cloud is weighted equally, any non-representative meteorological factors that might skew one cloud are less likely to skew the whole dataset, even if that one cloud had many data points sampled within it. For the reasons above, we believe that attempting to weigh clouds with greater horizontal extent more than other clouds would likely increase rather than decrease the uncertainties in our analysis.*

*Finally, regarding whether or not our approach might bias the results lower, we would like to re-clarify that in this study we estimated what the local radiative fluxes would be in homogeneous cloud cover conditions, as based on our collective ACI value and results from the McComiskey et al. (2008) model. Therefore, in this method, smaller clouds were not being considered more for the radiative flux than larger clouds, per se. However, it is true that not all the individual cloud points used to derive the ACI value covered the surface homogeneously. Therefore, we cannot rule out the possibility that patchy clouds or cumuliform clouds might have had different microphysical responses to aerosols, potentially biasing the ACI values (and thus flux estimates) either higher or lower by their inclusion. We have modified the text in section 3.2 of the manuscript to hopefully better clarify this uncertainty:*

*"*The McComiskey et al. (2008) output was also based on the assumption of homogeneous, unbroken clouds…. However, … although unbroken clouds are **frequently** observed **in the Arctic and subarctic**, the ACI value we use was determined **from samples that included some clouds within** broken cloud system**s, which may possibly have different microphysical responses to aerosols**. Periodic broken cloud conditions, cloud heterogeneity (McComiskey et al., 2008), and the patchiness of smoke will all reduce the net cloud albedo radiative forcing over wider spaces and times.*"*

*(a) If we wanted to somehow take horizontal aerial extent into account, we would need a way to quantify it, and this would be a very difficult thing to do. Video was only available for ARCTAS, and while some combination of flight notes and photos were available for the other campaigns, they offer only incomplete information on cloud size. Meanwhile, many of the clouds sampled were unobservable from remote sensing data because they were either present under another cloud layer and/or there were no available data*

*at that time for a variety of reasons. From a combination of flight notes, photos, and video, we were able to determine in nearly all instances whether a cloud was stratiform or cumuliform, but were only able to estimate cloud sizes from ARCTAS during the case study, when the entire cloud was small enough to be fully observable by video prior to sampling (and even then sizing was difficult due to the amorphous 3-D structure of the clouds). Without information on cloud horizontal extent, we cannot weight clouds by this information, and do not trust that transect number is an accurate reflection of cloud horizontal extent.*

7) Page 22833, lines 8-15 - Do the LWCs relate more to Re or N(liq), which may tell you something about the mixing processes?

*This is an interesting idea, but unfortunately we had limited data for testing the influence of homogeneous vs. heterogeneous entrainment in most cases. Only 2 clouds in the multi-campaign analysis had more than 2 transect aggregates with which to make correlation plots, and even in these 2 cases sample size was low (n=4 and n=8). In the n=8 smoke case mentioned in the text, there was a closer relationship with $r_e$ than $N_{liq}$, which does suggest that entrainment might have had an influence on this particular cloud. The text has been changed as follows:*

"Within the multi-campaign analysis, 2 of the 8 biomass burning clouds contained aggregated transects, as did 4 of the 16 background clouds. One background cloud in the case study included aggregated transects. To assess the impact of cloud transect aggregation on our analysis, we calculated differences in ACI values using the maximum and minimum values of $N_d$ within the aggregated samples. Calculated differences in ACI values were 1%, indicating that uncertainties caused by aggregation had only minor impacts on our results.

LWC among aggregated clouds was generally similar (within 30% of each other). **However**, in some cases it was more variable**;** in one biomass burning aggregation, the set of 8 related cloud transects had LWCs ranging **from** 0.12-0.54 g $m^{-3}$**. The relationship of LWC with $r_e$ suggests that entrainment could have influenced LWC variability within this particular cloud. Although we cannot constrain the influence of entrainment to a high degree of certainty within an individual cloud aggregate, as discussed in section 3.1, the ACI values derived across all clouds did not deviate from adiabatic values calculated from cloud parcel theory.**"

8) Page 28334, line 8 – Here, do you mean high-quality or high-resolution?

*The reviewer refers to the following text:*

"During the two FIRE.ACE campaigns, high quality aircraft chemical data for completely characterizing air mass sources were not collected…."

*We will change "*high-quality*" to "*the combination of relevant high-quality

and/or high-resolution*" here. There may have been some poor quality CO data and major ion data taken during the UW FIRE.ACE campaign, but after speaking with some of the people present on those flights, the first author was told that the data either didn't actually exist of were of poor quality since a chemist was not onboard the flights to ensure quality (a lot has been forgotten since the campaign took place 17 years ago, and the PI has since passed away). On the NRC FIRE.ACE campaign, mercury, ozone, and $SO_2$ data were taken, but these tracers were not useful alone in determining with confidence whether a sample with high particle number was primarily impacted by biomass burning or some other aerosol source. Major ion and $CHBr_3$ data were also collected, but at low temporal resolution. We are not aware of any other relevant high-quality/high-resolution chemical data collected during either campaign.*

9) Page 22835, line 15 – In the literature, there tends to be a generic use of the term Aerosol-Cloud Interactions that pervades the indirect effect. Are you not just assessing the effect of the BB aerosol on cloud? Is there an interactive aspect implicit in what you are assessing here? You do not deal with deposition resulting from precipitation altered by the aerosol in a meaningful way, other than to mention it at the bottom of page 22849. A few words of clarification would be helpful.

*Although we did not focus on the implications of our results for precipitation as much as we did on their potential radiative impacts, we chose to use the term ACI because it is frequently used in the literature to describe our method. However, we acknowledge that the term may be a source of confusion because the recent IPCC calls ACI the full link between aerosols and climate forcing, and ACI is also sometimes called the "Aerosol Cloud Index". Therefore, we now specify again in the text that ACI is defined by equation 1.*

*We also try to better explain why "ACI" is used instead of the term "indirect effect" by adding in the following text:*

One common way in which aerosol-cloud interactions (ACI) are quantified is by assessing how a cloud property changes relative to some aerosol tracer or, in this case, biomass burning aerosol tracer ($BB_t$). **The ACI term as defined by Eq. (1) was originally labeled the "Indirect Effect" (IE) (Feingold et al., 2001, 2003). Here, similarly to McComiskey et al. (2009), we use "ACI" instead of "IE" to differentiate the fact that the metric in Eq. 1 is more directly associated with aerosol-driven changes to cloud microphysics than with radiative forcing.**"

10) Page 22835, on line 26, you refer to CCN, which is not defined anywhere previously, including the abstract where it is mentioned as CCN. Please define it in the abstract.

*Done.*

On line 28, background values of 0.018 are referred to as being subtracted. What are the units and are you referring to CO or $CH_3CN$ or something else?

*We fixed the sentence so that now it is clear we meant 0.018 ppbv for $CH_3CN$.*

11) Page 22836, lines 21-24 - Both the UHSAS and the APS use sheath air to focus the particles for detection. The sheath air is normally dried and that can also help with the drying of the particles prior to detection.

*We now include this information:*

"UHSAS and APS measurements are not actively dried like PCASP samples are (Earle et al., 2011; Strapp et al., 1992), but sample humidity decreases significantly upon heating in the cabin and measurements are taken at dry relative humidity**; in addition, particles are exposed to dried sheath air prior to detection**."

12) Page 22837, lines 5-6 – Would you please clarify how this uncertainty can be "fully eliminated in model simulations"? It reads to me as if we don't need observations, since the model can solve the problem.

*Thanks for pointing that out – it certainly wasn't our intention to imply that observations were not necessary. Even modelers would likely agree that the in-situ data are vital because they are the most exact microphysical measurements available for model evaluation. The sentence has been rephrased from:*

"A third potential problem is the risk that a snapshot of a cloud in time is not representative of the net cloud properties over its lifetime (Duong et al., 2011). **This source of sampling error can only be fully eliminated in model simulations,** and it is best minimized in aircraft in situ data by resampling throughout the cloud's life cycle."

*to:*

"A third potential problem is the risk that a snapshot of a cloud in time is not representative of the net cloud properties over its lifetime (Duong et al., 2011). **Currently, only models can fully characterize cloud lifetime properties, but interpreting the model output can be challenging for other reasons. Within aircraft in situ data,** this source of sampling error is best minimized by resampling throughout the cloud's life cycle."

13) Page 22837, line 15 and 17 – insert "e.g." in front of these references, here and elsewhere (22840). The competition process was demonstrated 30 years ago.

*Done.*

14) Page 22838, lines 10-11 – It seems odd that there were no inversions topping the clouds. Even in the typically stable environment of the Arctic, the layers will be defined by slight inversions. How were they contained?

*We now rephrase:*

"With one exception (an ARCTAS-B background case from 8 July 2008), the stratiform clouds were not present below a **strong** temperature or moisture inversion."

15) Page 22838, line 19 – It is surprising to see CO up to 500 ppbv classified as out- of-plume, when the previous discussion referred to much lower values of CO as the reference for non-BB. What was the basis for identifying the plume?

*We think the reviewer is probably referring to page 22839, line 19? If so, the problem here was probably poor wording on our part. We were just trying to show non-extreme CO values here and not background concentrations, specifically. For clarity, we have changed the text from:*

*"*In Fig. 3, **we show that out-of-plume CO (CO < 500 ppbv)** is strongly related to the smoke tracer $CH_3CN$ and that it shows no correlation to the fossil fuel combustion tracer dichloromethane ($CH_2Cl_2$)…."

*To:*

*"*In Fig. 3, **we show that CO < 500 ppbv** is strongly related to the smoke tracer $CH_3CN$ and that it shows no correlation to the fossil fuel combustion tracer dichloromethane ($CH_2Cl_2$)…."

16) Page 22843, line 6 – Is ice "typically well mixed throughout" during the summer?

*We have taken this sentence out:*

*"*As noted previously, because the aircraft could only sample transects of clouds, we had to assume that the observed cloud phase was representative of the whole cloud. In the case study, all clouds were sampled at temperatures > 0 $^{o}$C, and this assumption holds well. ~~In Arctic stratocumulus clouds, ice is typically well mixed throughout (McFarquhar et al. 2007, 2011).~~ Where we expect this assumption to be most uncertain is in stratiform clouds in the multi-campaign analysis, which might have different properties in far-off, non-sampled portions. Uncertainties are also higher in clouds that were only transected horizontally, because mixed phase clouds in the Arctic frequently have vertical layers of ice and liquid particles (Morrison et al., 2012)."

17) Page 22844, line 22 – coagulation is usually a term associated with aerosol particles, whereas cloud processes refer to collision-coalescence.

*New text reads:*

"This narrowing is likely to lessen the eventual probability of **precipitation** (Tao et al., 2012), as is moves median droplet size further away from the 28 μm

effective diameter threshold at which collision-**coalescence** processes are thought to become efficient enough to induce precipitation (Rosenfeld et al., 2012)."

18) Page 22848-22849 – Can you briefly discuss how does CCN number vs. CCN hygroscopicity plays into the impact of BB on the ACI index?

*New text has been added into the conclusions, as suggested:*

"For comparison to the multi-campaign analysis, we also analyzed the 1 July 2008 ARCTAS case in the subarctic, where multiple clean and smoky clouds were found under similar meteorological conditions. **The case study smoke cases had a** combination of low cloud LWC**,** high **in-plume** aerosol concentrations**, and** very small cloud droplets. **From these samples we** derived an ACI estimate of 0.05 (95% confidence interval 0.04-0.06), **which is smaller than that of the multi-campaign analysis**. Based on **theory (e.g.,** Moore et al. (2013)**), as the number of smoke CCN increases (through some combination of enhanced aerosol number and/or increased hygroscopicity for existing particles), there is greater water vapor competition. This competition makes supersaturation development and cloud droplet activation increasingly difficult, which would reduce ACI values. Therefore,** we speculate that the 0.05 ACI case study value falls at the low-end of typical smoke ACI values for the larger subarctic/Arctic region. **Reductions in droplet activation and potential enhanced evaporation** would **also** limit the maximum magnitude of smoke cloud albedo effects."

19) Page 22850, lines 20-21 – sulphates are not necessarily "an additional organic component".

*Thanks for pointing out that typo. We have changed the text from:*

"Interestingly, previous studies have indicated that Arctic smoke aerosols also sometimes contain an additional organic component likely to be derived from smaller, non-biomass burning particles **such as** sulphates and marine particles (Earle et al., 2011; Zelenyuk et al., 2010)."

*To:*

"Interestingly, previous studies have indicated that Arctic smoke aerosols also sometimes contain an additional organic component likely to be derived from smaller, non-biomass burning particles **mixed with** sulphates and marine particles (Earle et al., 2011; Zelenyuk et al., 2010)."

20) Page 22851, line 1 – change "condensation of external particles onto" to "coagulation of external particles with".

*Done.*

Specific comments related to Section 3.3

21) Page 22845, lines 11-13 – Please add to your references: Leaitch et al., Elementa, (2013) and Tunved et al., ACP, (2013).

*Done, and an additional reference, O'Dowd et al. (2010) GRL, has also been added. Thanks, we had not previously been aware of those relevant references and they were very helpful in restructuring this section.*

22) Page 22845, lines 16-20 – During the time of Arctic Haze influence, 1) there are generally few particles smaller than about 80 nm, and 2) the presence of the larger particles inhibits the formation of smaller particles. So when the aerosol is dominated by Arctic Haze or BB influences, the "small background aerosols" are not directly significant for liquid cloud formation. However, during the summer, the air is quite clean and there is potential for such small particles to be important for clouds (e.g. Leaitch et al., Elementa, 2013). Please do not generalize here.

*The entire section has been substantially re-written, and the specific text the reviewer refers to has not been included. However, we have made an effort to better clarify the conditions in which the process in discussion could matter by adding in the following in the new text:*

"It is important to note that these small background particles are not ubiquitous throughout the year. They tend to accumulate mainly in the spring and summer, which is thought to be due to a combination of three reasons: 1) there is more sunlight available for the photochemical reactions key to new particle formation (Engvall et al., 2008; Tunved et al., 2013), 2) reduced sea ice and enhanced primary production likely lead to greater emissions of marine precursor gases and nanogels (Leaitch et al., 2013; O'Dowd et al., 2010; Tunved et al., 2013), and 3) during Arctic summer there tend to be fewer larger particles like smoke for these small particles to coagulate and condense upon. However, Arctic summertime smoke events do occur (e.g., Fuelberg et al. (2010); Iziomon et al. (2006)) and may be increasing (Moritz et al., 2012). In subarctic regions, wildfires actually peak in the summer (Giglio et al., 2006). Thus, while the influence of the small background particles on subarctic and Arctic smoke ACI values is probably fairly minor, it is possible that deviations from the linear ACI expectations derived here might occur during dilute summertime Arctic smoke events and in subarctic locations, especially where diluted smoke mixes over or near marine environments."

23) Page 22846, line 3 – Why do you use backscatter here instead of total volumetric scatter? The relative backscatter is higher for smaller particles, but their total scatter is generally smaller reducing sensitivity to them. What is the detection limit for the backscatter observations?

*As suggested, we now use submicron scatter (total scatter was not available in*

*ISDAC). This change does not substantially affect our results in any way. Note that because it was requested that we reduce space in section 3.3, this figure has been combined with Figure 10 and the ARCTAS-A data have been removed. Also, to show the full dataset more clearly we have now plotted the data on a log-log plot (see new figure 9). Detection limits are listed in Table 1.*

24) Page 22847 – The discussion of the rapid change in CN is hampered by 1) the absence of a discussion of the possibility of new particle formation (NPF) aided by a sharp reduction in the condensation sink (as indicated by the APS and OA; the backscatter observations appear to have a delayed response relative to the OA), 2) the failure to plot the data as vertical profiles rather than time series. It is difficult to understand from the time series the regions of mixing/transition region(s) in which the coagulation is apparently taking place. If you must retain this discussion, please make it easier for the reader by plotting the data as vertical profiles. The explanations that "Such a rapid change in CN(TSI) concentrations could be explained by either a sharp non-mixing transition zone or by rapid coagulation of the small particles onto the larger haze particles" seems to avoid the possibility that NPF associated with a small condensation sink may explain the rapid increase in CN. Certainly small particles will coagulate with larger particles if present together, but it seems that these layers are relatively de-coupled and that the higher CN concentrations after 69500 are more likely to be the result of NPF in very clean air.

*As suggested, this portion of the text has been taken out. It has been replaced with other references that better demonstrate the possibility of coagulation.*

25) Page 22848, lines 5-21 - Were there any CCN measurements of the BB particles that would suggest larger hygroscopicities (kappa values) than expected for a "pure" BB aerosol, exclusive of sulphate? How important an influence on the hygroscopicity would this coagulation be relative to the smaller amounts of sulphate found in the BB particles? You mention sulphate in Section 4, but not here.

*We now add the following text in bold.*

"Interestingly, the small marine particles appear to be fairly hygroscopic (Lathem et al., 2013; Lawler et al., 2014; Zhou et al., 2001), and they can be surface active (Lohmann and Leck, 2005). **One study using ARCTAS data showed that background aerosol values of the hygroscopicity parameter, κ, were on average nearly two times higher than average smoke κ values (0.32 ± 0.21 vs. 0.18 ± 0.13, respectively), although there was a high degree of variability and overlap in the κ values (Lathem et al., 2013)."**

*Regarding sulphates, we now add the passage the reviewer referred to in their comment above into the new section 3.3:*

"Previous studies also suggest that the small particles can condense upon larger particles (e.g., smoke) when such particles are present (Leaitch et al., 2013; Tunved et al., 2013). This coagulation process may explain why **Arctic smoke aerosols have been**

**shown to sometimes contain organic components likely to be derived from smaller, non-biomass burning particles mixed with sulphates and marine particles (Earle et al., 2011; Zelenyuk et al., 2010).**"