Aircraft-measured indirect cloud effects from biomass burning smoke in the Arctic and subarctic

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Dear Dr. Lynn Russell, editor:

Below, please see comments on the manuscript: "Aircraft-measured indirect cloud effects from biomass burning smoke in the Arctic and subarctic" by L.M. Zamora et al.

We appreciate the thoughtful and constructive comments from the reviewers. Their helpful suggestions and attention to detail have made this a substantially better paper, and we greatly appreciate all the time they put into the manuscript.

Following the reviewer's suggestions below, in numerous places we have now better clarified the text throughout the paper. Also following a referee suggestion, we have substantially reduced the size of section 3.3. Text in this section is down from 10 to 4 paragraphs. Additionally, we have removed figures 11-13, we have condensed figures 9 and 10 into one figure, and a figure has been removed from the associated Appendix (now Appendix A). We have also added in more information to substantiate this section and to make it more cohesive with the rest of the paper.

While developing responses to the reviewers, we also noticed and corrected a few errors in our underlying data from the previous analysis, which resulted in some minor updates in the new manuscript. These corrections are listed below:

1) The largest change to our results resulted from fixing a bug in the code used to derive the ARCTAS data presented in Figs. 5, 7, and 8 and in Tables 7 and 8. Due to this bug, we had previously used the ARCTAS 10-second averaged size distribution spectra for these figures/tables, rather than the 1-second spectra as intended. After correcting this error, and in combination with the other changes listed below, our new ACI value in the multi-campaign analysis has increased from 0.12 to 0.16, and the ACI value in the case study decreased slightly from 0.06 to 0.05. There were minor changes in the cloud droplet effective radii as well. These changes had only a small effect on our overall conclusions.
2) The two ISDAC smoky cloud instances were accidentally missing in the map in Figure 4 and in Table 7. These data have now been included in our results and the text has been updated. Their addition had only very minor impacts our results, and has not changed any of our conclusions.
3) While preparing responses to the reviewers, it was discovered that one of the background clouds in the case study was actually likely to be from the same cloud as one of the other background cases. Now these two cases have been merged into one cloud case. Again, this change had only very minor impacts on the results and did not change our conclusions.
4) The FSSP data from the NRC FIRE.ACE campaign have been modified slightly to increase data quality in response to reviewer 2's question 2. There were only two cloud cases from this campaign, and this change also did not have a noticeable impact on our results.

We now hope that the manuscript in its revised form will be acceptable for publication in "Atmospheric Chemistry and Physics." Below, we have given our responses in italics following the reviewer comments.

Reviewer Comments and Our Responses

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 μg m^{-3} and 0.12 μg 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).**″

Anonymous Referee #2

This paper uses in situ aircraft data on cloud properties from a variety of field campaigns in the Arctic and subarctic (ARCTAS, ISCCP, FIRE.ACE, and ISDAC) to determine the magnitude of subarctic and Arctic smoke aerosol-cloud interactions (ACI). Averaging the data over all campaigns gave an estimated ACI of ~0.12 (out of a maximum of 0.33). The data also included a subarctic case study from ARCTAS that included clean and smoke-polluted clouds in similar geographic areas and meteorological conditions. In this case study, the estimated ACI was 0.06. The authors explain the lower value in the case study as a result of the low liquid water content (LWC) of the clouds and the high aerosol concentrations, which would result in limited formation of droplets relative to the adiabatic value. They note that these ACI values could decrease short-wave radiative flux by 2-4 W m^{-2} or more under some low and homogeneous cloud conditions in the Arctic. The authors also show evidence that numerous background Aitken mode particles may interact with combustion particles, altering their properties.

General comments: This is a well-written paper on an important problem in climate science. The work appears to have been planned and performed well and the conclusions are generally supported by the evidence. I have some minor concerns that I have listed below that I would like to see addressed, but overall I recommend publication of the paper after these minor revisions.

Thank you.

Minor Comments:

1) P22825, L20-23: In this conclusion, the word "some" in "some low and homogeneous cloud conditions" is doing a lot of work. The text (P22843, L19-28 and P22844, L1- 7) makes clear that this 2 to 4 W m^{-2} estimate is only valid for a specific type of low, homogenous cloud layer over surfaces with an albedo of ~0.15. Given the limited applicability of this estimate of the impact, saying in the abstract and conclusions (P22849, LL21-25) that the impact is 2 to 4 W m^{-2} "or more" is misleading. The abstract and conclusions should make clear that this is not an appropriate value to assume for a regional impact, rather just an estimate of the impact under a very specific, but reasonable, set of subarctic conditions.

Thanks for pointing that out. Reviewer 1 also had a very similar comment (their comment #1). We have now tried to be more specific, and have added 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 between 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."

We also just wanted to clarify why we used the phrasing of -2 to -4 W m^{-2} "or more" since the reviewer mentioned that phrasing in their comment. Due to the non-representative cloud conditions in the case study, we believe that the ACI value of 0.05 used to derive the estimate of -2 to -4 Wm^{-2} is on the low-end of typical smoke ACI values for the greater Arctic/subarctic region. This hypothesis is stated in section 4, and is based on the information discussed in section 3.1. If we use the ACI value of 0.16 from the multi-campaign analysis instead of the 0.05 value from the case study, based on the McComiskey et al. model, the estimated change in local radiative flux would be larger (around -10 W m^{-2}). Therefore,

although we used the lower range of -2 to -4 W m^{-2} in the paper in order to be conservative, we felt the term "or more" was merited and important for the reasons stated above.

2) P22830, L21: Can you explain why the FSSP data were lower than the hot-wire probe measurements of LWC?

For the UW FIRE.ACE campaign, we now take only the data relevant to the days during which clouds were sampled in this study (as opposed to data representative of the whole campaign, which we had done before). Doing so now reduces the differences between the FSSP LWC and the hot-wire probe LWC from 16% to 8%. The data presented now are more representative of the data quality specific to the cases presented in this study.

The NRC FIRE.ACE campaign had a larger discrepancy with hot-wire LWC values than the UW FIRE.ACE campaign. We do not believe the discrepancy is due to deadtime/coincidence, which were corrected for (Baumgardner et al., 1985; Dye and Baumgardner, 1984). Icing and fogging of the FSSP probe are also not likely sources for the discrepancies because: a) according to the flight notes, periods were nulled out when the FSSP was known to be iced or fogged, b) we only looked at liquid phase clouds in this study, which reduced the risk of icing-related problems, c) we observed no significant differences in instrument performance in mixed vs. liquid phase clouds (phase determined by CPI data), and d) the difference between the FSSP and the King LWC observations were consistent within days and among days for nearly all of the campaign, which would be a counter-indication of fogging because we would not expect fogging to be so consistent.

Another possible reason for reduced LWCs as compared to the hot-wire probe data in the NRC FIRE.ACE campaign is that the FSSP used here (the FSSP serial number 96, or FSSP-96) was undersizing large particles. Based on a April 10 calibration in the middle of the NRC FIRE.ACE campaign, the FSSP-96 may have undersized particles with diameters > 30 microns by up to 20%. To test this, we looked at the 2 background cases from the NRC FIRE.ACE campaign. In one of these clouds we actually observed a noticeable fraction of particles with diameters greater than 30 μm in the CPI data whereas the other had smaller and more consistently-sized particles. However, when we analyzed how well the FSSP approximated King LWC values in the cloud with large droplets, it did not perform significantly worse than in the other cloud. In fact, it approximated King hot-wire values slightly better (slope = 0.73 vs 0.68, R^2= 0.93 vs. 0.95, n=865 vs. 81). That finding does not support the hypothesis that we were undersizing large droplets.

More information can be obtained if we compare the FSSP-96 probe with another FSSP (serial number 124, FSSP-124) available during the NRC FIRE.ACE campaign, which measured sizes from 5-98 μm diameter, overlapping with the FSSP-96 in the 4-47 μm diameter range. 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 also undersized particles >30 μm diameter. Therefore, we used the FSSP-96 data previously in this ACPD paper. However, during our sampling periods of interest in the NRC FIREACE campaign, the FSSP-124 data did appear to be of high quality based on the facts that the FSSP velocity ratios were not equal to 0.62 (a quality flag), and distribution and number concentrations of particles < 30 μm diameter were consistent with that of the FSSP-96.

To investigate further, we compared number spectra in the 6 predominantly liquid phase clouds observed on all flights 7-13 (dates were chosen to bracket the 2 relevant NRC FIRE.ACE clouds, which appeared on flights 8 and 12) (see Fig. R1, below). The FSSP-96 and the FSSP-124 had very similar spectra peak locations and number concentrations above 29 μm (Figure R1). That finding also does not support the hypothesis that the FSSP-96 was undersizing large droplets. However, there was a discrepancy in droplet numbers between the FSSP-96 and the FSSP-124, particularly in particles with diameters < 29 μm. If the FSSP-96 consistently underestimated droplet numbers, that could explain why this instrument underestimated LWC fairly consistently across days and cloud types.

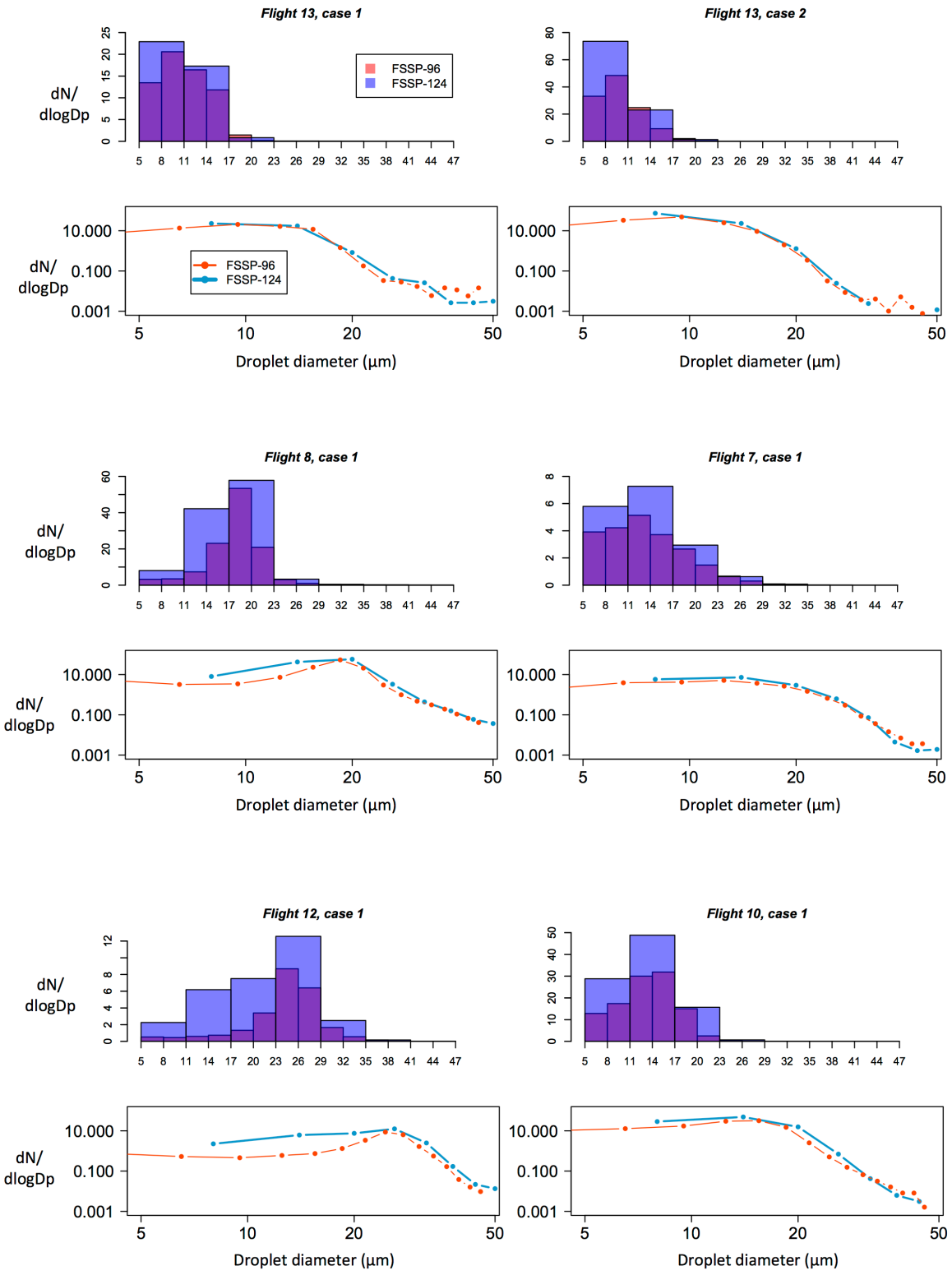

Figure R1. Droplet size distributions (the associated bottom panels containing line plots are the same data on a logarithmic scale) for 6 predominantly liquid clouds in the NRC FIRE.ACE campaign, including the two in this study (from flight 8 and 12). The FSSP-96 tends to have lower droplet numbers than the FSSP-124

in size ranges below ~29 µm.

On the flight with the best agreement for FSSP-96 and King LWC (flight 13), the FSSP-96 and FSSP-124 had similar droplet volume between 5-47 µm (their overlapping size ranges). On the flight with the least good agreement between the FSSP 96 LWC and the King LWC (flight 10), volume in the FSSP-96 was ~40% < that of the FSSP-124. This trend was consistent across the six cloud cases observed here (Fig. R2, below), and it suggests that the problem was with the FSSP-96 data and not with the FSSP-124 data.

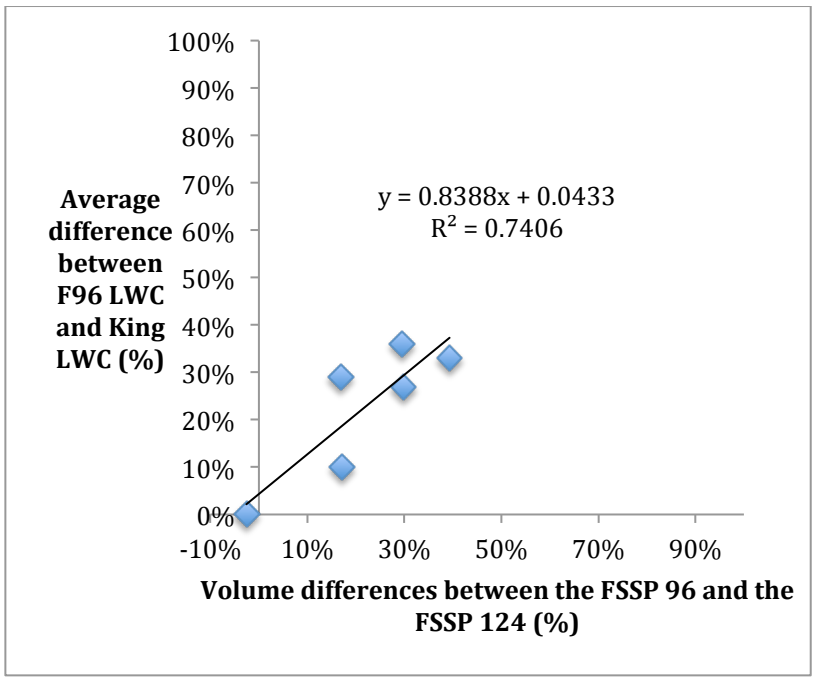

Figure R2. Average difference between FSSP-96 LWC and King LWC for the six clouds shown in Figure R1. Y-axis values were calculated from the difference between a 1:1 slope and the observed slope between FSSP-LWC vs. King LWC values (presented in percentages). If observations produced a 1:1 slope, it would correspond to a 0% value on the y-axis. The x-axis values is calculated from $(V_{F124} - V_{F96})/V_{F124}$, where V_{F124} and V_{F96} are the total volumes between 5-47 µm from the FSSP-124 and FSSP-96, respectively.

Based on the above information, we now have decided to use the FSSP-124 data for the 2 NRC FIRE.ACE cloud cases described in this study instead of the FSSP-96. The FSSP-124 data agree much better with King LWC values (slopes of 1.1 and 1.01 and R^2 values of 0.94 and 0.95) than the FSSP-96 data (slopes of 0.73 and 0.67, with R2 values of 0.94 and 0.96).

New text 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 throughout the paper. However, please note that the impact on the results is very minor, in part because there were only 2 distinct cloud cases that matched our background criteria from the NRC FIRE.ACE study.

3) P22835, L28: "background values of 0.018" – is this of CH3CN in ppbv? If so, make that clear.

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

4) P22837, L2: Using multiple BB tracers doesn't "minimize" the uncertainty, so much as it gives you a way of estimating the uncertainty in terms of the different resulting values.

We have changed the unclear wording here. However, to clarify our intended message in this sentence: our goal in using multiple tracers was not to estimate uncertainty, but rather to reduce biases from any one tracer. These biases are related to the fact that no tracer is a perfect estimate of the number of in-cloud aerosols that become cloud droplet nuclei. For example, in-cloud gas concentrations may not represent true aerosol number. CCN, aerosol number and aerosol chemical composition were generally measured near- but not in-cloud, and thus may not be truly representative of in-cloud dynamics. CCN likely

represents true cloud droplet nuclei better than aerosol number concentration, but the CCN-derived ACI estimates here are subject to more random error than the aerosol number estimates due to fewer sample numbers, and so forth. Therefore, to better clarify our intent here, the sentence has been changed from:

"… the magnitudes of derived ACI can vary depending on the BB_t tracers used. **To minimize the associated uncertainty**, we use a combination of up to six BB_t tracers to derive ACI, as available."

To:

"… the magnitudes of derived ACI can vary depending on the BB_t tracers used**, and any one tracer may be biased by random error and a variety of other reasons that may cause the tracer to imperfectly approximate actual cloud droplet nuclei**. To **reduce** the **biases inherent to any one tracer**, we use a combination of up to six BB_t tracers to derive ACI, as available."

5) P22841, L8-11: I don't think the fact that the results increase when two clouds are excluded is enough to say that non-linear processes "were indeed" affecting the ACI values. A less strong statement, "could have affected", would be more consistent with your evidence.

We have made the suggested change, as follows:

"That ACI values would increase to 0.08 (95% confidence interval 0.05-0.12) if the two biomass burning clouds were excluded suggests that non-linear processes **could have affected** the reduced ACI values in the case study."

6) P22867, Table 2: The column formatting of this table is odd – try cutting the redundant reference from the "Range" column and expanding the "Uncertainty" column. Also need an uncertainty value for the chilled-mirror hygrometer.

Done. The information on the chilled hygrometer has been removed since it was not used in the paper.

7) P22868, Table 3: Why doesn't this table have horizontal lines like Tables 1 and 2?

We will request of the copyeditors that this change be made. Thanks.

8) P22869, Table 4: Surely uncertainty data for the nephelometer and humidigraph exist somewhere, otherwise why should we trust the data at all?

With the various changes to the paper, we no longer present relative humidity data, so the information on relative humidity has been removed from Tables 1-4.

9) P22879, Figure 6: This caption needs more detail, like in Figure 8.

Done.

10) P22880, Figure 7: The caption should discuss the CO* as well, like in Figure 5.

Done.

11) P22881, Figure 8: The caption doesn't match the number or color of lines in the figure.

To better convey the information in this figure, we have changed the caption as suggested. Additionally, we have added a legend and changed the figure's color coding.

12) Typos: P22826, L3: Need a comma between "areas" and "such"

Done.

13) P22833, L5-6: How about "SO42-, and submicron organic aerosol, or OA, concentrations in ARCTAS, and by SPLAT II number concentration in ISDAC"? I'm not sure what "number composition" means.

Done, and we have changed "number composition" to "particle composition". The new sentence reads:

"In addition, **in all clouds we** assessed cloud pressure, location, temperature, **and** on-flight video (when available). **In** biomass burning cases **we also assessed** nearby aerosol conditions (as determined **in ISDAC by** SPLAT II **particle** composition **and in ARCTAS** by CH_3CN, black carbon (BC), submicron SO_4^{2-} and submicron organic aerosol**, or** OA**,** concentrations)."

14) P22835, L18: Appendix A is so short, you should just include it here.

Done.

15) P22837, L24: Instead of "in the text below", name the section (in this case Section 2.6).

Done.

16) P22838, L28: Again, name the section (3.1).

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17) P22814, L26: Should this be a separate section from the text above?

Apologies, we were unable to address this comment because there was no P22814, and we were not sure to which text the reviewer was referring.

18) P22844, L18: The order of Figure 6 and 7 should be switched, as you discuss Figure 7 before Figure 6.

There was likely some confusion here because we actually discussed figure 6 in two places. The first place it was discussed was on p.22840, l.25. The first mention of Fig. 7 was on page 22842, l.5. The next mention of Figure 6 was on page 22844, l. 17.

19) P22844, L29: I think it would be clearer to say, "increased in smoky conditions"

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Aircraft-measured indirect cloud effects from biomass burning smoke in the Arctic and subarctic

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Abstract

The incidence of wildfires in the Arctic and subarctic is increasing; in boreal North America, for example, the burned area is expected to increase by 200-300% over the next

50-100 years, which previous studies suggest could have a large effect on cloud microphysics, lifetime, albedo, and precipitation. However, the interactions between smoke particles and clouds remain poorly quantified due to confounding meteorological influences and remote sensing limitations. Here, we use data from several aircraft campaigns in the Arctic and subarctic to explore cloud microphysics in liquid-phase clouds influenced by biomass burning. Median cloud droplet radii in smoky clouds were ~40-60% smaller than in background clouds. Based on the relationship between cloud droplet number (N_{liq}) and various biomass burning tracers (BB_t) across the multi-campaign dataset, we calculated the magnitude of subarctic and Arctic smoke aerosol-cloud interactions (ACI, where ACI = (1/3)* d ln(N_{liq})/d ln(BB_t)) to be ~0.16 out of a maximum possible value of 0.33 that would be obtained if all aerosols were to nucleate cloud droplets. Interestingly, in a separate subarctic case study with low liquid water content (~0.02 g m^{-3}) and very high aerosol concentrations (2000-3000 cm^{-3}) in the most polluted clouds, the estimated ACI value was only 0.05. In this case, competition for water vapor by the high concentration of cloud condensation nuclei (CCN) strongly limited the formation of droplets and reduced the cloud albedo effect, which highlights the importance of cloud feedbacks across scales. Using our calculated ACI values, we estimate that the smoke-driven cloud albedo effect may decrease local summertime shortwave radiative flux by between 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. We lastly explore evidence suggesting that numerous northern latitude background Aitken particles can interact with combustion particles, perhaps impacting their properties as cloud condensation and ice nuclei.

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1 Introduction

The incidence of wildfires in the Arctic and subarctic is increasing dramatically (Flannigan et al., 2009; Moritz et al., 2012; Stocks et al., 1998), and in some areas, such as boreal North America, it is expected to grow by 200-300% over the next 50-100 years (Balshi et al., 2009). Already, periods of intense wildfires can increase regional aerosol

concentrations in the Arctic twofold (Warneke et al., 2010), and the impact of smoke is increasingly being recognized as a strong contributor to Arctic haze (Hegg et al., 2009, 2010; McConnell et al., 2007; Shaw, 1995; Stohl et al., 2006, 2007). Increases in biomass burning aerosols could have a large effect on cloud dynamics (Earle et al., 2011; Jouan et al., 2012; Lance et al., 2011; Lindsey and Fromm, 2008; Rosenfeld et al., 2007; Tietze et al., 2011); in turn, smoke-derived changes to cloud microphysics may result in changes to precipitation and regional heating that are strong enough to affect dwindling regional sea ice (Kay et al., 2008; Kay and Gettelman, 2009; Lubin and Vogelmann, 2006; Vavrus et al., 2010).

However, the interactions between smoke particles and Arctic clouds are poorly quantified, in part due to the confounding effects of meteorology and surface conditions (e.g., Earle et al. (2011); Jackson et al. (2012); Jouan et al. (2012)), and in part due to satellite sampling constraints over the Arctic, such as caused by the presence of many low contrast regions, multi-layer clouds (Intrieri et al., 2002), and reduced sunlight. 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). Following Eq. (1), ACI estimates for a given location can be derived from aircraft measurements of cloud droplet number, N_{liq}; they can also be derived from ground-based or remote sensing retrievals of changes in cloud properties such as droplet effective radius (r_e) or cloud optical depth (τ) at constant liquid water path (LWP) (Feingold et al., 2001; McComiskey et al., 2009):

$$ACI = \frac{1}{3}\frac{d\ln N_{liq}}{d\ln BB_t} = -\frac{\partial \ln r_e}{\partial \ln BB_t}\Big|_{LWP} = \frac{\partial \ln \tau}{\partial \ln BB_t}\Big|_{LWP} \quad (1)$$

The ACI term as defined by Eq. (1) was originally described as 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 microphysical responses than with radiative forcing.

The maximum value of ACI as derived from Eq. (1) is 0.33. An ACI value of 0.33 corresponds with the 1.0 maximum possible change in $\ln N_{liq}$ relative to $\ln BB_t$, which would occur if every aerosol were to nucleate a cloud droplet. The first term of Eq. (1) is

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divided by 3 in order to correspond with the last two terms, which are derived at constant LWP from the following theoretical relationships: $r_e \alpha LWP/\tau$ (Stephens, 1978) and $\tau \alpha N_{liq}^{1/3}$ (Twomey, 1977). Note that although each term in Eq. (1) should equal each other term, in practice measurement-derived biases can cause apparent differences between the terms. This issue will be discussed further in later sections.

One study convincingly demonstrated that smoke reduces cloud droplet effective radius and enhances cloud albedo in Arctic liquid clouds (Tietze et al., 2011). In that study, modeled BB_t concentrations were combined with remote sensing of cloud properties, enabling the authors to reduce meteorological bias by basing their conclusions on tens of thousands of clouds sampled over a variety of meteorological conditions throughout the Arctic. Smoke ACI values derived from relative changes in cloud r_e were estimated at between 0.04-0.11 out of a maximum 0.33. (Note however that in that study, clouds were binned by temperature and pressure, rather than by LWP as in Eq. (1) above.)

However, despite being able to conclusively demonstrate a smoke cloud albedo effect, Tietze et al. (2011) noted that they might have underestimated the magnitude of satellite-derived ACI values because of difficulties constraining aerosol concentrations and locations. They cite a study by Costantino and Breón (2010), where it was demonstrated that not co-locating aerosol-cloud layers in the vertical column dramatically lowered ACI estimates from 0.24 to 0.04 over marine stratocumulus clouds influenced by African biomass burning. This bias seems to be apparent in many ACI estimates globally; from a literature search, McComiskey and Feingold (2012) revealed that remote sensing-derived ACI values worldwide are lower than those derived from in-situ, modeling and/or ground-based studies. They also showed that in addition to errors in co-location of clouds and aerosols, the comparatively low spatial resolution of remote sensing observations can further enhance the low bias in ACI estimates.

In the Arctic, these biases can be substantial. In a study in Northern Finland, ACI estimates derived over the same general time period and location from both ground-based and remote sensing methods were ~0.25 and 0.09±0.04, respectively (Lihavainen et al., 2010); a more than two-fold difference. For reference, the range of Arctic remote sensing-derived ACI estimates for all aerosol sources is -0.01 to 0.09 (Lihavainen et al.,

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2010; Tietze et al., 2011); in situ, ground-based, and model estimates range between 0.05-0.3 (Garrett et al., 2004; Lihavainen et al., 2010; Zhao et al., 2012). The degree of bias at other global sites has led McComiskey et al. (2012) to assert that the albedo effect can only be assessed accurately from aircraft or ground-based in situ data.

To better understand the impacts that expected increases in smoke will have on the Arctic, it is important to better constrain remote-sensing and model estimates of smoke-specific ACI in the Arctic using in situ aircraft data. The biggest challenge in obtaining representative aircraft-based ACI values is the fact that they are more prone to uncertainties caused by the influences of poorly constrained meteorological factors (Shao and Liu, 2006) than other methods due to logistical limitations in sample size. We confront this issue in two ways. First, we focus on a case study day from the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) campaign (Fuelberg et al., 2010; Jacob et al., 2010) in which several clouds were sampled under very similar conditions. We derive ACI estimates for all clouds that were either verifiably clean or are clearly influenced by biomass burning aerosols, and contrast the observed cloud properties. Second, to increase sample size, we consolidated data from four separate aircraft campaigns in the Arctic. In addition to ARCTAS, these datasets include: the First ISCCP (International Satellite Cloud Climatology Project) Regional Experiment Arctic Clouds Experiment *(*FIRE.ACE), which included portions flown by the University of Washington Convair-580 (UW FIRE.ACE) and the Canadian National Research Council Convair-580 (NRC FIRE.ACE) (Curry et al., 2000), and the Indirect and Semi-Direct Aerosol Campaign (ISDAC) (McFarquhar et al., 2011). We then compare these findings with those from the ARCTAS case study.

2 Methods

2.1 Dataset description

The dates and flight locations of data used in this study are shown in Fig. 1, and the data used are listed in Tables 1-4. The ARCTAS, FIRE.ACE, and ISDAC datasets have each been extensively described previously (e.g., Curry et al., 2000; Fuelberg et al., 2010; Jacob et al., 2010; Korolev et al., 2003; McFarquhar et al., 2011; Rangno and Hobbs,

2001; Soja et al., 2008). However, to our knowledge, they have never been compared directly to each other. Here we note only briefly a few relevant points about the datasets and how they are inter-compared.

First, during the ISDAC and FIRE.ACE flights, multiple passes inside clouds were often obtained, and aerosols were intentionally sampled above- and below-cloud. In contrast, during ARCTAS there was very limited resampling of a given region and generally only one pass through a cloud was obtained. This difference in sampling impacts our results only in that there are not as many vertical profiles through the ARCTAS clouds as in the other datasets. Second, the UW FIRE.ACE dataset contains some gaps in positional data (latitude, longitude, and altitude), which range most frequently between 1-10 seconds, with rare instances of gaps >1 minute. If the data were out-of-cloud and if the gap in positional data is <1 minute, we linearly interpolate the latitude, longitude, and/or altitude. Otherwise, occasional gaps > 1 minute and data without positional information were excluded. Thirdly and most importantly, we have made our best effort to use data that are as comparable as possible between campaigns. However, when high quality measurements are not available from the same instrument in all campaigns, we use the most similar measurement available and we discuss the uncertainties this raises in the text.

2.2 Cloud presence and phase

2.2.1 ARCTAS

In ARCTAS, cloud liquid water content (LWC) was determined from droplet size spectra gathered with the CAPS-CAS instrument (Baumgardner et al., 2001) 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

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qualitatively consistent trends with the CAPS-CAS LWC data. Liquid phase cloud presence was defined by LWC values ≥ 0.01 g m^{-3} (Matsui et al., 2011b), a value that corresponds well with cloud presence verified from the on-flight video. Because neither ice water content (IWC) nor cloud particle images were directly measured during ARCTAS, we are unable to accurately verify cloud phase at temperatures < 0 °C in the ARCTAS dataset. Therefore, we limited our focus within the ARCTAS dataset to clouds present at temperatures > -0.5 °C (i.e., those clouds highly likely to be in the liquid phase). We also excluded clouds that video indicated were affected by drizzle or ice precipitation from cloud layers above.

2.2.2 FIRE.ACE and ISDAC

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

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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 μ m.

During ISDAC, LWC was determined from cloud droplet probe (CDP) data. These data agreed within 15% of the bulk probe values. Following Earle et al. (2011), FSSP data were used on days when high-quality CDP data were unavailable; the FSSP data are estimated to agree with CDP data to within 20%. 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).

For comparability with ARCTAS clouds, the presence of liquid clouds in the FIRE.ACE and ISDAC datasets was determined by simultaneous measurements of LWC > 0.01 g m^{-3}. Also, for inter-campaign comparisons we focused on clouds sampled for ≥ 20 s in order both to increase representativeness of the average measured properties of the clouds and to enhance meteorological similarity of clouds. Sometimes entrainment from outside air caused pockets of low- to no-LWC (i.e., LWC <0.001 g m^{-3}) within a cloud body; these pockets of air were not included when determining the average cloud droplet effective radius.

There is no consistent definition for cloud phase in the literature. In remote sensing studies for example, cloud phase is usually determined by cloud radiative properties – thus, clouds with some mixed particles can be included in "liquid" or "ice" phase classifications if they are mostly liquid or mostly ice (e.g., Baum et al. (2012), Platnick et al. (2003)). Due to instrumentation limitations, aircraft studies sometimes also define a cloud with small fractions of ice particles as being a "liquid" cloud (e.g., Korolev et al. (2003)). Alternatively, distinct portions of a cloud may be classified as different phases if a primarily liquid portion of a cloud is far away (~1-2 km) from a mixed portion of a cloud mass (McFarquhar et al., 2007; Zuidema et al., 2005).

Here, we define liquid cloud phase by the lack of any ice particles in the CPI data throughout the entire cloud transect, based on a roundness criterion (Lawson et al., 2001). When possible (i.e., in the NRC FIRE.ACE and ISDAC datasets), we verified that there was no detectable ice water along the cloud transects. This relatively stringent definition

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of liquid phase clouds is used to describe as best as possible the liquid phase end-member cloud characteristics. Because aircraft cloud transects can only sample a portion of a cloud, we must assume that the portion of the cloud sampled is representative of the rest of the cloud. This may introduce uncertainties, particularly in persistent large-scale stratus clouds. Nonetheless, as discussed in Sect. 3.1, we believe that errors from this assumption are not likely to have a large impact on our results.

2.3 Cloud microphysical properties

We used aircraft vertical profiles to assess cloud droplet effective radius (r_e), cloud liquid water path (LWP) and cloud optical depth (τ), and to gather information on aerosol properties above and below cloud. The r_e was derived by Eq. (2), following Hansen and Travis (1974):

$$r_e = \frac{\int r^3\, n(r)\, dr}{\int r^2\, n(r)\, dr} \qquad (2)$$

where r is the radius, and n(r) is the cloud particle size distribution. LWP is defined as the vertical integral of LWC from the base to the top of the cloud. LWP values were only determined when vertical profiles through the cloud were available, thus providing the cloud base and top heights. We define τ following Peng et al. (2002) as:

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$$\tau = \frac{3}{2}\frac{LWC\ H_c}{r_e \rho_w} \qquad (3)$$

where H_c is cloud thickness (again only available in vertical cloud transects) and ρ_w is the density of water. In addition to vertical transects, we also used horizontal transects within clouds to obtain information on horizontal variability of within-cloud properties and to obtain increased sample numbers for r_e.

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. Clouds were determined as being related in part by a

combination of time and location sampled. Here, the range of distance and time between clouds deemed as related or the same ranged from 0.4 -76 km and several seconds to 2.5 hours apart, depending on the conditions and cloud type (the 2.5-hour time frame included 8 separate transects through a stratus cloud). In addition, in all clouds we assessed cloud pressure, location, temperature, and on-flight video (when available). In biomass burning cases we also assessed nearby aerosol conditions (as determined in ISDAC by SPLAT II particle composition and in ARCTAS by CH_3CN, black carbon (BC), submicron SO_4^{2-} and submicron organic aerosol, or OA, concentrations). 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.

2.4 Air mass classification

For this work, distinguishing smoke-influenced from background cloud conditions is critical. During ARCTAS, background conditions were selected by a combination of in-cloud gas concentrations (average CO < 123 ppbv and average acetonitrile (CH_3CN) < 0.14 ppbv) and near-cloud SO_4^{2-} and BC concentrations (< 0.3 µg m^{-3} and < 0.12 µg C m^{-3}, respectively). In ideal cases, "near-cloud" air masses were defined as half the width of the cloud if it was a vertical profile, and within 10 s before and after the cloud if it was a

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horizontal transect. However, sometimes the presence of a neighboring cloud or the vertical changes in the aircraft track forced us to use slightly smaller samples.

The 123 ppbv CO cutoff value represents the upper quartile range of time periods with concurrently low CO, CH_3CN, and BC (all separate indicators of combustion), and the CH_3CN cutoff is the median for these values. 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.

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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. For comparison, Lance et al. (2011) used a concentration of >200 ppbv CO for "polluted" (mostly biomass burning) cases.

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During the two FIRE.ACE campaigns, the combination of relevant high-quality and/or high-resolution aircraft chemical data for completely characterizing air mass sources were not collected, and remote sensing products useful for air mass classification were also unavailable. As a result, biomass burning-derived haze events were indistinguishable from anthropogenic pollution events in the FIRE.ACE datasets.

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Therefore, we only use FIRE.ACE clouds sampled under non-polluted background conditions for inter-comparison with the other datasets.

Because within-cloud gas concentrations were not available, we used average near-cloud (as defined above) aerosol concentrations to define "background" conditions in the FIRE.ACE data. To reduce the risk of any potential humidification effects, we excluded near-cloud air masses that had any observations of cloud particles in the CPI or that had LWC values ≥ 0.001 g m^{-3}.

To classify background air masses, we used Passive Cavity Aerosol Spectrometer Probe (PCASP) aerosol concentrations (CN_{PCASP}) directly adjacent to the cloud. The PCASP measures dehumidified particles with diameters between 0.12-3 µm. Previous authors have noted the presence of large numbers of small nucleation- to Aitken-mode particles (between ~15-85 nm) in the spring- and summer-time Arctic that appear to have natural sources (Garrett et al., 2004; Howell et al., 2014; Leaitch et al., 2013; Leck and Bigg, 1999; O'Dowd et al., 2010; Ström et al., 2009; Tunved et al., 2013; Zhao and Garrett, 2015). However, the relatively large minimum size cutoff of the PCASP (~120 nm) excludes these particles, while including low altitude particles from pollution and biomass burning sources, which tend to be in the accumulation mode (Earle et al., 2011; Lathem et al., 2013; Warneke et al., 2010). Thus, CN_{PCASP} tends to be a fairly good indicator of non-background conditions.

To be classified as background, air masses had to have CN_{PCASP} concentrations of ≤ 127 particles cm^{-3} (Shantz et al., 2012). This CN_{PCASP} cutoff is a more stringent criterion for determining clean conditions than those adopted by Jackson et al. (2012), Earle et al. (2011) and Peng et al. (2002), where respective values of < 200, 250 and 300 particles cm^{-3} were used, but the criterion applied here appears to exclude biomass burning and pollution aerosols fairly effectively (Table 6). However, the upper 95% CH_3CN concentrations are higher than typical background conditions, indicating that our chosen cutoff value is generally, but not completely, effective at removing air masses influenced by smoke. Therefore, the FIRE.ACE samples have a more uncertain background classification than the ARCTAS and ISDAC datasets, where actual chemical tracers verify the presence of pollution and biomass burning aerosols. For ISDAC samples,

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"background" conditions were determined by out-of-cloud CN_{PCASP} concentrations, in order to be consistent with the FIRE.ACE campaigns. However, the TSI aerosol concentrations (CN_{TSI}) and backscatter values were not used to assign a background classification (see Sect. 3.2 for further details).

A "biomass burning" classification was assigned in ISDAC data when a cloud had contact with discernable amounts of biomass burning aerosols, as determined by single particle mass spectrometer, SPLAT II (Zelenyuk et al. 2009; Zelenyuk et al. 2015), based on the mass spectral analysis of individual aerosol particles (Fig. 2). This method has been similarly employed to determine biomass burning influence in the ISDAC dataset previously (Earle et al., 2011; McFarquhar et al., 2011; Shantz et al., 2014).

2.5 Assessment of indirect effects from biomass burning

As mentioned before, the impact of smoke aerosols on cloud droplet activation was assessed by looking at aerosol-cloud interactions (ACI) of biomass burning aerosols on cloud droplet number. The ACI values were derived from changes in cloud droplet number relative to measured biomass burning tracers, BB_t, following Eq. (1) and using a non-parametric Kendall robust line-fit method. The Kendall robust line-fit model (also commonly known as the Theil-Sen method) (Sen, 1968; Theil, 1950) derives a linear model of a dataset from the median of the slopes between each two points in the dataset. While this method is not as commonly used as linear regressions, it performs similarly when data are normally distributed. In cases when the data are not normally distributed, this method is more appropriate than a linear regression because it reduces the impact of outliers.

As previously mentioned, ARCTAS was the only campaign where biomass burning gaseous tracers were directly quantifiable in-cloud (here we use BBt = CH_3CN (de Gouw et al., 2003) and BBt = CO (Tietze et al., 2011)), measured in ppbv. Both CO (Bian et al., 2013) and CH_3CN have appreciable background concentrations in the Arctic (as can be seen in Fig. 3a). Therefore, approximate background CO and CH_3CN concentrations of 99.2 and 0.088 ppbv, respectively, were subtracted prior to deriving ACI values from Eq. (1) in the case study. These background values were derived from the mean of the Kendall robust line-fit method analyses of ARCTAS CCN and CN_{PCASP} equivalent

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concentrations vs. CO (or CH_3CN) concentrations. In the multi-campaign analysis, background values of 0.018 ppbv CH_3CN were subtracted, due to lower background concentrations in the cleanest samples. 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 ~0.14 ppbv at altitudes of ~8 km (Kupiszewski et al., 2013; Warneke et al., 2009; A. Wisthaler, personal communication, 2015). A maximum error of 0.038 ppbv in background CH_3CN would equal at most 18% of the CH_3CN signal in biomass burning samples. For that reason, and because CH_3CN was only one of six tracers used to derive ACI values, the range of possible background CH_3CN concentrations is expected to have only minor impacts on the analysis. Arctic background CO is more consistent than CH_3CN, and in that case, the differences in background CO as computed from CN_{PCASP} vs. CCN line-fit analyses (93.0 and 105.4 ppbv, respectively) led to only a 2.6% change in the derived ACI values.

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Because the in-cloud CO and CH_3CN values were not available in the ISDAC or FIRE.ACE campaigns, we also compared aerosol tracers of smoke/polluted particles adjacent to the cloud as a BB_t quantity. The aerosol tracers used were CN_{PCASP} concentrations, backscatter at 550 nm, BC concentrations, and when available, CCN (not available in the UW FIRE.ACE campaign). For comparison to the PCASP, aerosol concentrations with diameters > 4 nm were measured with a TSI 3775 in ISDAC. Aerosols with diameters > 3 and 10 nm were measured during ARCTAS from TSI models 3025 and 3010, respectively. Because CN_{PCASP} values were not measured during ARCTAS, we combined APS and UHSAS sized aerosol data collected during that campaign into a similar size distribution as the CN_{PCASP} measurements (0.124-3.278 μm). 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.

There are some limitations of the ACI approach. First, a systematic bias can be introduced when aerosol and cloud properties are averaged or co-located in low spatial or temporal resolution datasets (McComiskey and Feingold, 2012). This particular systematic bias is generally not a large concern for in-cloud aircraft studies such as this

one where gas and/or aerosol measurements and N_{liq} measurements are either collected simultaneously or in very close proximity. Secondly, the magnitudes of derived ACI can vary depending on the BB_t tracers used, and any one tracer may be biased by random error and a variety of other reasons that may cause the tracer to imperfectly approximate actual cloud droplet nuclei. To reduce the biases inherent to any one tracer, we use a combination of up to six BB_t tracers to derive ACI, as available.

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 in aircraft in situ data by resampling throughout the cloud's life cycle. Resampling was sometimes, but not always, carried out for individual cloud cases presented here, and was not specifically carried out throughout the lifetime of the cloud. However, based on the results presented in Duong et al. (2011), the magnitude of this type of error is unlikely to have a large impact on our results, although we cannot with full confidence assess how cloud life stage might have impacted the way aerosols were interacting with the clouds.

The fourth limitation with the ACI method is that N_{liq} has a sublinear relationship with CCN (e.g., Morales et al. [2011]; Morales and Nenes [2010]), with particularly noticeable deviations from linear behavior expected when a cloud contains high CCN concentrations (e.g., Moore et al. [2013]). This behavior is driven by increased competition for water vapor, which in turn decreases cloud supersaturation and reduces the tendency to form additional drops. Because ACI values are typically derived from linear-type regressions, apparent ACI values can be reduced if clouds with high CCN are included in the analysis. We discuss the potential for this type of interaction where applicable in the text. Finally, the most difficult problem to address is the potential bias introduced if one does not account for meteorological conditions (Shao and Liu, 2006). We discuss the relationship of derived ACI with meteorology in sections 2.6 and 3.

2.6 Overview of surface and meteorological conditions

Ambient conditions such as cloud type and presence of drizzle from an overlying cloud

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deck were determined from available video, photos, flight notes and AVHRR images. Although in situ chemical and physical measurements were primarily used to determine end-member situations (i.e., where only smoke or only background air were the dominant sources of aerosols interacting with clouds), in some cases we discuss out-of-cloud aerosols with potentially more mixed sources. In these cases we supplemented chemical and physical data with 5-day HYSPLIT back trajectories (Draxler, R.R. and Rolph, G.D. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://www.arl.noaa.gov/HYSPLIT.php), NOAA Air Resources Laboratory, College Park, MD)) to determine recent air mass history. Using video, photos, and flight notes, clouds were also classified as either stratiform or cumuliform. Stratiform clouds were present at 1-3 km altitude. 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. In our dataset, none of the biomass burning cases were present below an inversion either; such inversions occurred only in four of the clean background cases, indicating generally unimpeded aerosol mixing from above and below for biomass burning clouds in these data. The cumuliform clouds were also found between 1 and 3 km, and although they were less optically thick than the stratiform clouds, optically thin ($\tau < 15$) and multi-layer clouds dominated all samples.

Across all clouds sampled during the four campaigns, there was substantial variation between cloud properties (Table 7) and the physical locations of the clouds (Fig. 4). For example, background clouds were primarily sampled over open-ocean and at higher latitudes, whereas the smoky clouds were primarily sampled at lower latitudes over land. For this reason, in addition to comparing median characteristics of all background and clean cases, we also focus on a case study where multiple clean and smoky clouds were observed under very similar meteorological and surface conditions (section 3.1).

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3 Results

3.1 Indirect effects of smoke in Arctic liquid phase clouds

On 1 July 2008 during the ARCTAS-B campaign, a variety of small cumuliform clouds were sampled during flight 18 over inland Saskatchewan, Canada. The physical

characteristics of the clouds were very similar (Table 8), being small (~0.7 km high, and ~0.2-7 km wide) non-precipitating clouds present between 1680 and 2650 m altitude, and far from any major temperature or water vapor inversions. All clouds were liquid phase, with low median LWC values of 0.02 g m^{-3} (the implications of which is discussed further down). All clouds had temperatures ranging from -0.1 to 3.1°C. All were sampled within 97 km^2 and 5.2 hours of each other, during which time each cloud experienced similar northeasterly wind direction.

Despite being exposed to similar meteorological and surface conditions, aerosol inputs to these clouds ranged significantly, with average CH_3CN and PCASP equivalent particle numbers ranging between 0.092-0.55 ppbv and 107-3001 cm^{-3}, respectively. The large range in chemical properties was due to the aircraft track, which repeatedly covered areas up- and downwind of local fresh smoke plumes from the Lake McKay fire. This fire is comprehensively described in the combination of Cubison et al. (2011), Alvarado et al. (2010), and Raatikainen et al. (2012).

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) (see Kondo et al., 2011 for further discussion on use of this tracer during ARCTAS). Given that CO has both pollution and biomass burning sources, this finding indicates smoke was the dominant aerosol contributor on that day, not pollution. Back trajectories also support this conclusion (Alvarado et al., 2010). Of the clouds sampled during this flight, two clouds met the classification criteria for being biomass burning influenced, three were classified as intermediate, and two met the ARCTAS background criteria.

As shown in Fig. 5, smoke is clearly correlated with reduced cloud droplet radius in the seven clouds studied (with an average 59% reduction relative to background clouds, Table 8). As expected, there was a concurrent increase in cloud droplet number (Fig. 5). Based on this increase, we compute a combined median ACI of 0.05 (bootstrapped 95% confidence interval 0.04-0.06) across all tracers shown in Fig. 5.

Although linear regressions were not used to derive ACIs, we plot them for each tracer in Fig. 5 to show the degree of variation between individual tracer ACI values. Other

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researchers have previously noted differences in calculated ACIs when these interactions are computed from different tracers (e.g., McComiskey et al. (2009), Lihavainen et al. (2010) and Zhao et al. (2012)), and these differences probably reflect a combination of measurement error and how well a given tracer approximates the sub-population of aerosols that are participating in cloud droplet activation (Lihavainen et al., 2010). As plumes age, there may also be increasing uncertainty in biomass burning aerosol co-location with gaseous tracers such as CO and CH_3CN, as these are subject to different depositional processes (Hecobian et al., 2011). However, in this case the fires were relatively fresh so this issue is unlikely to be an important source of uncertainty.

ACI estimates can also sometimes be influenced or even overwhelmed by systematic differences in local meteorological conditions associated with cleaner versus more polluted clouds (Hegg et al., 2007; Shao and Liu, 2006). For the case study, that possibility is unlikely because of the relatively small area and time frame considered and the similar meteorological conditions in which the clouds were sampled.

However, because case study smoky clouds had a combination of very low LWC, very high aerosol concentrations from a fresh fire, and consequently, very small droplet sizes (Fig. 6), it is likely that smoky case study clouds were less sensitive to further additions of smoke aerosols than clouds with lower aerosol concentrations. Such non-linear behavior is predicted when high CCN levels cause increased competition for water vapor, which in turn decreases cloud supersaturation and reduces the tendency to form additional drops (e.g., Moore et al. [2013]; Morales et al. [2011]; Morales and Nenes [2010]). Additionally, possible enhanced entrainment of outside air in smoky clouds compared to background clouds (Ackerman et al., 2004; Bretherton et al., 2007; Chen et al., 2012; Lebsock et al., 2008) could enhance droplet evaporation and further reduce ACI values from the expected adiabatic ACI maximum value at a given aerosol level.

Because in-situ ACI derivations assume linearity in the response of N_{liq} to BB_t, and such as assumption does not hold well at high CCN levels, we would expect to derive lower in-situ ACI estimates if clouds with very high CCN levels are included in the analysis (Rosenfeld et al., 2014). That ACI values would increase to 0.08 (95% confidence interval 0.05-0.12) if the two biomass burning clouds were excluded suggests that non-

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linear processes could have affected the reduced ACI values in the case study. For reference, at case study smoky CN_{PCASP} equivalent concentrations of ~2,000-3,000 cm^{-3}, modeled adiabatic ACI values were ~0.06-0.16 (Moore et al., 2013). The range in modeled ACI values depended on factors such as cloud vertical velocity and CCN hygroscopicity (the CCN spectrum). Given these model uncertainties and our estimated case study ACI value, any potential effects of entrainment were not clearly noticeable in our data.

For these reasons, although the 1 July 2008 case is in some ways ideal in that the clouds were sampled in very similar environmental conditions, it is not necessarily representative of typical cloud conditions in the Arctic. The clouds were present relatively far south in the subarctic (52-56°N) and were cumuliform compared to the more dominant Arctic stratus type clouds. Moreover, the case study clouds were subjected to fresh concentrated smoke rather than aged diluted smoke, as one would expect at higher latitudes. Therefore, as explained above, we expect case study clouds already affected by high smoke concentrations to have reduced sensitivity to additional smoke, particularly given the low LWC of the case study clouds.

To assess the impact of smoke on liquid clouds more generally, we compared background and biomass burning cloud properties sampled over the larger region shown in Fig. 4. This more expansive set of clouds includes a broader range of high-latitude meteorological conditions, making it more representative of overall conditions in the Arctic region. However, the greater heterogeneity also makes trends in the data more difficult to interpret, as we cannot describe in full detail the degree to which meteorological influences affected each cloud given the limitations of the datasets.

Despite the uncertain meteorological influence, we see qualitatively similar trends to those in the 1 July 2008 ARCTAS case study (Fig. 7). We find a 3.7 µm (42%) median reduction in r_e between the smoky and background cases (Table 7). Concurrently, median N_{liq} increased from 41 droplets cm^{-3} in background clouds to 338 droplets cm^{-3} in smoky clouds. Within stratiform-only and cumuliform-only liquid clouds, groupings that are somewhat more comparable meteorologically, the mean r_e differences are 2.5 and 6.4 µm (n= 13 and 14), respectively. However, the combined median ACI estimate from all

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tracers shown in Fig. 7 is 0.16 (95% confidence interval 0.14-0.17). This value is three times that of the case study, which is further evidence to suggest that cloud sensitivity to aerosols in the case study was lowered by aerosol-driven adiabatic reductions in cloud supersaturation (and possibly enhanced entrainment).

Observed smoke-driven reductions in liquid cloud droplet size and increases in cloud droplet number in both the case study and the multi-campaign analysis are in line with several other studies in the Arctic. Peng et al. (2002) found a similar difference in r_e of 4.8 μm to the multi-campaign analysis in two combined datasets in the Arctic (one of which was the NRC FIRE.ACE dataset), in conditions where PCASP values were > and < 300 particles cm^{-3}, although they did not specifically focus on biomass burning-related samples. Tietze et al. (2011) also found significant changes in LWP, τ, and r_e using remote sensing cloud observations combined with a modeled biomass burning tracer. In contrast, Earle et al. (2011) did not see a reduction in r_e in biomass burning-influenced clouds based on selected ISDAC samples. They attributed this finding to a combination of meteorological and microphysical factors. It is possible that some of the differences with our study are also caused by reduced contrast between selected clean and polluted cases, as their cutoff for defining clean conditions was higher than ours, and they did not include any samples that met our background criteria (which were only present during the 4 April 2008 ISDAC flight). Also note that the biomass burning-influenced cloud cases assessed by Earle et al. (2011) did not overlap with the clouds assessed in this study.

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 °C, and this assumption holds well. 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). We cannot fully rule out that non-sampled portions of the clouds in the multi-campaign analysis contained ice particles, or that different vertical layers had different r_e values. However, if the 6 ISDAC and FIRE.ACE background clouds that were either stratiform or that contained only horizontal transects are excluded,

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the results of the multi-campaign analysis are nearly the same (ACI = 0.15 and median background cloud r_e = 7.0 vs. 7.6 μm). Thus we do not believe that uncertainties in cloud phase had a major impact on our results.

3.2 Implications for radiation and precipitation

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 um) 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 winter 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 frequently observed in the Arctic and subarctic, the ACI value we use was determined from samples that included some clouds within broken cloud systems, 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. 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).

In contrast to the subarctic, in the Arctic high surface albedo will lessen the expected impact of the cloud albedo effect. Although future sea ice losses and associated reductions in surface albedo may affect the relative importance of the cloud albedo effect

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on Arctic clouds, others (e.g., Garret et al. (2004)) have suggested that in the Arctic, a more important impact of reduced cloud droplet size may be greater longwave opacity, which can lead to enhanced snow melt. Relatedly, smaller droplets may affect cloud lifetime either by extending it via reduced precipitation (the "second indirect effect" (Ackerman et al., 2000; Albrecht, 1989)) or by reducing it via enhanced water vapor competition and evaporation, as may have occurred in the case study.

Cloud droplet spectra from the 1 July 2008 ARCTAS case study clouds are shown in Fig. 6. Although sample size is small, the presence of smoke appears to narrow the droplet spectra from a dispersion of 0.84 in background clouds to 0.55 in smoky clouds, as calculated by the ratio between the standard deviation of the size distribution and the mean droplet radius. 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/coagulation processes are thought to become efficient enough to induce precipitation (Rosenfeld et al., 2012).

Cloud droplet spectra from the multi-campaign clouds are shown for comparison in Fig. 8. There is not as obvious a narrowing of spectra as for the case study, but median droplet concentrations in smoky clouds never reached above 28+ µm diameter, whereas median droplet diameter in background clouds did reach above this point (Fig. 8). Also, small droplet concentrations (those most susceptible to evaporation) increased in smoky conditions, and rainfall was only noted in clean conditions, as shown in Fig. 8 by elevated (>0.1 cm^{-3}) cloud droplet concentrations with diameters >50 µm (King et al., 2013). Therefore, although clouds outside the case study suffer large uncertainties related to their collection over heterogeneous conditions, their droplet distributions support the hypothesis of smoke-induced reductions in drizzle.

3.3 Interactions of background aerosols with dilute biomass burning particles: a potential uncertainty in ACI values

As mentioned previously, large numbers of nucleation- and Aitken-mode particles are frequently observed in the spring and summer Arctic and subarctic (Engvall et al., 2008; Leck and Bigg, 1999; Ström et al., 2009; Zhao and Garrett, 2015). These particles are thought to have a marine origin via some combination of new particle formation from

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marine gases (Allan et al., 2015; Leaitch et al., 2013; O'Dowd et al., 2010; Tunved et al., 2013) and direct oceanic nanogel emissions (Heintzenberg et al., 2006; Karl et al., 2012, 2013; Leck and Bigg, 1999; Orellana et al., 2011). Chemical data from the ARCTAS dataset also show the presence of numerous small particles with a natural background source (Fig. 9).

Previous studies also suggest that the small particles can condense upon larger particles (e.g., smoke) when such particles are present (Engvall et al., 2008; 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 derived from smaller, non-biomass burning particles mixed with sulphates and marine particles (Earle et al., 2011; Zelenyuk et al., 2010). To get some idea of how important the background particles may be, we estimated the maximum mean aerosol volume change that would occur if high concentrations of small background aerosols were to mix with and condense upon diluted smoke particles. Concentrations of background particles were estimated at 5000 cm^{-3} (based on high-end values observed in Fig. 9 and at another Arctic site (Ström et al., 2009)). Diluted smoke concentrations were estimated at 450 particles cm^{-3} (low-end values from Fig. 9). Volumes were calculated from the size ranges observed in ARCTAS background and smoky aerosols (see Appendix A for details). In this hypothetical scenario, we estimate that background aerosols could increase dilute smoke aerosol volume by up to 2-15%, although volume increases are likely substantially less in most air masses.

Interestingly, the small Arctic 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). Previous studies also suggest that volume increases alone might affect Arctic particle hygroscopicity, independent of chemistry (Moore et al., 2011). Given this information, we cannot rule out that upon condensation, the small background particles might act as surfactants or otherwise modify smoke CCN characteristics, causing deviations from the

ACI value as derived in section 3.1 at low smoke concentrations. This hypothesis is difficult to test because, excepting three intermediate instances in the case study, the data presented in Section 3.2 only included background and high smoke conditions.

However, the nucleation- and Aitken-mode 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 factors: 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 such as 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 the subarctic, wildfires peak in the summer (Giglio et al., 2006). Thus, although the influence of the small background particles on subarctic and Arctic smoke ACI values is probably minor, deviations from the linear ACI expectations derived here might occur during dilute summertime Arctic smoke events and in subarctic locations: for example when smoke is diluted over or near marine environments.

4 Discussion and Conclusions

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The challenge of separating the influence of meteorology and aerosol indirect effects on clouds introduces relatively large uncertainty in our understanding of how smoke impacts clouds. Using in situ aircraft data, we quantified these impacts in both a subarctic cumulus cloud case study and in a multi-campaign data assessment of clouds north of 50°N. The multi-campaign assessment suggests an ACI value of 0.16 (95% confidence interval 0.10-0.13), which is on the high end of previous satellite-based assessments (0.04-0.11) (Tietze et al., 2011). Given a known low bias in remote-sensing-derived estimates of ACI (e.g., McComiskey et al. (2012)), our findings suggest that smoke-derived increases in cloud albedo may be higher than previously derived in the region. We reduced confounding meteorological effects by including data from as wide a

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geographic region as possible, applying very stringent conditions to identify clean and smoky clouds, and reducing the impact of outliers on ACI derivations by using the Kendall robust line-fit method instead of normal linear regressions. However it is important to note that meteorological effects are still imperfectly constrained in this assessment due to inherent limitations in the in situ dataset size and content.

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.

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. In those regions, a more important effect of smoke might be its inhibition of precipitation and cloud lifetime effect, as evidenced by the observed reductions in cloud droplet radius of ~50% in both the case study and the multi-campaign assessment.

Smaller cloud droplets can have various consequences. Smoke-driven reductions or delays in precipitation may affect the distribution of aerosol and moisture deposition. Longer cloud lifetime could impact not only Arctic albedo but also longwave radiation

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(Stone, 1997), and previous studies suggest that even small changes in the above parameters may affect sensitive Arctic sea ice (Kay et al., 2008; Kay and Gettelman, 2009; Lubin and Vogelmann, 2006; Vavrus et al., 2010). Additionally, changes in cloud cover might also have indirect effects on ocean photosynthesis and biogeochemistry (Bélanger et al., 2013). It is our hope that the improved quantification of smoke-derived ACI values will help quantify these impacts in future model studies.

One obvious limitation of our study is that we do not address the impacts of smoke on existing mixed and ice phase clouds. Additionally, we cannot account for the ways in which smoke might have affected sample phase. For example, ice nuclei presence might facilitate the conversion of an otherwise liquid phase cloud into a mixed phase cloud that was excluded in this assessment. Alternatively, we could have included liquid clouds in our assessment that might otherwise have been present as mixed or ice phase clouds if not for the inhibition of freezing by soluble smoke compounds via the Raoult effect (discussed in Tao et al. (2012)).

Finally, we have presented evidence to suggest that coagulation of the numerous nucleation- and Aitken-mode background particles frequently present in clean summertime Arctic air masses might increase the volume of diluted smoke aerosols by up to 2-15%. Previous studies suggest that such interactions with background particles may increase smoke aerosol hygroscopicity, which in turn could cause deviations from the ACI value derived here. 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.

Appendix A:

Calculations for maximum potential contribution of background aerosol to diluted smoke aerosol volume

We first estimate the volume of smoke particles at dilute concentrations of 450 particles cm^{-3}. Arctic/subarctic smoke aerosol size distributions were taken from Kondo et al. (2011) and Sakamoto et al. (2015), where lognormal aerosol size distributions were

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characterized by geometric mean diameters of 224±14 nm and 230 nm and geometric standard deviations of 1.33±0.05 and 1.5, respectively. From the corresponding size distributions, we estimate smoke aerosol volumes of ~2.9-6.0 μm^3 per cm^{-3} of air at smoke concentrations of 450 cm^{-3}.

The degree to which aerosol properties can be affected by the collection of Arctic nucleation- and Aitken-mode background particles onto larger smoke and pollution particles also depends in part on the size ranges and concentrations of the background particles. These can be quite variable (Engvall et al., 2008) (also see Fig. A1). To estimate average background concentrations, we use the observed geometric mean ratio range in 6-year Svalbard summertime data (Engvall et al., 2008), which indicated that Aitken-mode particle concentrations were ~1.5-3 times greater than those of accumulation-mode particles. Given this range in ratios, we would expect background particle concentrations to be ~675-1350 cm^{-3} at smoke concentrations of 450 cm^{-3}. We then combine the expected small background aerosol concentrations with ARCTAS background aerosol spectra from events from 12 April, 10 July, and 13 July 2008 (Fig. A1) for particles < 80 nm in diameter. Based on these values, small background aerosol volume is estimated at 0.012-0.114 μm^3 cm^{-3}. A comparison of this volume with the previously estimated smoke aerosol volume suggests that background aerosols could contribute only ~0.2-4% of total diluted smoke aerosol volume in average summertime conditions. This estimate does not account the fact that all else being equal, small particles are usually more likely to coagulate onto the largest sized particles (Seinfeld and Pandis, 1998), which would reduce the contribution to average particle volume even further.

Alternatively, we can estimate what the background aerosol volume might be if particle concentrations were as high as 5000 cm^{-3}. Although such events are not common in the Arctic and subarctic, similar high-end concentrations of background particles are observed in Figure 9 and have been observed elsewhere in the Arctic as well (Ström et al., 2009). Again assuming the same range of particle size distributions observed in Fig. A1, the small background aerosol volume at 5000 particles cm^{-3} is estimated to be between 0.092-0.422 μm^3 per cm^{-3} of air. Thus, in this case background aerosols could

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full size spectra for the smoke aerosols, we aerosols, we combined the known size ranges of particles with diameters >523 nm with average same-day smoke size distributions of smoke particles <523 nm from the boundary layer. The boundary layer was the only location with full size spectra data (blue line in Fig. B2), and while it does appear to have different size spectra for particles with >523 nm diameter, quality flagged UHSAS data from the 3-5 km haze layer (not used in this calculation) suggest that the peak at 100-200 nm diameter is similar. Based on this estimated smoke size spectra and the concentrations of smoky haze particles from Fig. 11 (~ 900 particles cm^{-3}), haze layer smoke aerosol volume for the event measured in Fig. 11 was estimated at ~ 5.232 μm^3 per cm^{-3} of air.

add at most 2-15% of total aerosol volume in diluted smoke with concentrations of 450 particles cm^{-3}.

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3.3 Interactions of biomass burning particles with background aerosols - potential impacts on clouds

Previous authors have noted the presence of large numbers of small, low-scattering Aitken particles in the Arctic (Garrett et al., 2004; Howell et al., 2014; Leck and Bigg, 1999; Zhao and Garrett, 2015), which other studies have suggested may be marine in origin (Heintzenberg et al., 2006; Karl et al., 2012; Leck and Bigg, 1999; Orellana et al., 2011). New particle formation may be another source of the high Aitken particle number; marine processes can also be a substantial source of new particles (e.g., Allan et al., 2015). Either way, chemical data from the ARCTAS dataset (shown in Fig. 9), confirm that these Aitken particles were numerous and that they appear to have a natural background source.

Fortunately, the relatively large minimum size cutoff of the PCASP (~120 nm) excludes these background particles (Fig. 10), and the CN_{PCASP} concentration seems to accurately indicate the presence of particles from pollution and biomass burning sources (Figs. 9 and 10), independent of whether small background aerosols are present. This finding is in-line with previous studies that found low altitude Arctic biomass burning aerosols primarily in the accumulation mode (Earle et al., 2011; Warneke et al., 2010) and other studies that found the peak size of marine background particles to be between 25-80 nm (Leck and Bigg, 2005).

However, although the PCASP seems to effectively exclude the individual small background aerosols, there is evidence to suggest that these background particles are interacting with the larger accumulation-mode biomass burning and pollution particles, and perhaps changing their properties. In Figs. 9 and 10, we plot all non-cloud high-quality data from ARCTAS and ISDAC. These data from multiple air masses show that there are two distinct populations of aerosols: small particles in the clean background having low CO, CH_3CN, and backscatter, and larger, combustion-derived particles. In both campaigns there is a clear separation between low scattering background air masses and polluted air masses.

Two possibilities could explain the dual distributions. First, it is possible that two

separate air masses were observed that did not passively mix during the sampling periods (otherwise we would expect passive mixing to blur together the two distributions observed in Figs. 9 and 10). Another possibility is that the small background particles were coagulating onto larger aerosols when air masses having different aerosol sources came into contact.

At first, the larger scatter in aerosol-backscatter optical properties in ARCTAS-B samples compared to ARCTAS-A suggested the distribution was caused by two non-mixing separate air masses, as did the fact that most days were characterized by either all small, weakly scattering particles, or all larger, scattering particles. However, upon closer analysis, it seems that the greater range in backscatter at a given CN concentration in ARCTAS-B is more likely caused by differences in smoke optical properties. Most of the ARCTAS-B samples with backscatter > 2.5 Mm^{-1}, and with a high relative particle number compared to ARCTAS-A at a given backscatter level were from fresh Lake McKay smoke samples on 1 July 2008 (Fig. 9, bottom row). Previous work has shown that these plumes were evolving, with ongoing evaporation of primary smoke particles and formation of secondary smoke particles (Cubison et al., 2010). Those resulting changes likely explain the large range in particle number at high backscatter levels in the fresh plumes from that day. Also, despite the scatter in the ARCTAS-B samples shown in Fig. 9, there was still separation between low scattering background air masses and polluted air masses, as there was in ARCTAS-A and ISDAC.

To better understand the reason for the dual distributions in Figs. 9 and 10, we focused on a few cases when aerosol properties transitioned as the aircraft moved between clean and polluted air masses. On 1 July 2008 (downwind of the ARCTAS-B biomass burning case study) for example, the aircraft passed through several smoky haze layers into a clean air mass (Fig. 11). Both video and differential absorption LIDAR (DIAL) data confirm that the presence of the upper haze layer at 3-5 km was geographically widespread (Fig. 12). During the transition into cleaner air, CN_{TSI} number rapidly increased by ~1400 particles cm^{-3} within a 63 m vertical transition zone, and these particles were found to be fairly small (Fig. 11). 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.

As shown in Fig. 11c, there were some weak temperature and moisture inversions that might have inhibited mixing of these air masses to some degree (Fig. 11c). However, at the transition zone, both backscatter and OA concentrations changed at a much slower rate than the CN_{TSI} concentrations (Fig. 11), suggesting that mixing was in fact taking place over a broader scale. Although data were not available in the exact time period of interest, DIAL data taken shortly after the transition also suggest that the smoke layer was probably undergoing broad diffuse mixing (Fig. 12). In addition, despite the fact that high quality data were unavailable for size classifications < 0.5 μm diameter, it is clear that large particles contributed a higher relative percentage in the polluted layers than in the clean air mass (Fig. 11b), which is what we would expect if coagulation were occurring. Although not definitive, these observations suggest that coagulation was occurring during mixing.

Similar observations were made during a transect over the Bering Strait on 12 April 2008 (Fig. 13), when the airplane passed through a clean region with high CN_{TSI} and low backscatter sandwiched between two more polluted layers with higher CN_{TSI} and backscatter. Unfortunately, size distributions of particles with diameters < 0.5 μm were unavailable during this portion of the transect as well, but the available data indicate that there was again a higher fraction of larger particles in the polluted air masses (Fig. 13b). On either side of the clean air mass, submicron SO_4^{2-} concentrations began to rise, which back trajectories suggest was the result of transport from Asia. In the border regions, there was a sudden drop in CN_{TSI} of ~700-1000 particles cm^{-3} over an ~140 m vertical distance. As in the previous case, submicron SO_4^{2-} concentrations and backscatter also changed, but at a slower rate than CN_{TSI}. DIAL observations indicate that mixing of aerosols was occurring in a vertical zone of at least 500 m below the clean air mass.

If coagulation were occurring, it likely does not contribute to a large change in smoke aerosol volume during most smoke haze events. For example, we estimate that background aerosols could contribute only ~1-4% of the total smoke aerosol volume for smoke aerosols in the transition zone in the 1 July 2008 case presented in Fig. 11. (details on this calculation can be found in Appendix B). Although by volume this impact is fairly minor, Lohmann and Leck (2005) showed that marine-derived background particles are highly surface-active, and that they would likely activate at lower supersaturations than

anthropogenic material or fresh smoke. Therefore, if these background particles act as surfactants or if they otherwise modify smoke CCN or IN characteristics, the coagulation of these particles onto larger smoke particles might impact cloud droplet formation or the way the presence of smoke affects cloud phase changes. These background particle effects would probably be largest in air masses with very diluted smoke and/or higher concentration background particles. For example, if concentrations of smoke from the above case were diluted by half (to ~450 cm^{-3}) and background aerosol concentrations were doubled to ~5000 cm^{-3} (these values are within the spread of data shown in Figs. 9 and 10), background aerosols could contribute up to 16% of smoke aerosol volume if coagulation were to occur.

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-derived particles. These ultrafine particles are thought to activate at relatively low supersaturations (Lawler et al., 2014; Lohmann and Leck, 2005), and thus may have some effect on smoke aerosol properties such as hygroscopicity. The importance of this process is not well understood, but any changes in the effectiveness of smoke CCN and IN

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Thus, the condensation of external particles onto biomass burning aerosols merits further study.

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The Kendall robust line-fit model (also commonly known as the Theil-Sen method) (Sen, 1968; Theil, 1950) derives a linear model of a dataset from the median of the slopes between each two points in the dataset. While this method is not as commonly used as linear regressions, it performs similarly when data are normally distributed. In cases when the data are not normally distributed, this method is more appropriate than a linear regression because it reduces the impact of outliers.

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Appendix B:

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PMS airborne Passive Cavity Aerosol Spectrometer Probe (PCASP)-100X

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Temperature	Rosemont 102 probe	–65 to +35 °C	±1°C

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DMT continuous-flow, streamwise thermal-gradient CCN counter (reported between 14-37% supersaturation)

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Total dry backscatter (550 nm)	TSI 3563 Integrating Nephelometer	> 0.1 M m^{-1}	±0.5 M m^{-1}

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Relative humidity	LiCor Hygrometer	0-90%	n.a.

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Figure 10. The relationship between out-of-cloud aerosol concentration and backscatter at 550 nm given different lower-end particle size cutoffs in the a, b) ARCTAS and c, d) ISDAC datasets. Ultrafine aerosols appear to dominate the high aerosol number concentration/ low backscatter particles seen in a-c, as shown by their disappearance when a diameter cutoff of 140 nm is used (d). Measurements are from the following

instruments: a) TSI 3025, b) TSI 3010, c) TSI 3775, and d) PCASP. To show detail, some high values along both axes are not shown.

Figure 11. Relationships between (a) bulk CN_{TSI}, backscatter, and submicron OA concentrations, (b) APS aerosol concentrations in bins from 0.58-3.0 μm, (c) and altitude, temperature and relative humidity along the ARCTAS-B flight track from 1 July 2008 (shown in panel d), for data within the area boxed in red (shown in higher resolution in panel e) as the airplane moved north. Data are shown as a function of seconds UTC since the start of the date on which measurements began.

Figure 12. Images of the haze layer sampled at ~3-5 km altitude in Fig. 11 from a) on-board aircraft video and b) the DIAL instrument (data from http://science.larc.nasa.gov/lidar/arctas/dial_18.html). Arrows point out the presence of the haze layer sampled in Fig. 11.

Figure 13. Relationships between (a) bulk CN_{TSI}, backscatter, and submicron SO_4^{2-} concentration, (b) APS aerosol concentrations in bins from 0.58-3.0 μm, (c) and altitude, temperature and relative humidity along the ARCTAS-A flight track from 12 April 2008 (shown in panel d), for data within the area boxed in red (shown in higher resolution n panel e) as the airplane moved north. Data are shown as a function of seconds UTC since the start of the date on which measurements began.

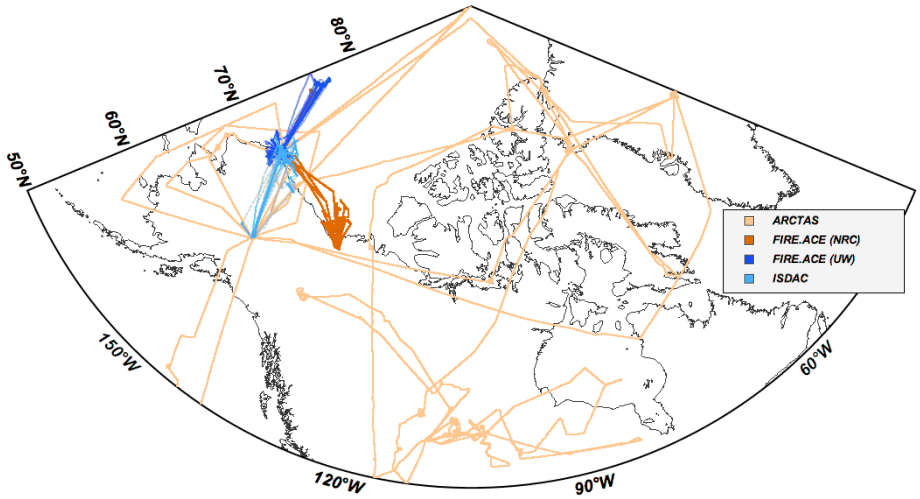

Figure 1. Sampling locations for the following campaigns: ARCTAS (light orange), NRC FIRE.ACE (dark orange), UW FIRE.ACE (dark blue), and ISDAC (light blue). The locations of clouds sampled are shown in Fig. 4.

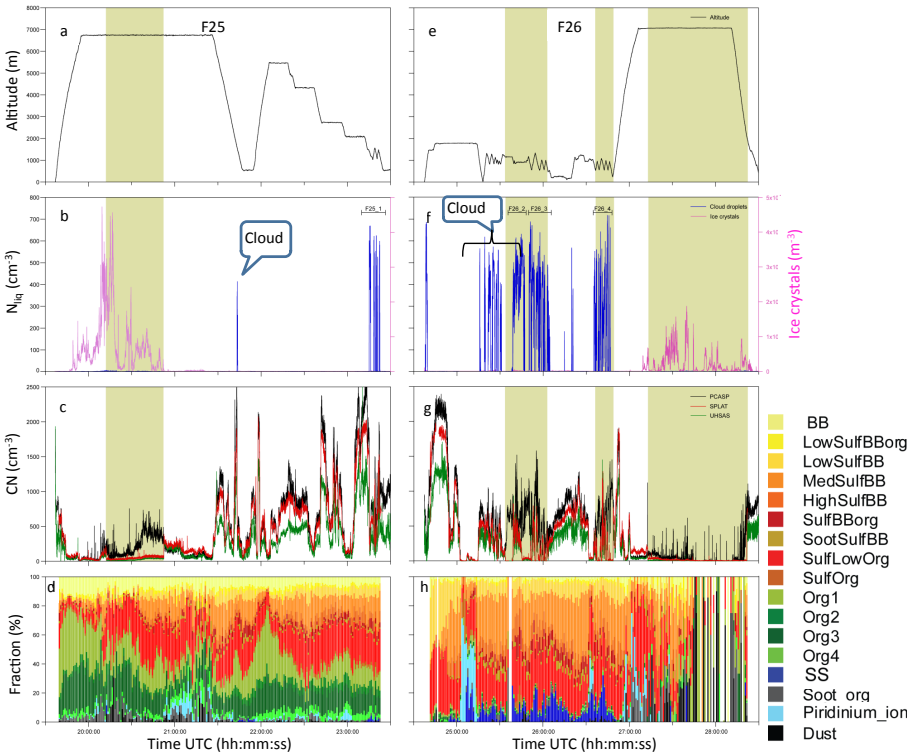

Figure 2. ISDAC 2008 aerosol and flight characteristics near and in selected clouds influenced by biomass burning from 19 April (left) and 20 April (right). Flight characteristics shown include: a) altitude, b) LWC (blue) and IWC (pink), c) aerosol concentration from the PCASP (black), SPLAT (red), and UHSAS (green) instruments, and d) bulk aerosol SPLAT chemical composition. Tan shading indicates SPLAT sampling through the in-cloud CVI inlet.

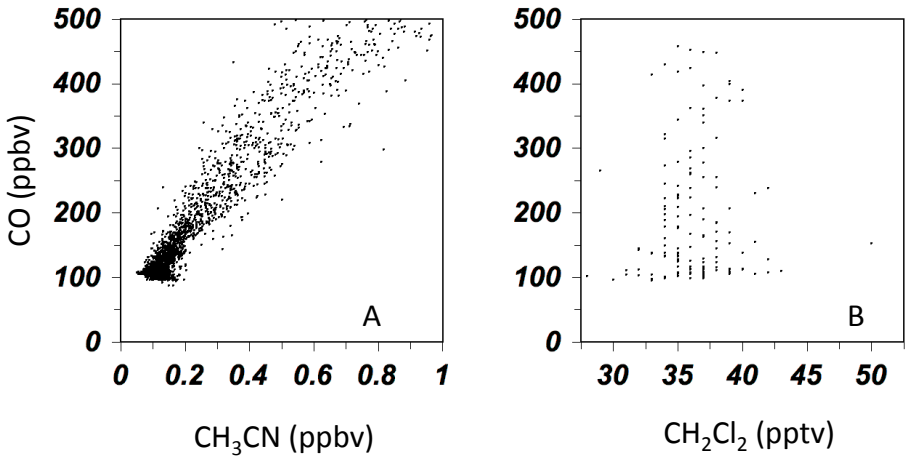

Figure 3. Carbon monoxide (ppbv) during the 1 July 2008 ARCTAS-B flight as a function of a) the biomass burning tracer CH_3CN (ppbv) and b) the fossil fuel combustion tracer CH_2Cl_2 (pptv).

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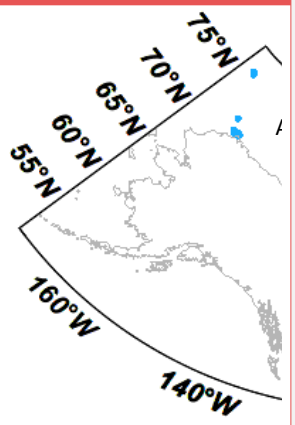

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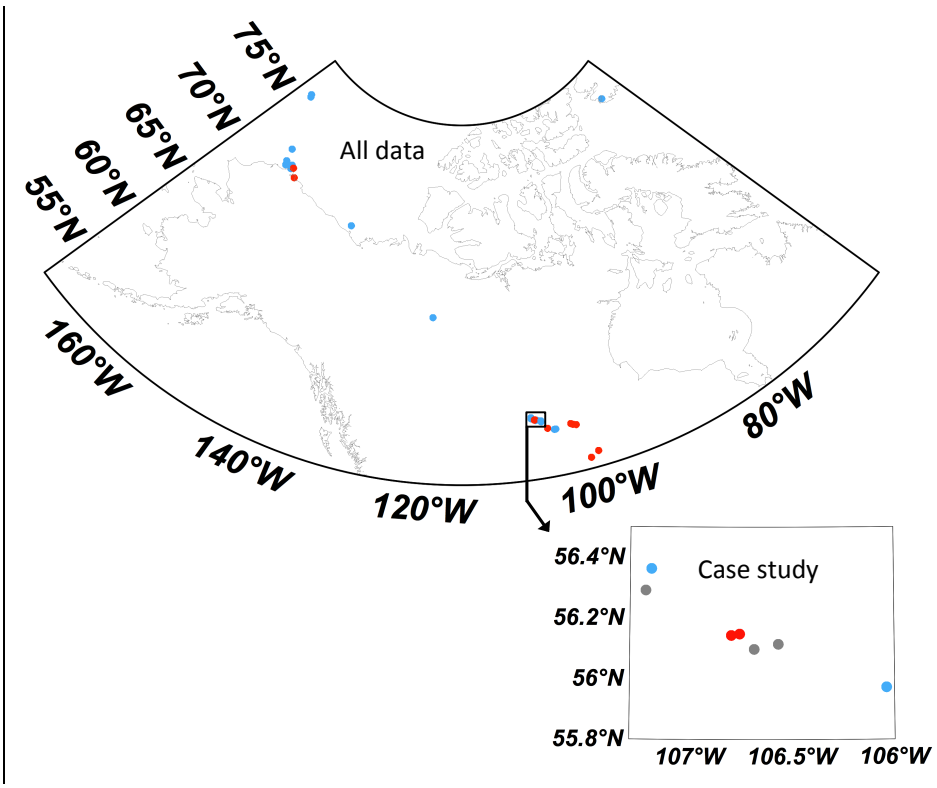

Figure 4. Map of cloud sample locations from all campaigns. Red points indicate biomass burning samples, blue cases indicate background samples, and grey points indicate intermediate samples.

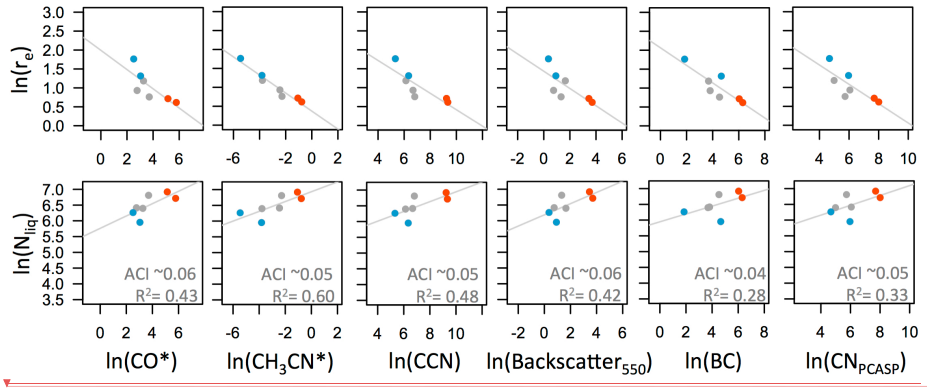

Figure 5. Based on seven samples from the ARCTAS-B 1 July 2008 case study, here we show the relationships between $\ln(r_e)$ (top row) and $\ln(N_{liq})$ (bottom row) and $\ln(BB_t)$ derived from six indicators (where BB_t = CO (ppbv) (* indicates background values of 99.2 ppbv have been subtracted), CH_3CN (ppbv) (* indicates background values of 0.088 ppbv have been subtracted), CCN (cm^{-3}), backscatter at 550 nm (Mm^{-1}), BC (μg C m^{-3}), and CN_{PCASP} equivalent values (cm^{-3}), as calculated from UHSAS and APS measurements. Biomass burning samples are noted in red, and background samples are noted in blue. To show variation between tracers, linear regressions and associated ACI estimates are shown in light gray (but note that final ACI values are not derived from individual regressions, but rather a combination of all six tracers).

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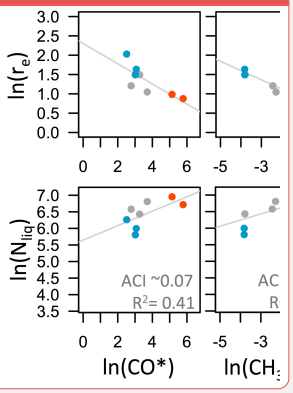

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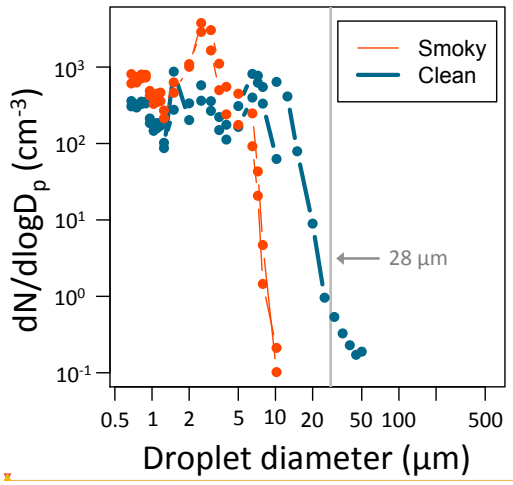

Figure 6. Mean cloud droplet size distributions (µm) for individual case study biomass burning clouds (thin orange lines) and clean background clouds (thick blue lines). The 28 µm line is marked in grey.

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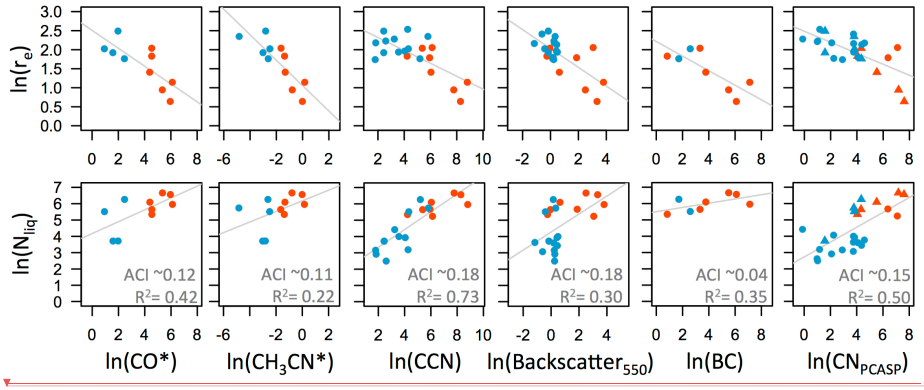

Figure 7. Same as in Fig. 5, but for data from the multi-campaign analysis. As in Figure 5, CO* indicates that background values of 99.2 ppbv have been subtracted. For CH_3CN, the * indicates background values of 0.018 ppbv have been subtracted (due to low background CH_3CN levels in some of the samples).

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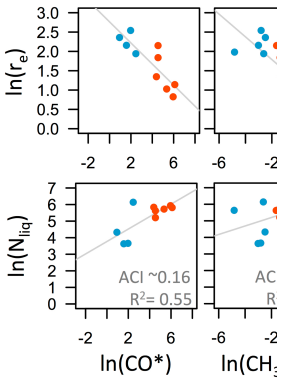

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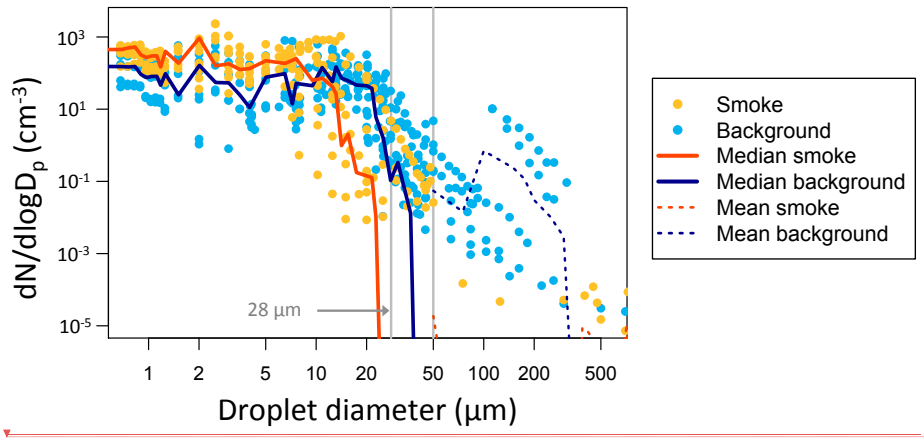

Figure 8. Mean cloud particle size distributions (µm) for all non-case study biomass burning clouds (yellow dots) and clean background clouds (light blue dots). The 28 and 50 µm lines are marked in grey. Thick red and darker blue lines indicate median values for binned size classes for smoky and clean clouds, respectively, including zero values not shown on the log-log plot. Due to the high number of zero values above >50 µm diameter, the mean values above this level are also shown (dashed lines) for comparison.

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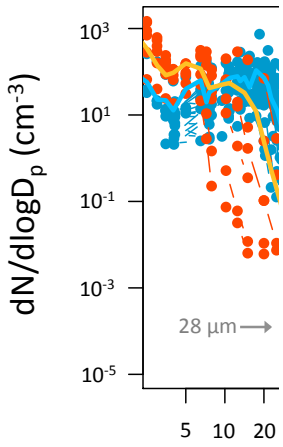

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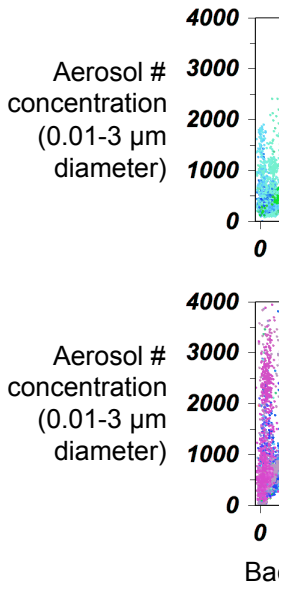

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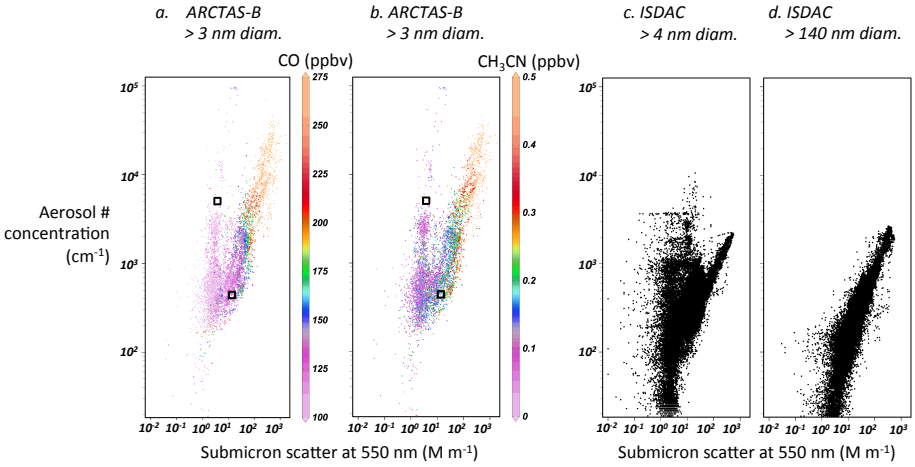

Figure 9. Log relationships between ARCTAS-B and ISDAC aerosol number concentration and submicron scatter. In panels a and b, the combustion tracer CO, and the biomass burning tracer CH_3CN in out-of-cloud air masses are also shown. The black squares in panels a and b indicate where background aerosol concentrations of 5000 cm^{-3} and dilute smoke concentrations of 450 cm^{-3} would be relative to other points. Measurements are from the following instruments: a and b) TSI 3025, c) TSI 3775, and d) PCASP. ARCTAS-B summertime samples were taken at altitudes < 5.2 km; ISDAC samples were taken at <3.65 km due to TSI 3025 instrument limitations. All quality-flagged data were excluded, as well as suspicious ISDAC values within 17 km and <1 km altitude of the Fairbanks, Alaska airport. Very small background aerosols appear to dominate the high aerosol number concentration/ low scatter particles seen in a-c, as shown by their disappearance when a diameter cutoff of 140 nm is used (d).

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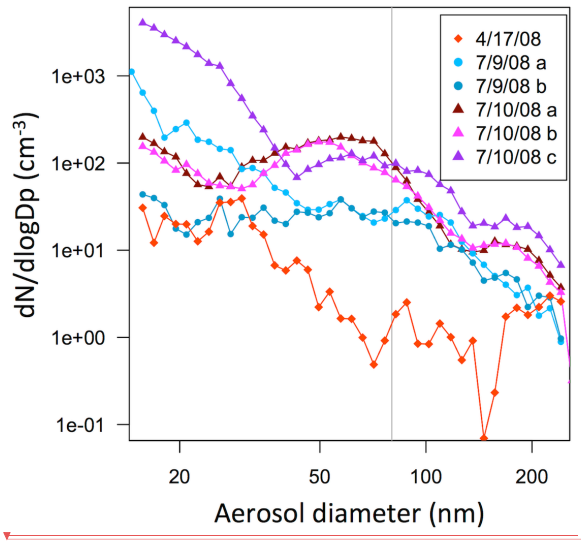

Figure A1. Mean out-of-cloud aerosol particle size distributions for several ARCTAS background aerosol events. Some days had multiple background aerosol events; these are distinguished by color and the letters a-c. The light grey line shows the 80 nm cutoff used here to distinguish Aitken mode particles from accumulation mode particles.

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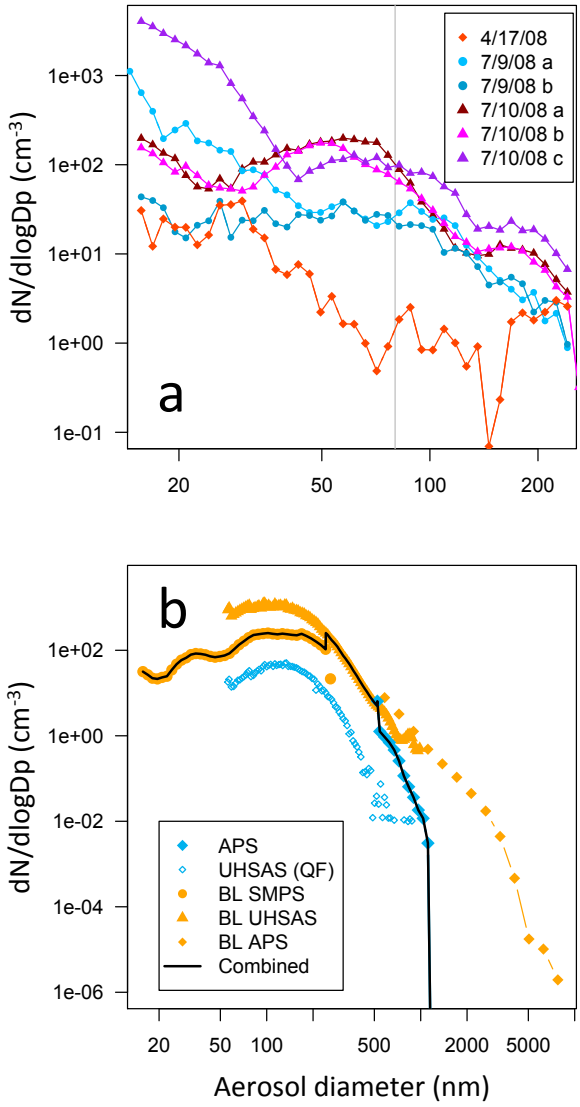
Deleted: Panel b) APS and UHSAS values (in blue) are from the 3-5 km altitude haze layer described in Fig. 11. Other data (shown in orange) are the boundary layer (BL) smoke spectra. The UHSAS data from the 3-5 km altitude haze layer had a quality flag, and were not used to determine the estimated size spectra (shown as the black line). Note that panels a) and b) have different axes values.

Figure 10. The relationship between out-of-cloud aerosol concentration and backscatter at 550 nm given different lower-end particle size cutoffs in the a, b) ARCTAS and c, d) ISDAC datasets. Ultrafine aerosols appear to dominate the high aerosol number concentration/ low backscatter particles seen in a-c, as shown by their disappearance when a diameter cutoff of 140 nm is used (d). Measurements are from the following instruments: a) TSI 3025, b) TSI 3010, c) TSI 3775, and d) PCASP. To show detail, some high values along both axes are not shown.

Figure 11. Relationships between (a) bulk CN_{TSI}, backscatter, and submicron OA concentrations, (b) APS aerosol concentrations in bins from 0.58-3.0 μm, (c) and altitude, temperature and relative humidity along the ARCTAS-B flight track from 1 July 2008 (shown in panel d), for data within the area boxed in red (shown in higher resolution in panel e) as the airplane moved north. Data are shown as a function of seconds UTC since the start of the date on which measurements began.

Figure 12. Images of the haze layer sampled at ~3-5 km altitude in Fig. 11 from a) on-board aircraft video and b) the DIAL instrument (data from http://science.larc.nasa.gov/lidar/arctas/dial_18.html). Arrows point out the presence of the haze layer sampled in Fig. 11.

Figure 13. Relationships between (a) bulk CN_{TSI}, backscatter, and submicron SO_4^{2-} concentration, (b) APS aerosol concentrations in bins from 0.58-3.0 μm, (c) and altitude, temperature and relative humidity along the ARCTAS-A flight track from 12 April 2008 (shown in panel d), for data within the area boxed in red (shown in higher resolution n panel e) as the airplane moved north. Data are shown as a function of seconds UTC since the start of the date on which measurements began.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7/9/08 a
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1e+03
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1e+01
1e+00
1e-01
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b
APS
UHSAS (QF)
BL SMPS
BL UHSAS
BL APS
Combined
1e+02
1e+00
1e-02
1e-04
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Aerosol diameter (nm)