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.

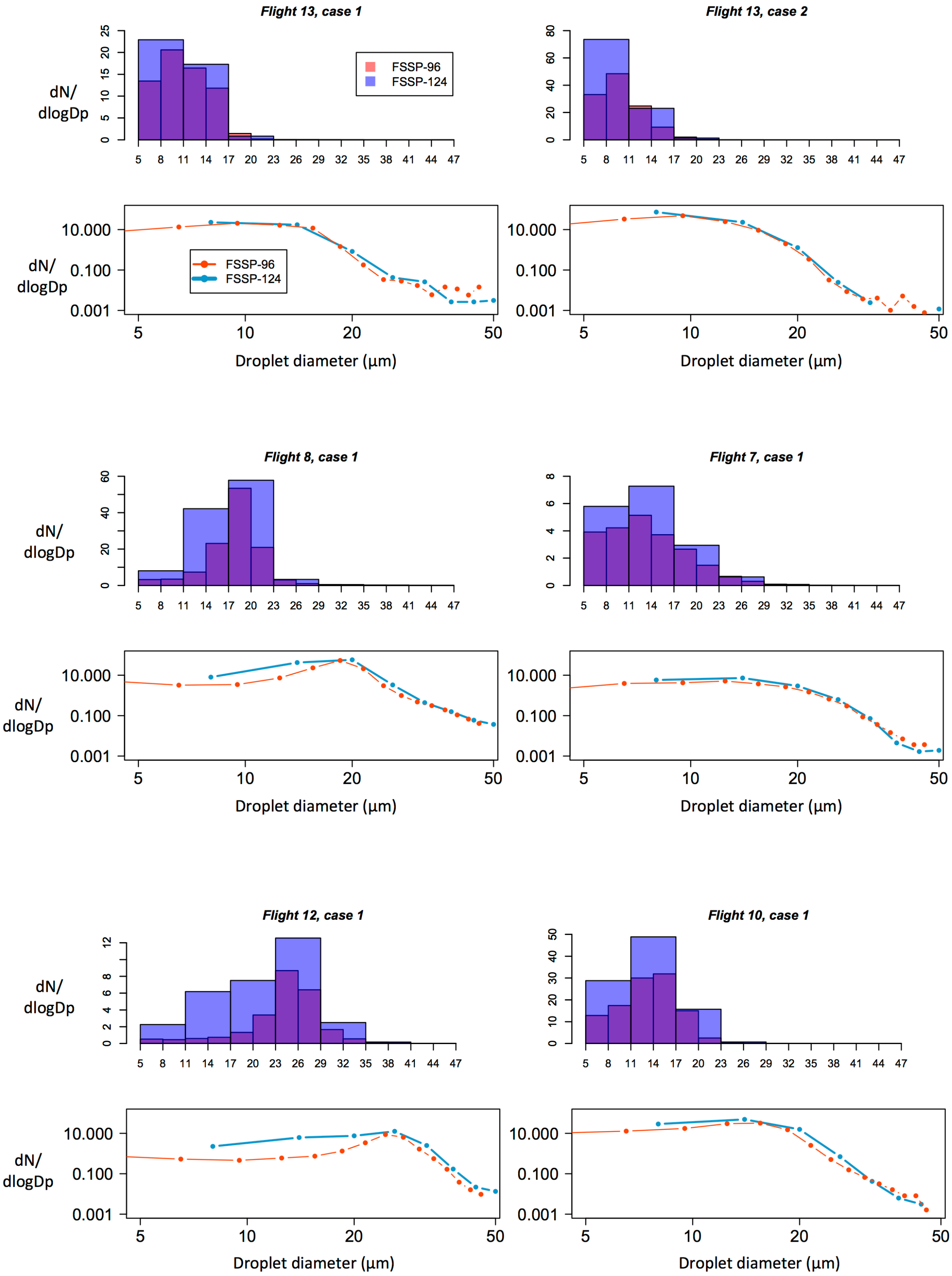

Figure R1. Droplet size distributions (the associated bottom panels containing line plots are the same data on a logarithmic scale) for 6 predominantly liquid clouds in the NRC FIRE.ACE campaign, including the two in this study (from flight 8 and 12). The FSSP-96 tends to have lower droplet numbers than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

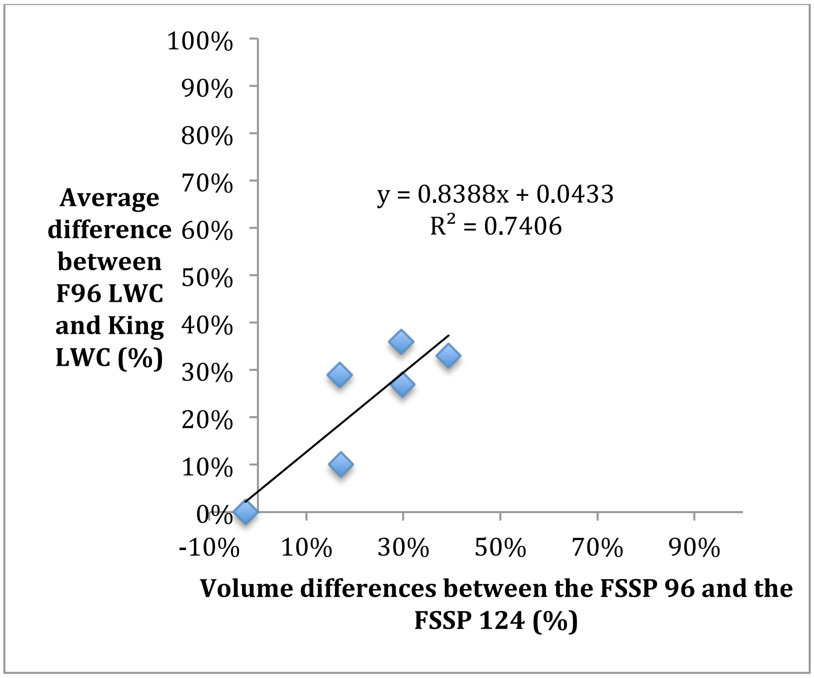

Figure R2. Average difference between FSSP-96 LWC and King LWC for the six clouds shown in Figure R1. Y-axis values were calculated from the difference between a 1:1 slope and the observed slope between FSSP-LWC vs. King LWC values (presented in percentages). If observations produced a 1:1 slope, it 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).

> *Do4ne.*

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”

Done.