# The observed influence of local anthropogenic pollution on northern Alaskan cloud properties

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# Response to reviewers

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*Original Referee comments are in italic*

> manuscript text is indented, with added text underlined and ~~removed text crossed out.~~

We would like to thank the reviewers for their detailed and helpful comments. We revised the manuscript and responded to all of the reviewers' comments. In addition to the reviewers' suggestions, we modified:

- We removed Fig. 4 in order to shorten the manuscript and because it did not contribute to the main focus of the article.
- The OLI data set included some clouds sampled directly after aircraft take-off. Due to their low cloud base (as low as 23 m) they were more likely to be impacted by surface emissions and biased the comparison to NSA where the lowest cloud base sampled was 178 m. Therefore we removed all clouds less than 3 km in distance to the airport which increased the minimal cloud base for OLI to 221 m. We added:

  > Data obtained during take off and landing have been removed to avoid skewing the comparison by sampling aerosols and clouds at much lower altitudes than elsewhere.

- We refer to aerosol observations of the PCASP no consistently to as 'PCASP concentration' and avoid the use of the term 'accumulation mode concentration', because the PCASP does not cover the full accumulation mode size range.

- Several smaller changes, mostly with respect to grammar and wording. Please see the attached pdf of the manuscript with all text changes highlighted for details.

Please note that a companion paper to this study has just been published in ACP Discussions (Creamean et al., 2017).

# 1 Reviewer III

*General comments:*

*This paper is well written, and the figures are mostly easy to understand. I like how they provided the ACI value for their observations, which will be useful for comparison to other locations and aerosol types, and possibly for model parameterization. As best I can tell, the findings are not majorly novel, but it is clear that the authors worked hard to make their findings useful to the community, and there is a lot of good information that would be nice to have available in a publication.*

*However, I do have a few concerns that need to be addressed before I would recommend this paper for publication. There was repeated mention of various differences between the sites, but I was not satisfied with the lack of discussion of the meaningfulness of these differences (see specific comments below). For the reader to understand and evaluate the author's conclusions, more clarifications, appropriate statistical analysis, and/or assessment of errors is needed. I also disagree with the author's interpretation of the influence of oil-field particles on local clouds. However, perhaps I am just misunderstanding something that will become clearer once additional analysis and/or clarification has been provided.*

*Specific comments:*

*Most important comments:*

*1) There were many instances in the text where a better discussion on meaningfulness is needed to support the author's statements. Here are some examples:*

*P5l25: "We manually inspected the vertical profiles of rBC and carbon monoxide (CO), which together constitute a good tracer for biomass burning" rBC and CO together don't necessarily make good tracers for smoke? anthropogenic combustion processes also create those tracers, and photochemistry affects CO concentrations in the Arctic. Can you discuss the uncertainties related to this statement, and its potential impacts on your*

*results?*

We agree that the description was too brief. We extended:

> Therefore, we manually inspected the vertical profiles of rBC and ~~carbon monoxide (CO)~~ CO , which together constitute a good tracer for biomass burning (Warneke et al., 2009, 2010). Typically, these layers are found aloft (Roiger et al., 2015), allowing us to use vertical profiles obtained by the aircraft to aid in their identification. For each spiral obtained at the two sites, elevated layers with CO $\geq$ 0.1 ppmv or rBC $\geq$ 20 ng kg$^{-1}$ were flagged as corresponding to forest fires. Local emissions, on the other hand, are expected to be found in a layer connected to the surface.

We are confident that our approach to remove forest fires from the data set is feasible: As shown in Creamean et al. (2017) and stated in the discussion of Fig 3, we did not find enhanced CO concentrations in the OLI region. Further, rBC values were increased in the OLI region, but the increase was small in comparison to forest fires. Finally, local emissions are expected to be connected to the surface while transported emissions are found aloft. To make this more clear, we added to the discussion of Fig. 3

> The differences between CO and rBC concentrations attributed to forest fires and the concentrations measured in the OLI region show that our approach to use CO and rBC to separate observations impacted by forest fires is feasible .

To make clear that we might have removed too many clouds, we added to the end of the section:

> For the spirals, data identified as originating from forest fire either from manual inspection or according to HYSPLIT, were removed from subsequent analysis. With this approach, we likely removed more clouds from the analysis than required. This, however, ensures that the analysis of the remaining clouds is not biased by influences from forest fires.

*P6l17-19: How meaningful is the difference in skewness at the two sites? Can you demonstrate a statistically significant difference between the skewness, and/or discuss the impact of outliers/sample number/errors, etc.?*

We replaced the violin plots with real histograms so that the reader can see outliers better. In order to test more features than only the skewness, we applied the two sample Kolmogorov–Smirnov test in order to show that the distributions are significantly different. The text has been modified accordingly.

> For both instruments, the distributions within the 90 km circle belonging to each site are skewed towards higher concentrations and the distributions of

both sites are significantly different (1% confidence interval) according to the two sample Kolmogorov–Smirnov (KS) test (Massey, 1951) .

*P6l23-24: If you keep this sentence, please provide more details on how meaningful this difference is (including the amount of difference, if it is statistically significant, and if these numbers are trustworthy given that a) particles of this small size are on the low end of the detectability range for both instruments, and b) that there is typically some error involved in the measurements themselves, and both samplers would ideally be calibrated correctly for these comparisons.)*

We added this quantity to Figure 3 so that the reader can evaluate the difference by him/herself. We also state that the uncertainty might be enhanced:

> Further, the difference between ~~both~~ the two CPC instruments, which ~~depends on~~ equates to the concentration of CN between 3 and 10 nm diameter, is enhanced ~~east of OLI (not shown) .~~ in the OLI region and the distribution is significantly (KS-test) different to the one at NSA (Fig. 3.c). Because this quantity is stemming from the difference in two instruments at the limit of their measurement range, the data is used here only in a qualitative way. Freshly emitted soot has been shown to be larger than ~~this (> 20 nm ), so this range is~~ 15 nm (Zhang et al., 2008), so particles in the 3 to 10 nm size range are likely due to in situ nucleation of aerosol particles from gas phase precursors (i.e., formation of new particles as compared to secondary aerosol formation, where gases condense onto preexisting aerosol, Kulmala et al., 2012). Nucleated aerosols typically have sizes below 3 nm, but quickly grow via condensation and coagulation to sizes $> 3$ nm (Colbeck and Lazaridis, 2014). This source of nucleated aerosol particles from petroleum and gas extraction activities has been reported by Kolesar et al. (2017) for emissions transported from OLI to NSA.

Regarding CPC calibration, we added additional information to the instrument section:

> CPC calibration activities included verifying inlet flow rate with a low pressure-drop bubble flow meter, and determining the size-dependent particle counting efficiency, according to methods defined in Hermann et al. (2007) and Mordas et al. (2008).

*P6l32-P7l2: "In contrast to traces from forest fires, carbon monoxide (CO) concentrations were not found to be significantly enhanced in the OLI region (not shown)." I think some clarification here is needed. Please specify what you mean by traces (tracers?) of forest fires. Do you just mean rBC or are you including CN from the CPC instrument (or MODIS/HYSPLIT information?). Some discussion of the uncertainties in this statement would also be useful. For example, BB smoke can produce large amounts of brown carbon not necessarily detected by a nephelometer; there are many other sources of*

*CN besides forest fires; what CO changes would be constituted as meaningful; were background CO values already high to begin with (which might mask small local changes)?; etc. By "significantly enhanced" do you mean you did some statistical analysis? What was it? In general, I don't think you have presented enough evidence for this statement to be meaningful or useful to the reader yet.*

Regarding CO, we removed the word 'significant'. A detailed analysis of CO is out of the scope of this article, please see Creamean et al. (2017) for more information about CO observatiosn during ACME V and the difference between forest fires and local emissions. Furthermore, we replaced 'traces' with 'air masses'. Regarding black carbon, please note that this quantity was measured with a SP2 which is designed such that it is not impacted by coating of the particles. We modified

> In contrast to ~~traces~~ CO concentrations sampled in air masses originating from forest fires, ~~carbon monoxide (CO ) concentrations were not found to be significantly enhanced~~ low altitude CO concentrations in the OLI region were not enhanced relative to background values (Creamean et al., 2017). The differences between CO and rBC concentrations attributed to forest fires and the concentrations measured in the OLI region ~~(not shown)~~ show that our approach to use CO and rBC to separate observations impacted by forest fires is feasible .

*P7l3-7: How do you know this change is significant? I might have missed it, but I didn't see any information presented on sample number.*

We added the number of observations to Figure 3 and also applied a Kolmogorov–Smirnov test to show the significance of the difference of the distributions. We added

> ... and the difference in the distributions is significant according to the KS-test with 1% confidence interval .

*P7l28: What do you mean by "significant"? If there was a significance test you used to make this statement, what was it?*

We removed the word 'significant'.

*P8l2-3: Some kind of statistical analysis would be helpful here? at minimum are the differences in means significant? Also, a "shift" implies movement from one state to another, and you haven't presented the information to show that movement occurred. Instead, what you show is that there is a secondary peak at NSA that is not present at OLI. My opinion, based on the information provided, is that one can only speculate on the reasons for this difference and not assign a cause/mechanism.*

We reworded the sentence to replace the word "shift":

When comparing 2D histograms of liquid effective radius and liquid water content for OLI and NSA (Fig. 4, a, b), ~~a shift towards~~ OLI values are shown to feature smaller $r_{eff}$ ~~can be clearly seen in the measurements obtained in close proximity to OLI~~ for the same LWC.

In order to show for what range of LWC values the difference of reff is significant, we added the new Figure 5:

Fig. 5.a reveals that this difference is statistically significant for most LWC > 0.1 g $m^{-3}$ according to a Welch's t-test (Welch, 1947) with a 5% confidence interval (Note that a 5% confidence interval is always used in this study unless noted otherwise). [...] For TOI, the mean $r_{eff}$ is significantly different than those at NSA for LWC > 0.02 g $m^{-3}$.

*P10l3-6: "It can be seen that C is decreased at OLI in comparison to NSA by up to one order of magnitude for constant LWC and reff." Since sample numbers are kind of hard to extract from Fig. 5, it might be helpful to mention the sample numbers provided for this comparison.*

Please see the caption of Fig 4 for the total number of observations, we also added that information to the plot title. We think that with the updated colormap regions with e.g. less then 1% data (corresponding to 9-16 samples) can be easily identified.

*Also, what are the errors in C to begin with? Perhaps it would be more appropriate to compare the overall site values of C, instead of single bin values to reduce random errors and biases from samples from a single cloud dominating one of the bins. At minimum, the authors should qualify this statement with the uncertainties involved. One suggestion is to move Fig 6 to the supplement, and just present mean differences between the sites in the main text. For the following sentence, "Interestingly, differences in C are largest for reff smaller than 16 µm" again, what is the statistical basis for this statement? To me, it looks like all the samples ¿ 16 um have a very small sample number for comparison.*

Regarding the uncertainty of $C$, we added:

Note that our approximation does not consider the impact of turbulence and droplet charge on $C$. This might lead to considerable uncertainties, which have—to the authors' best knowledge—not been fully quantified. Because we are interested how $C$ is modified in the OLI region, we ~~show the difference~~ focus on the ratio of $C$ ~~between both sites in~~ determined at NSA and OLI which should reduce the uncertainty of $C$. Fig. 6 shows the ratio between NSA and OLI of $C$ as a function of $r_{eff}$ and LWC.

Regarding the Figure, we decided to keep it in the manuscript, because a comparison of overall site values for $C$ would not work given that $C$ spans easily more than 10 magnitudes depending on LWC and reff (see text). Therefore, a general comparison would

be dominated by differences of LWC and reff. However, we agree that the statement regarding $C$ for reff > 16 um is based on poor statistics and removed it.

*P10l11-12: "C is reduced by 1 to 1.5 orders of magnitude at OLI. The offset is surprisingly constant for LWC larger than 0.01 g m-3 " Can you trust the data at LWC < 0.01 g m-3? Other studies define LWCs below that value as not even being in-cloud (e.g., Matsui et al. (2011)). Do you have statistical basis for this statement?*

In accordance with other studies (e.g. Lance et al., 2011; Hobbs and Rangno, 1998), we decided to use the number of drops for cloud definition, mostly because this is the quantity directly observed by the cloud probes. Our threshold of 10 drops per cm3 (which was criticized as too high by the other reviewer) translates for our data set into a minimum LWC of approx 0.001 g/m3. In order to show the full data set, we decided to show LWC values as little as 0.001 g/m3. However, we agree that we have to explain that and added to the discussion of Fig. 4:

> Note that LWC values below 0.01 g $m^{-3}$ are defined as not in-cloud by some studies (e.g., Matsui et al., 2011; Leaitch et al., 2016), but we decided to show the full data set because the in-cloud definition used here (> 10 droplets $cm^{-3}$) can result in LWC as low as 0.001 g $m^{-3}$ and we wanted to make sure that all cloud data points are included in the analysis.

*P12l21-22: "since no enhanced PCASP particle concentrations are found to be correlated to droplet sizes in the emissions-impacted 9 to 12 µm range (unlike for rBC and CN), there is no indication that local emissions are directly altering liquid clouds to have smaller reff as a result of PCASP-sized particles." Since no correlation was actually shown, the authors should either show the correlation or its relevant statistics, and discuss its meaningfulness, or they should use a different word and present some discussion on the statistical basis for the new statement.*

We reworded that sentence:

> The fact that the response of $r_{eff}$ to PCASP aerosol concentrations ~~is very similar~~ is—for constant LWC—almost monotonic for both sites is likely because the PCASP covers the aerosol size range most relevant to droplet nucleation ~~. It should be noted that the monotonic decrease in PCASP concentration with increasing droplet size~~ and is consistent with the first indirect effect. ~~However, since no enhanced PCASP particle concentrations are found to be correlated to droplet sizes in the emissions-impacted 9 to 12 $\mu$m range (unlike for rBC and CN), there is no indication that local emissions are directly altering liquid clouds to have smaller $r_{eff}$~~ A different behaviour would indicate that clouds react differently to the same PCASP concentration (which covers most of the accumulation mode size range, see also Sec. ~~as a result of PCASP-sized particles. Even~~ 6).

We added a new Figure (12) to the manuscript in order to discuss the impact of PCASP concetration on cloud properties better:

> Analysis of the relationship between clouds and HYSPLIT concentrations, rBC and CN shows that there are some, but not all, clouds at OLI impacted by local pollution. Because enhanced rBC and CN concentrations are expected to be good indicators of anthropogenic activity, they are used to isolate clouds impacted by anthropogenic emissions. Clouds, whose mean below-cloud rBC or CN concentration is above the median concentrations shown in Fig. 3 (4.1 ng $kg^{-1}$ and 1122 $cm^{-3}$, respectively), are identified as potentially impacted. When using this criterion, 10 of 24 (3 of 16) clouds at OLI (NSA) are identified as potentially locally influenced (Fig. 12). For NSA, two of the three clouds corresponded to either extremely low CN ($< 20$ $cm^{-3}$) or rBC ($<$ 1 ng $kg^{-1}$) values, making a connection to anthropogenic activities unlikely. But, the clouds classified as anthropogenic at OLI correspond mostly to enhanced concentrations of rBC (Fig. 8) and CN (Fig. 10) and mid-sized $r_{eff}$ (5 - 15 $\mu$m). Fig. 12 shows how the PCASP concentrations of the potential locally affected clouds compares to the clouds classified as affected by forest fires (these clouds are removed in all other Figures except Fig. 14) and the remaining, non-classified clouds referred to as 'other'. Note that forest fire emissions were also present in the vicinity of NSA, but cloud measurements from these time periods did not pass the quality control measures implemented (continuously ascending or descending profiles). It is striking that the clouds classified as associated with forest fire have a significant (t-test), 6-times larger linear mean PCASP concentration than the clouds classified as locally affected at OLI (510 vs 80 $cm^{-3}$). Despite this big difference, clouds classified as locally affected still feature PCASP concentrations significantly larger than the clouds classified as other (35 $cm^{-3}$). We conclude that CN and rBC particles, which were used to classify local clouds, have the potential to grow to accumulation mode particles measured by the PCASP. For NSA, however, the mean PCASP concentration for clouds classified as other is not significantly (t-test) different from the clouds classified as locally affected at OLI. This is also true when including the three clouds classified as locally affected (from which only one is potentially local as discussed above). This is consistent with the findings of Fig. 3 which shows that the general PCASP concentration background is enhanced at NSA in comparison to OLI.

*For general discussion of Figs. 8-11: What is the influence of random error? Was there bias, e.g., from a single outlier cloud containing all the values in one or more of the bins, etc.?*

We agree that Figs. 8-11 are potentially affected by outliers. Consequently, we reduced the size of the bins such that most pixel show only observations of a single data point. By this, the trend and the variability of the data can be seen. We added:

> The bin sizes in Fig. 7 were reduced in order to investigate the variability between clouds. This reveals that only a subset of clouds is associated with local pollution according to HYSPLIT.

*P16l7-8: Does the statement that, "collision-coalescence and precipitation rates are reduced by up to two orders of magnitude around OLI" still hold after careful consideration of the impacts of sample number and potential biases?*

We applied a t-test to show for which LWC values the difference is significant:

> Doing so reveals that, for constant LWC, $\bar{C}$ is reduced by 1 to 1.5 orders of magnitude at OLI. The offset is significant and surprisingly constant for LWC larger than 0.01 g m$^{-3}$. [...] Averaging over all $r_{eff}$ enhances the effect and leads to differences of up to two orders of magnitude for $R$ as a function of LWC. This effect is statistically significant for LWC $>$ 0.02 g m$^{-3}$.

Please see above for a discussion of uncertainties of $C$.

*2) I did not agree with the author's 3rd conclusion (on page 17), that local emissions affect cloud properties. Their arguments were as follows:*

*a) "HYSPLIT simulations show that 62% of all cloud observations around OLI can be traced back to local emission sources." First, I am not clear on what was meant by "cloud observations around OLI can be traced back to local emission sources" (see specific comment #17 below). I do agree that OLI is more likely to contain oil-field related particles than NSA, based on proximity and the rBC, CN, and HYSPLIT analyses the authors presented. However, this argument alone is not enough to indicate that oil-field related emissions are impacting clouds. The authors present evidence to show that higher CN at OLI is due to elevated small (<100 nm) particle concentrations, and they mention that these particles are likely too little to act as CCN. As the authors mention, rBC is not necessarily a good indicator of CCN either. Moreover, the range of below-cloud PCASP aerosol concentrations, which are in the size ranges where droplet nucleation would be more likely, were similar between sites (Fig. 4).*

Thanks for pointing this out. Please see your comment 17 for a response with respect to HYSPLIT.

We added a new figure (12) to show how PCASP particles can change cloud properties, see above for discussion. We also added a new panel to Figure 3 showing that the increase in CN is particularly related to particles in the 3 to 10 nm range. We also added a section describing how sub-100 nm particles might impact cloud properties to the discussion of Fig. 14:

> In addition, some data points lie above the 1:1 line which might indicate that particles smaller than the PCASP size range (i.e. $<$ 100 nm) are acting as

CCN (Leaitch et al., 2016). Further, the assumption that the below-cloud aerosol properties govern the cloud microphysical properties might not be true for all clouds depending on sub-cloud vertical mixing. Therefore, we identified all clouds where the above-cloud PCASP concentration is larger than below-cloud (red dots in Fig 14), and indeed half of these clouds are above the 1:1 line. When using the above-cloud concentration for these clouds, only two of these clouds are above the 1:1 line. However, there are still 11 more clouds above the 1:1 line. Since these clouds generally feature PCASP concentrations < 50 $cm^{-3}$, the fact that they are above the 1:1 line could be related to increasing sampling errors for small concentrations. They may also confirm the finding of Leaitch et al. (2016) that aerosols below 100 nm can act as CCN for thin clouds.

*b) "Reduced reff (between 9 and 12 μm) of OLI clouds correspond to increased CN and rBC concentrations" Convincing supporting evidence for the validity and meaningfulness of this trend has not yet been presented (see specific comment #1).*

See response to specific comment #1 above.

*c) "the mean size of cloud-associated rBC particles is smaller at OLI which is consistent with freshly emitted, less aged particles" There were smaller particles at OLI, and these small particles probably were less aged, but don't the CN and PCASP data indicate that these particles are likely too small to act as efficient CCN?*

We removed that argument because it was actually wrong: Because the SP2 does not measure the coating of black carbon particles, smaller particles as measured by the SP2 do not indicate fresher emissions. However, they indicate a different source:

~~This is consistent with aging of rBC during atmospheric transport, and supports the idea that rBC measurements around OLI are associated with local emissions from Prudhoe Bay and not transported fire emissions.~~ rBC can originate from biomass burning as well as anthropogenic sources, but particle size is smaller for the latter (Schwarz et al., 2008).

In addition, we added that sub-100 nm might also impact cloud properties as discussed above.

*d) "clouds [at OLI] were found to be frequently connected to enhanced CN and accumulation mode concentrations" I think the authors meant "associated with" instead of "connected to"? But again, the presence of smaller particles at OLI does not necessarily indicate that they are participating in cloud processes. As I mentioned, the range of below-cloud PCASP aerosol concentrations, which are in the size ranges where droplet nucleation would be more likely, were similar between sites (Fig. 4). As the authors mentioned in the text, particles <100 nm in diameter are likely to be less efficient CCN. In summary, there wasn't convincing evidence that oil-field aerosols did have a discernable*

*impact on cloud properties.*

We changed the wording as suggested and removed "connected to".

Please see above for the more general issues raised. We modified the conclusions accordingly:

> Finally, ~~while no enhanced concentrations of larger accumulation mode particles were observed for OLI~~ the clouds identified as most likely influenced by anthropogenic activities have significantly higher PCASP concentrations for OLI than for the remaining clouds (Fig. ~~3), clouds there were found to be frequently connected to enhanced CN and accumulation mode concentrations~~ 12). However, the PCASP concentration of local clouds is not significantly higher than at NSA which might be related to a higher background of PCASP particle concentrations at NSA.

*More minor comments (not in order of importance):*

*3) P2l23: It might be helpful to note upfront that liquid clouds are not that common in the Arctic, compared to ice and mixed phase clouds?*

While the reviewer is right that there are more mixed phase and ice clouds year round, low altitude liquid clouds are frequent in summer as temperatures are typically above freezing in Northern Alaska. We added to the introduction of the data set:

> During ACME-V, 156 (60%) of the 258 vertically sampled clouds were classified as liquid (see below for thresholds), showing that liquid clouds are frequent during the summer time in Northern Alaska.

*4) P5l23: "While Arctic Haze was not observed during ACME-V, transported emissions from forest fires can contribute significantly to summertime aerosol loading in the Arctic?" Earlier Arctic haze was defined as long-range transport of aerosol particles from lower latitudes. Here it seems like the long-range transport of smoke is excluded from consideration as Arctic haze. I think it would help to clarify this sentence, especially since smoke does contribute to what most people define as Arctic haze (e.g., Warneke (2010)). Also, how do you know that what you are referring to here as Arctic haze (Anthropogenic pollution from lower latitudes? Long-range aerosol transport?) was not observed during ACME-V? Is this statement based on the fact that typical precipitation and air mass transports during the summer season make the presence of long-range aerosol transport less likely, or on some extra analysis you did to determine this? Please specify.*

We modified that statement, because long range transport was indeed present during ACME-V as found by Creamean et al. (2017), but it will likely not impact our analysis. However, we decided not to use the term 'Arctic Haze', because it is usually related to winter and spring time long range transport. For this, we added a section to the end of

the paragraph and also state that we treat long range transport separately from forest fires.

> For ACME-V, Creamean et al. (2017) classified only four flights as impacted by long range transport from lower latitudes not related to forest fires. During these flights, only a single cloud was sampled in the vicinity of OLI or NSA which had one of the lowest aerosol concentrations measured in the whole data set. Therefore we are confident that our analysis is not strongly impacted by this kind of long range transport events.

*5) Last paragraph of section 2: This paragraph is a little confusing, and it would help if the authors were more clear about their methods. What was the order and/or priority of the steps to ID long-range smoke transport? From where were the CO data obtained? It says the vertical profiles of rBC and CO were manually inspected ? but what were the criteria relating to those parameters, and what were those criteria specifically used to do? Were the 5 locations where HYSPLIT back trajectories were obtained chosen based on MODIS fire locations?*

We extended the paragraph to make the methodology more clear:

> Transported emissions from forest fires can contribute significantly to summertime aerosol loading in the Arctic ~~(Law and Stohl, 2007)~~ (Law and Stohl, 2007); (Creamean et al., 2017) . Therefore, we manually inspected the vertical profiles of rBC and ~~carbon monoxide (CO)~~ CO , which together constitute a good tracer for biomass burning (Warneke et al., 2009, 2010). Typically, these layers are found aloft (Roiger et al., 2015), allowing us to use vertical profiles obtained by the aircraft to aid in their identification. For each spiral obtained at the two sites, elevated layers with CO $\geq$ 0.1 ppmv or rBC $\geq$ 20 ng kg$^{-1}$ were flagged as corresponding to forest fires. Local emissions, on the other hand, are expected to be found in a layer connected to the surface. Note that the data impacted by forest fires were only removed for spirals above OLI, NSA, and TOI. For clear-air observations during level flight legs between sites, it is generally impossible to determine whether a layer is connected to the surface or elevated. [...] For the spirals, data identified as originating from forest fire either from manual inspection or according to HYSPLIT, were removed from subsequent analysis.

Regarding MODIS, the five locations chosen were based on the general location of fires detected from MODIS. For the daily HYSPLIT simulations, the five source points were turned on or off, depending on the fire activity in that area for that day, as indicated by MODIS. We modified

> . These sources were toggled on or off on a daily basis in correspondence to thermal anomaly observations in the corresponding region (see Fig. 5 of

Creamean et al. (2017) ) from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Aqua and Terra satellites obtained using brightness temperature measurements in the 4 and 11 $\mu$m channels (Giglio et al., 2003; Giglio, 2013).

*6) Fig1: Maybe include the MODIS fire locations?*

This is an excellent suggestion. However, we decided only to show the locations of the fires assumed in HYSPLIT in order to avoid cluttering of the Figure. See Figure 5 of Creamean et al. (2017) for a map of the forest fire locations used.

*7) P6l11-12: "The data presented are limited to observations obtained below 500 m in order to demonstrate the impact of local emissions and reduce the impact of forest fires." Up to this point in the text, the reader has not been presented with any strong evidence that smoke (or long-range anthropogenic pollution, for that matter) was not present below 500m. To avoid confusing the reader, please provide this information.*

Thank you for pointing this out. We modified the text:

~~The data presented~~ . As discussed above, removal of data potentially impacted by forest fires is only possible for the spirals. Therefore, the data presented in Fig. 3 are limited to observations obtained below 500 m ~~in order to demonstrate the impact of local emissions and reduce the impact~~ , because transported emissions of forest fires were typically at higher altitudes during ACME-V (for a detailed study of aerosol properties during ACME-V see Creamean et al. (2017)) .

*8) P6l29-31: It would be helpful to use consistent units here to aid comparisons with the data in this study. Also, the rBC concentrations referenced from Zamora et al., were for background air masses specifically thought to not be influenced by smoke (see their Table 6). From their Figure 7, smoky rBC values ranged somewhere between the order of 1-103 ng/m3 (average appears on the order of 102 ng/m3). Also, the majority of rBC values presented in Figure 3 are much below the median 20-30 ng/kg values from Roiger et al. So what I interpret the rBC from Figure 3 to mean is that the region was very clean with respect to rBC, except near OLI.*

We agree with the reviewer and modified the section.

rBC background concentrations appear to be similar to ~~values found by Zamora et al. (2016) (1-16 ng/m3) and Roiger et al. (2015)(median 20-30 ng /kg) for summertime transported forest fire plumes in the Arctic. Other studies (Warneke et al., 2009; Schwarz et al., 2010) found up to one order of magnitude higher rBC concentrations in the Arctic which is more similar to the maximum values we observed around OLI~~ background observations made by Zamora et al. (2016) and Roiger et al. (2015). It should be noted that

> emissions related to forest fires led to concentrations as high as 600 to 1000 ng $kg^{-1}$ during ACME-V (mostly at altitudes above 500 m, Creamean et al., 2017) which were also observed in other data sets (Warneke et al., 2009); (Schwarz et al., 2010); (Zamora et al., 2016). Consequently, the emissions from anthropogenic sources in the OLI region are about a magnitude lower

Regarding the Roiger paper, the 20-30 ng/m3 values correspond to mean values connected to forest fires, i.e. they are not background values. Their Fig 12a, blue dashed line shows their background values are actually very similar (< 5ng/kg) to ours.

Regarding the Zamora paper, we confirmed with the corresponding author of that study that Fig 7 shows data in the range between 2 to 1235 ng C/m3 (and not $\mu$g C/m3 as stated in their caption) which is consistent with our findings.

*9) P7l7-13: Your expectation that that there might be more smoke influence at the NSA site would be consistent with literature observations of general smoke particle sizes being larger than the PCASP minimum size range of 100 nm (e.g., Kondo et al. (2011); Sakamoto et al. (2015)). However, I am confused by your statement that, "An alternative explanation could be ... more aerosol processing by precipitation (e.g. Hoppel et al., 1990)." I believe that the Hoppel et al. reference refers to non-precipitating cloud processing of aerosols, correct? So do you mean "processing by clouds" instead of "processing by precipitation"? In which case, why did you mention collision-coalescence and precipitation being the causes for the differences? Or did you mean "removal" or "scavenging" here instead of "processing" (in which case why the subsequent sentence regarding aerosol in-cloud processing)? Other readers might be confused as well, so clarification here would be helpful.*

We added the suggested references

> Transported forest fire aerosols are often larger than the PCASP detection threshold of 100 nm as shown by Kondo et al. (2011) and Sakamoto et al. (2015).

Regarding the study by Hoppel, the reviewer right that we picked the wrong reference by accident. We replaced the reference with Feingold et al. (1996)

*10) P7l18: "While mean particle size generally increases with decreasing CN concentration, ?" Looking that figure 4, this is not clear to me. I suggest taking this statement out, since it doesn't seem that necessary. Or, if you disagree, you could try plotting on a log-scale to see if the pattern emerges more clearly or switching the x and z axes to show the trend better?*

Because it did not belong to the main focus of the article, we removed the figure and the corresponding statement.

*11) P7l19-20: "the variability of PCASP mean size is rather low for CN concentrations > 600 cm-3 , which is consistent with the idea that particles have already experienced growth" Are there other things that might cause the same trend? Might be worth mentioning.*

Same as above: we removed the figure and the statement.

*12) Fig.4: why was the y-axis cut off at 4000? To provide aerosol context for the cloud data? To provide better context for the study as a whole, you might consider plotting on a log scale to include all the data, or at least mentioning in the caption that a fraction of the data are excluded for whatever your reason.*

Same as above: we removed the figure and the statement.

*13) P9l6-7: "It is not possible to study the cloud life cycle using aircraft in-situ observations, but the potential for impact on cloud life cycle can be estimated by?" I suggest moving your qualifiers in the last paragraph of this section up here so that you can better clarify upfront that what you are estimating is a factor important to cloud lifetime, but your estimate is not necessarily indicative of cloud lifetime itself.*

We followed the reviewer's suggestion and changed the order.

*14) P10l4-5: "This is caused by reduced broadening of the drop size distribution towards large drops at OLI (not shown), consistent with cloud chamber experiments (Gunn and Phillips, 1957)." Maybe I am not understanding something, but shouldn't that kind of broadening be apparent in Fig. 5? Why not mention that here? Also, what specifically is consistent with this cloud chamber experiment?*

Figure 5 (now 4) shows only the effective radius which does not provide any information about the breadth of the drop size distribution. Regarding the cloud chamber experiment, we decided to keep the reference for historical context, but extended:

> This is caused by reduced broadening of the drop size distribution towards large drops at OLI ( ~~not shown~~ Fig. 5.b ), consistent with ~~cloud chamber experiments (Gunn and Phillips, 1957)~~ the experiments of Gunn and Phillips (1957), who produced similar results when ingesting polluted background air into their cloud chamber.

Please see the new Figure 5.b for information on the breadth of the distribution.

*15) P10l9: "typical reff values are reduced at OLI in comparison to NSA for the same LWC." I don't see that consistently in Fig. 5. What about at high LWCs, for example? Did you mean within a specific LWC range? If so, please specify.*

We added a new Figure 5a to highlight the relevant LWC range and to apply a statistical test:

> Fig. 5.a reveals that this difference is statistically significant for most LWC > 0.1 g $m^{-3}$ according to a Welch's t-test (Welch, 1947) with a 5% confidence interval (Note that a 5% confidence interval is always used in this study unless noted otherwise).

*16) P10l13: more info on this droplet fall velocity parameterization would be helpful.*

A detailed description would be out of the scope of the paper and parallel the original publication. However, we added:

> ... which provides a continuous solution over the entire drop size range in dependence of the Best and Reynolds number

*17) P11l6: "For OLI (NSA), 62% (16%) of all ACME-V cloud observations can be traced back to surface emissions originating from the Prudhoe Bay oilfields." I am confused here. How did were these values calculated? What does it mean that a certain fraction of cloud observations were traced back to surface emissions? That some fraction of the aerosol particles in the air masses containing the clouds were likely to have originated from the oilfields? Please clarify in the text.*

Thanks for pointing this out. We clarified:

> For OLI (NSA), ~~62~~ 50 % (16%) of all data points observed within clouds during ACME-V ~~cloud observations~~ can be traced back to surface emissions (i.e. mass concentration > 0 according to HYSPLIT) originating from the Prudhoe Bay oilfields.

Note that the reduction from 62 to 50% is due to the exclusion of extremely low clouds at OLI as discussed above.

*18) Fig. 7 and text citing it: It is not clear to me how one can get the mass concentration at a certain location unless the emissions at the point source are known. Is this a relative mass concentration? Was emissions information available? If so, please provide it. Also, what are the errors in the co-location of the modeled plume and the actual plume? Please discuss. If you don't have this information, I recommend removing this figure and just stating in the text that HYSPLIT back trajectories indicate a higher influence of oil field aerosols at OLI than at NSA x% of the time, and that one would expect higher oil field aerosol concentrations at OLI than at NSA due to proximity of the source. Figs. 8 and 9 probably provide more trustworthy information on aerosol concentrations anyways (please correct me if I am wrong).*

Because HYSPLIT provides a relative unit (mass per volume), we do not have to know the emission rate. We added to the end of the paragraph:

> Note that HYSPLIT provides only a relative emission rate in 'mass $m^{-3}$'

because the actual emission rate in the Prudhoe Bay region is unknown.

We decided to keep the Figure, because it also shows the variability for similar reff and LWC due to the smaller bins. We agree that HYSPLIT uncertainties might be high:

> Note that given the coarse resolution of the forcing model (1°) and the complexity of the Arctic boundary layer, HYSPLIT is used here only in a qualitative way, and not to select locally impacted clouds.

*19) P16l8-9: Regarding the statement that, "As a consequence, the breadth of the size distribution of liquid droplets has to be smaller at OLI", why not just provide the size distribution data from the campaign?*

The size distribution depends strongly on the LWC. Therefore we decided to show the mean standard deviation of droplet size as a function of LWC in the new Figure 5.b.

*20) p. 17l10-12: "While forest fire cases have typically higher aerosol concentrations and consequently droplet concentrations, their inclusion into the estimation of ACI does not substantially alter the found relationship." I disagree with this statement. Based on the arguments from specific comment #2 above, the authors admission that they might not have been able to completely exclude smoke influenced air masses from the study, and the similar ACI values for smoke influenced cases, for all we know, smoke was driving the relationships? or did I miss something?*

We are not sure whether we understand the reviewer here properly. We updated Figure 14 to show that clouds potentially impacted by local emissions are between forest fires and the remaining clouds with respect to aerosol concentration. However, the sample size is too small to estimate ACI for them separately. We added:

> As already discussed, clouds associated with local emissions have lower PCASP (and likely accumulation mode) concentrations than forest fires, but still have larger concentrations than for the other clouds.

We added to the end of the section that the goal is only to investigate whether there are differences in the mechanism how aerosols change cloud properties.:

> Therefore, we conclude that a ~~significant difference of~~ difference in ACI between local emissions and forest fires cannot be found , given the limited data set. This refers only to the mechanisms through which aerosols change cloud properties, and does not imply that local emissions do not change cloud properties.

We also added the standard error of ACI obtained by the fit in order to show that the uncertainty is larger than the difference.

*Technical corrections:*

*P3l15: You cite a 1993 paper for 2015 observations. Please clarify what you meant by citing this paper here.*

This is not a paper, but a data set. We modified the reference to make this more clear.

*P6l15: Just a suggestion, get rid of "For the SP2", since people may not remember what SP2 is, and it is not necessary to the sentence.*

Changed to from the SP2 probe.

*P6l20: Suggest combining this sentence with the following sentence to make it clear what specific pattern(s) is/are being deemed similar (the skewness and spatial distribution as opposed to the median?).*

We modified the sentence to indicate it is about spatial patterns:

> CN measurements from the CPC show a ~~similar pattern~~ spatial pattern similar to the SP2 even though the increased values are distributed over a larger area

*P6l2-4: The units in this sentence are confusing. Also, can you clarify in the manuscript that the latter two numbers are for wet deposition scavenging (or some other kind of scavenging)?*

We agree that the units are confusing, but they are in accordance with the HYSPLIT manual. The numbers are indeed for wet deposition scavenging, we added this information to the text.

*P8l1: suggest rewrite to "As mentioned above, clouds known to be impacted by forest fires have been removed" to better reflect the uncertainty you discussed in the previous section.*

Thanks for the suggestion, changed.

*Fig. 5: please describe in the caption what the dashed line indicates. Also suggest adding in the term "NSA", since that is what you talk about in the text referring to this figure. Also, I found the label "number of observations [%]" to be very confusing. I recommend changing this to something like, "% of total observations."*

Good suggestions, we modified the figures and the caption accordingly.

*Fig.6: The caption says OLI-NSA, but the figure says NSA-OLI? Also, can you make the axes consistent between Fig. 5 and 6? That would help enable comparisons between the two. Lastly, in the caption it says that, "The green dots highlight data points with less than five observations." Please clarify in the caption whether those 5 observations include points at both sites added together, or at each site individually.*

Thanks for noting the wrong caption and varying axes, we fixed that. The dots indicated less that 5 observations for one of the sites. However, we use the dots now to indicate whether the difference is significant or not.

*Suggest using site abbreviations throughout the paper and in the figures after they have been defined, instead of using a mix of the site names and the abbreviations to avoid confusion.*

Excellent suggestions, we modified the figures.

*P12l5: Please clarify: what "this" do you refer to when you say, "This means?"?*

We replaced 'This means that' with 'As a consequence'

*Fig 12: concentration not cooncentration on the x-axis.*

*P17l19: from not form.*

Both fixed, thanks.

# 2 Reviewer III

*The authors use airborne observations from June-September, 2015 within 90 km of two DOE-ARM sites (North Slope Alaska or NSA and Olitok Point or OLI) to study potential effects of changes in aerosol particles on Arctic liquid water clouds. The aerosol observations are limited to physical measurements with a PCASP (particles larger than about 100 nm) and a condensation particle counter (CPC; particles large than about 3 nm as well as measurements of black carbon (rBC). The main objective is to see if clouds formed on particles from anthropogenic activities in the OLI area result in differences in cloud microphysics compared with cloud formed on particles observed in the NSA area. The topic is relevant for ACP, the paper is well organized and interesting, and I believe the overall results are useful. However, the paper is not currently ready for publication.*

*Major comments:*

*1) The paper leads up to section 6 (Quantification of cloud aerosol interaction) by accumulating information suggesting the microphysics of the OLI clouds are impacted by local emissions. That notion is then set aside in section 6 based on the calculated ACI indices. However, the ACI calculation in inappropriate for these observations if particles smaller than 100 nm are nucleating droplets. Drawing a 1:1 line in Figure 12 indicates some points above. Assuming the measurements are reasonably accurate, which the authors do not discuss, then relatively few droplets are nucleated on particles smaller than the lower limit of the PCASP. The larger deviations above the 1:1 line are towards lower aerosol*

*concentrations, which would be consistent with Leaitch et al. (ACP, 2016) if there are sufficient particles smaller than 100 nm to explain the deviations. As it stands, the ACI discussion tells us only that there is some impact of PCASP-sized particles on the NSA cloud observations, which has already been mentioned in connection with Figure 10 and is not the focus of the paper. The fundamental result could be more clearly shown using Figure 12 with straight concentrations rather than natural logarithms. What do Figures 8, 9, 10 and 11 tell us that cannot be found from a modified Figure 12?*

It was not our intent to set the notion "local emission impact cloud properties" aside with Fig. 12 (now 14) and we think that this is not supported by the measurements. The intent of the ACI discussion is to investigate how the mechanisms connecting PCASP concentrations to clouds are different when connected to local and forest fire emissions. To make this more clear, we added

> Therefore, we conclude that a ~~significant difference of~~ difference in ACI between local emissions and forest fires cannot be found , given the limited data set. This refers only to the mechanisms through which aerosols change cloud properties, and does not imply that local emissions do not change cloud properties.

We agree that the argument on how local emissions change cloud properties was not convincing. Therefore, we identified clouds potentially impacted by local emissions and show that they have larger aerosol concentrations at OLI than the remaining clouds (Fig 14). The new Figure 12 discusses that in greater detail. Moreover, we agree that sub-100 nm particles could have impacted the analysis:

> In addition, some data points lie above the 1:1 line which might indicate that particles smaller than the PCASP size range (i.e. $<$ 100 nm) are acting as CCN (Leaitch et al., 2016). Further, the assumption that the below-cloud aerosol properties govern the cloud microphysical properties might not be true for all clouds depending on sub-cloud vertical mixing. Therefore, we identified all clouds where the above-cloud PCASP concentration is larger than below-cloud (red dots in Fig 14), and indeed half of these clouds are above the 1:1 line. When using the above-cloud concentration for these clouds, only two of these clouds are above the 1:1 line. However, there are still 11 more clouds above the 1:1 line. Since these clouds generally feature PCASP concentrations $<$ 50 cm$^{-3}$, the fact that they are above the 1:1 line could be related to increasing sampling errors for small concentrations. They may also confirm the finding of Leaitch et al. (2016) that aerosols below 100 nm can act as CCN for thin clouds.

*2) Concerning Figure 6 and related discussion on page 10, I have the following questions and remarks:*

*a) The differential collection growth rates (OLI minus NSA) range between 0.1 and 1*

*(units of kg/sm3). The mean collection growth rates vary from less than 1e-12 to about 5e-6 with the same units. How can the differential rates be higher than the mean rates? Are the differential values ratios (e.g. OLI-NSA/NSA) rather than absolute values?*

Thanks for pointing this out, the difference is actually in log scale, i.e. the plot shows actually a ratio. We updated figure and text accordingly.

*b) On lines 3-4, you say that C is lower at OLI compared with NSA for constant LWC and Reff. This is very difficult to see in 6a. Regardless of whether the differentials (OLI-NSA) are absolute or ratios, they are higher, not lower. Also, how do I look at constant LWC and Reff in these plots?*

Sorry, we missed to update the Figure caption as we swapped OLI and NSA last minute, it is NSA/OLI. The values are larger for NSA as stated in the text correctly. We changed the caption accordingly. The axes of the plot are LWC and reff, therefore every pixel of the plot is for constant reff and LWC (or at least for a small interval).

*c) On lines 10 and 11, you say that the mean value of C (averaged for Reff) is 1 to 1.5 orders of magnitude reduced at OLI for a constant LWC. Yet the mean OLI growth curve lies to the right of the NSA curve, which indicates a higher mean collection growth rate.*

We apologize, when doing the last minute swap of the sites, we also overlooked to update the legend. The description in the text is correct, we updated the figure legend.

*d) The same apparent discrepancies are present for the rainfall rates in Fig. 6b.*

Same as above.

*3) The aerosol observations are restricted to below 500 m-msl. Please indicate the altitude range for the cloud observations. Please indicate how you know that the aerosol below cloud was connected with the cloud above and not isolated by temperature inversions, which can happen in the relatively stable environment of the Arctic.*

This is a misunderstanding, the 500 m limit is only applied to the aerosol observations presented in Figure 3 in order to avoid contamination by forest fires. We use only cloud observations in the vicinity of the sites where vertical profiles were obtained. Therefore we can remove forest fires by looking at elevated layers. For the legs between the sites we cannot do that because we do not know whether an observed aerosol layer is elevated or not. Therefore we applied a 500 m threshold instead. For Figs 8-14, the aerosol concentration is obtained directly under the cloud, whether the cloud base is below or above 500 m does not matter. See also Figs. 2 and 6 in Creamean et al 2017. We updated the description of Fig 3 accordingly:

~~The data presented~~ . As discussed above, removal of data potentially

impacted by forest fires is only possible for the spirals. Therefore, the data presented in Fig. 3 are limited to observations obtained below 500 m ~~in order to demonstrate the impact of local emissions and reduce the impact~~ , because transported emissions of forest fires were typically at higher altitudes during ACME-V (for a detailed study of aerosol properties during ACME-V see Creamean et al., 2017) .

Regarding the range of cloud observations, we added:

Cloud base varied between 178 m and 5346 m with a median of 1498 m.

*Minor comments:*

*4) Page 2, lines 4-5 - Aerosol number concentration or mass concentration? If number, what sizes? This statement is very simplistic.*

We clarified "cloud condensation nuclei (CCN) concentration"

*5) Page 3, line 18 - what are 'bulk' probes?*

Bulk probes measure bulk cloud properties such as LWC. We removed that statement because bulk probes are not used in this study.

*6) Pages 3 and 4 - If not described, references are needed for how the CDP, DCDP and OAPs were evaluated and calibrated during the study.*

We added:

For the 2DS probe, the evaluation of particle sizing and sample area determination was done following Korolev et al. (1991). The sample areas of CDP and FCDP were determined by their manufacturer using the technique described by Lance et al. (2010). The droplet size response for CDP and FCDP was calibrated weekly using glass beads in field. In addition, LWC was measured by a multi-element water content system (WCM-200) and used for evaluating the in-flight performance of the 2DS, CDP and FCDP (King et al., 1978, 1981, 1985).

*7) Page 4, line 6 - A droplet threshold of 10/cc may not be appropriate for Arctic summer clouds (e.g. Leaitch et al., ACP, 2016), and it does not allow consideration of situations such as discussed by Mauritsen et al. (ACP, 2011). Please discuss.*

Thank you for this comment. We tested also the use of lower thresholds. For $5 \times 10^6$ $m^{-3}$, the number of clouds increased only by two and they were classified as associated with forest fires. For even lower thresholds, we found artifacts in the cloud classifications. In addition, we see a change in cloud properties mostly for LWC > of 0.1 g/m3, making us confident that clouds with very small LWC are not relevant for our study.

However, we agree, that a discussion about the threshold is necessary, so we added:

> We also evaluated the use of a lower cloud threshold (5 $cm^{-3}$ in accordance with Hobbs and Rangno (1998)), but this only increased the number of observed clouds by two, both of which were classified as impacted by forest fire (see below). In order to avoid sampling errors due to small sample sizes, we use the larger threshold (10 $cm^{-3}$) in the following.

*8) Page 4, line 10 - what are 'tiny' particles?*

Please see the line above in the manuscript:

> The habit classification scheme differentiates between spherical particles, tiny particles which are too small to be classified and various forms of ice crystals.

*9) Page 5 - Describe the inlet for particles measured with the CPCs.*

We added:

> Aerosol particles were sampled through an isokinetic inlet with an upper size cut of 5 $\mu$m (Zaveri et al., 2010; Dolgos and Martins, 2014)

*10) Page 5, line 11 - How was the PCASP calibrated during the study?*

We added:

> The PCASP was calibrated using both size selected ammonium sulphate particles and monodisperse polystyrene latex (PSL) spheres. The sizing accuracy was checked weekly in the field using PSL particles following Cai et al. (2013).

*Is the lower detection limit truly 100 nm (e.g. Liu et al.: Response of Particle Measuring Systems airborne ASASP and PCASP to NaCl and latex particles, Aerosol Sci. Technol., 16, 83-95, 1992)?*

The lower detection limit of the PCASP used is 90 nm, but we omitted the 90-100 nm bin due to the low counting efficiency.

*11) Page 5, line 20 - How as the SP2 calibrated?*

We added:

> The applied SP2 calibration methods using ambient BC and fullerene soot are described in detail by Gysel et al. (2011) and Irwin et al. (2013). The fullerene soot and PSL calibration were performed twice during this field campaign and the sensitivity of the SP2 was found to be stable to around

10% for fullerene soot particles resulting in an estimated SP2 measurement uncertainty of 10%.

*12) Page 5, line 23 - While Arctic Haze is not common during the summer, how can you be certain it was not observed? PCASP number concentrations of 150-200/cc may be representative of Arctic Haze (e.g. Leaitch et al., J. Atmos. Chem, 9, 187-211, 1989).*

The reviewer is right, long range transport was actually found by Creamean et al. (2017) for AMCE-V. We decided not to use the term 'Arctic Haze', because it is usually related to winter and spring time long range transport. However, this does not impact our cloud analysis, we updated:

> For ACME-V, Creamean et al. (2017) classified only four flights as impacted by long range transport from lower latitudes not related to forest fires. During these flights, only a single cloud was sampled in the vicinity of OLI or NSA which had one of the lowest aerosol concentrations measured in the whole data set. Therefore we are confident that our analysis is not strongly impacted by this kind of long range transport events.

*13) Page 5, line 35 - A particle density of 6 g/cm3 is very high. The density of submicron particles is usually less than 2.5 g/cm3. Please explain.*

As stated, this was a HYSPLIT default parameter when deposition is turned on. Additionally, we use the HYSPLIT results only qualitatively, thus, the deposition locations would not change with changing particle density.

*14) Page 6, line 12 - aerosol data*

Added.

*15) Page 6, line 23 - reference for size of freshly emitted soot?*

Thank you for asking for a reference, because the reference actually states 15 nm. We updated:

> Freshly emitted soot has been shown to be larger than ~~this (> 20 nm ), so this range is~~ 15 nm (Zhang et al., 2008), so particles in the 3 to 10 nm size range are likely due to in situ nucleation of aerosol particles from gas phase precursors (i.e., formation of new particles as compared to secondary aerosol formation, where gases condense onto preexisting aerosol, Kulmala et al., 2012)

*16) Page 6, line 26 - qualify 'quickly' by assuming sufficient gaseous precusors.*

We extended:

> Even though the CN dominating the CPC observations are likely too small to act as ~~a~~ CCN, these small particles can grow to accumulation mode quickly given sufficient gaseous precursors , potentially creating a particle population capable of acting as CCN (Jaenicke, 1980).

*17) Page 7, line 22 - smallER*

Thanks, fixed.

*18) Page 7 - In Fig. 4, there are interesting similarities between the small group of yellow points associated with each site. Both groups show increases in PCASP size with increasing CPC. This curious group is related to cloud for the OLI case but not the NSA case. Can you identify a connection?*

Because it did not belong to the main focus of the article, we removed the figure.

*19) Page 9, line 1 - Indicate the reason for the 16 um line in the caption of Fig. 5. Also, in Fig. 5, please add a line showing how LWC vs LER varies assuming the mean droplet number concentration for each location. What are those mean droplet concentrations?*

We added a description of the dashed line to the caption of Figure 5 (now 4). We added a new Figure 5 to show how mean reff and the standard deviation of the drop size distribution changes as a function of LWC. We added:

> Fig. 5.a reveals that this difference is statistically significant for most LWC $>$ 0.1 g $m^{-3}$ according to a Welch's t-test (Welch, 1947) with a 5% confidence interval (Note that a 5% confidence interval is always used in this study unless noted otherwise). [...] For TOI, the mean $r_{eff}$ is significantly different than those at NSA for LWC $>$ 0.02 g $m^{-3}$. Fig. 5.b reveals that not only the mean $r_{eff}$ is reduced at OLI, but also the breadth of the distribution as shown by the standard deviation. This difference is significant for most data points with LWC $>$ 0.1 g $m^{-3}$.

*20) Page 12, lines 1-3 - Is there evidence that OLI emissions impacted any of the NSA observations?*

Indeed, this cannot be excluded because both our Hysplit simulations and Kolesar et al. (2017) found found that emissions can make it to NSA. Therefore we extended:

> The HYSPLIT simulations (Fig. 7) show that the mass concentration originating from local pollution sources ~~is on average more than two order of magnitudes~~ can be a substantially higher at OLI than at NSA. These simulations indicate that relative to NSA, the number of clouds impacted by local emission is ~~increased~~ higher at OLI and these clouds are impacted by a larger amount of aerosol particles by mass. However, an impact of

local emissions on cloud properties is also possible at NSA, although less frequently than at OLI.

We added to the conclusions:

Given the limited data set, we found ten of 24 clouds at OLI, but only one of 16 clouds at NSA which might be impacted by anthropogenic emissions.

*21) Page 12, lines 9-10 - Reduced Reff with increased rBC is not so clear; these plots have a qualitative aspect to them. At the higher LWC (> 0.1 g/m3) that may be true, but below 0.1 it appears that the opposite may be true.*

Because we decreased the pixel size, this effect is now visible for all LWC, albeit not all clouds are impacted

*22) Page 12, lines 15-16 - When averaged over a large number of observations. Also, the "notion" is commonly anticipated for clouds with higher LWC (roughly >0.1 g/m3; e.g. Leaitch et al., JGR, 1992) when effects of evaporation, dissipation and precipitation are reduced factors.*

The updated Figure shows smaller bins, i.e. there is less averaging. We reworded that statement:

However, for OLI there are more enhanced rBC concentrations (> 4 ng $kg^{-1}$) for intermediate $r_{eff}$-values (5-10 $\mu$m) consistent with Fig. 3.a. It is interesting to note that this intermediate region is consistent with enhanced local particle concentrations according to HYSPLIT.

We also added the proposed reference.

*23) Page 12, Lines 18-20 - The use of monotonic is not justified here.*

True, we removed the word monotonic.

*24) Page 13, lines 8-9 - How many CCN are needed for cloud formation?*

We removed Fig. 4 and the corresponding discussion in order to shorten the manuscript and because it did not contribute to the main focus of the article.

*25) Page 15, header for section 6 - You are not discussing an 'interaction' here, only a potential impact of the aerosol on the cloud.*

We decided to stick to the term "aerosol cloud interaction" to be consistent with existing literature (e.g. Coopman et al., 2016; Zamora et al., 2016). In addition, clouds do have an impact on aerosols, e.g. by processing and through dynamical feed-backs. Therefore the term interaction is appropriate.

# The observed influence of local anthropogenic pollution on northern Alaskan cloud properties

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**Abstract.**

Due to their importance for the radiation budget, liquid-containing clouds are a key component of the Arctic climate system. Depending on season, they can cool or warm the near-surface air. The radiative properties of these clouds depend strongly on cloud drop sizes, which are governed by the availability of cloud condensation nuclei. Here, we investigate how cloud drop sizes are modified in the presence of local emissions from industrial facilities at the North Slope of Alaska. For this, we use aircraft in-situ observations of clouds and aerosols from the 5th Department of Energy Atmospheric Radiation Measurement (DOE ARM) Program's Airborne Carbon Measurements (ACME-V) campaign obtained in Summer 2015. Comparison of observations from an area with petroleum extraction facilities (Oliktok Point) with data from a reference area relatively free of anthropogenic sources (Utqiaġvik/Barrow) represents an opportunity to quantify the impact of local industrial emissions on cloud properties. In the presence of local industrial emissions, the mean effective radii of cloud droplets are reduced from 12.2 to ~~9.8~~ 9.4 $\mu$m, which leads to ~~a suppression of~~ suppressed drizzle production and precipitation. At the same time, concentrations of refractory black carbon and condensation nuclei are enhanced below the clouds. These results demonstrate that the effects of anthropogenic pollution on local climate need to be considered when planning Arctic industrial infrastructure in a warming environment.

## 1 Introduction

Liquid-containing clouds are a significant modulator of the Arctic climate system's radiation budget. Their properties impact both shortwave and longwave radiative transfer, resulting in seasonally-dependent influences that include both net cooling and warming of the Arctic surface (Intrieri et al., 2002; Shupe and Intrieri, 2004), and various forms of cloud feedbacks (Colman, 2003). At the same time, liquid cloud droplet number concentration and size are influenced by the number of available cloud condensation nuclei. It has been proposed that this has an effect on cloud albedo, life cycle and longwave emissivity (Twomey, 1976; Albrecht, 1989; Garrett and Zhao, 2006). Long range transport of aerosol particles from lower latitudes in winter and

early spring (Arctic haze) and episodic forest fires in summer can lead to higher aerosol concentrations (Shaw, 1995; Law and Stohl, 2007), which have been found to modify liquid and mixed phase cloud properties (Garrett et al., 2004; McFarquhar et al., 2011; Jackson et al., 2012; Zamora et al., 2016). Besides these transported emissions, the Arctic is an environment that is generally relatively clean (Quinn et al., 2002, 2009) which makes clouds particularly susceptible to an increase in aerosol cloud condensation nuclei (CCN) concentration (Platnick and Twomey, 1994). In comparison to other regions, there are only few sources of local anthropogenic emissions north of the Arctic Circle, which are mainly related to ship traffic and petroleum as well as natural gas extraction facilities (Law and Stohl, 2007). While emissions from ships are expected to rise due to the retreating sea ice, emissions from resource extraction are expected to remain at present day levels (Peters et al., 2011) with an estimated 13% of the world's untapped oil resources located in the Arctic (Gautier et al., 2009). Local emissions by Arctic petroleum and natural gas extraction facilities have been observed and quantified by aircraft campaigns (Brock et al., 2011; Roiger et al., 2015). These emissions are mostly associated with flaring, but also by regular internal combustion engines. Ødemark et al. (2012) found that black carbon (BC), which is particularly created by flaring (Stohl et al., 2013), results in a modeled positive net radiative forcing of petroleum and natural gas extraction, mainly due to deposition of BC on the snow. Kolesar et al. (2017) showed that emissions from the Prudhoe Bay area result in in-situ particle growth events in BarrowUtqiaġvik (formerly known as Barrow), located around 300 km west of the Prudhoe Bay region. Although these previous studies have demonstrated the potential impact from industrial activities in the Arctic, in-situ aerosol and cloud observations have not been combined in order to study local sources of emissions.

In this work, we show how cloud properties are altered by aerosol particles originating from local anthropogenic pollution from industrial activities in the Prudhoe Bay area in northern Alaska (Fig. 1), and investigate the influence on processes impacting the cloud life cycle. Even though the work is limited to observations from the North Slope of Alaska, the results are broadly applicable to other Arctic regions with significant industrial activities (e.g. Siberia), although exact details of the types of aerosol effects will be influenced by aerosol concentration, size, and composition. Because of their importance in regulating the surface and top-of-atmosphere energy budgets, we focus here on liquid clouds. Increased cloud droplet concentrations in the Prudhoe Bay, Alaska area were previously reported by Hobbs and Rangno (1998) although that study could not directly connect these increased concentrations to locally produced aerosol particles due to a lack of aerosol measurements. In this study, we fill this gap by using airborne cloud property and aerosol observations obtained during the US Department of Energy Atmospheric Radiation Measurement (DOE ARM) program's 5th ARM Airborne Carbon Measurements (ACME-V) campaign to study the influence of local pollution on Arctic liquid clouds. An enhanced understanding of the influence is crucial to evaluate the role of clouds and aerosols in changing Arctic which is warming faster than other regions (Jeffries et al., 2013).

In Section 2 we provide background information on the ACME-V campaigns along with details on the various data sets used to conduct our analysis. Following this, we analyze analyse observed aerosol particle (Section 3) and cloud (Section 4) properties, before combining these to evaluate the interactions between locally-produced aerosols and clouds in Section 5. This evaluation is carried further in Section 6 where we attempt to quantify observed aerosol-cloud interactions. Finally, we provide a summary and concluding remarks in Section 7.

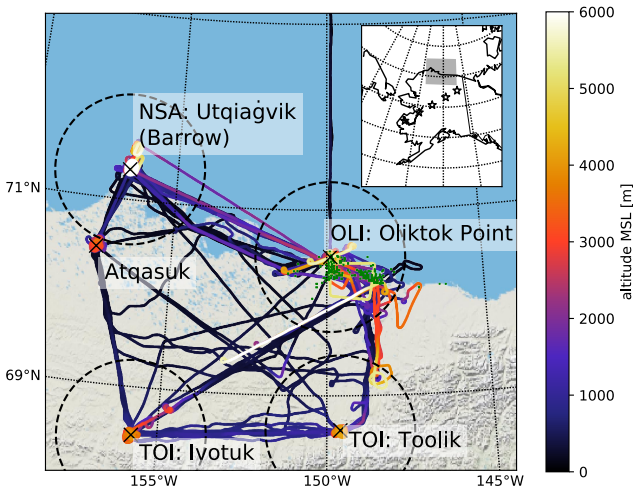


**Figure 1.** Overview of all flights of the ACME-V campaign. Color shows altitude m MSL. The dashed circles indicate 90 km radii around the sites (black crosses), the ~~red~~ green dots indicate active oil wells (Data obtained from http://doa.alaska.gov/ogc/publicdb.html in March 2017). The grey inlet shows the location of the region in Alaska and the five assumed sources for forest fire emissions (stars).

## 2 Data set

The ACME-V aircraft campaign took place from June ~~to September~~ 1st to September 15th 2015 ~~(ARM, 2016)~~ (Biraud, 2016; ARM, 2016) and consisted of 38 research flights of the ARM Gulfstream G-159 (G-1) aircraft of the ARM aerial facility (Schmid et al., 2014, 2016). Since the campaign targeted trace gas measurements from local and regional sources, a majority of the flight time was spent below 200 m above mean sea level (MSL). However, spirals up to an altitude of 6,000 m were flown in the vicinity of two ARM surface observatories in northern Alaska, Utqiaġvik (formerly known as Barrow or ARM's North Slope of Alaska site, NSA, 71.323°N, 156.616°W) and Oliktok Point (OLI, 70.495°N, 149.886°W). Additional spirals were flown at Toolik (68.628°N, 149.598°W), Ivotuk (68.483°N, 155.754°W), and Atqasuk (70.467°N, 157.436°W) in order to ~~characterize~~ characterise cloud and aerosol properties (Fig. 1). In this work, we compare data within 90 km of OLI and NSA. These two sites form an ideal opportunity to study the effects of local emissions on cloud properties: While OLI is surrounded by industrial activities related to oil and natural gas extraction (with the majority closer than 90 km), no substantial local sources exist in the vicinity of NSA and previous studies have shown only limited advection ($8 \pm 2\%$) of air masses passing through the Prudhoe Bay area to NSA (Kolesar et al., 2017). Despite substantial differences in aerosol properties, the two coastal sites lie only 250 km apart, resulting in very similar synoptic scale forcing, as can be seen from the high correlation between both sites for pressure, temperature, humidity, and wind (Fig. 2). For both sites, north-easterly to easterly winds prevailed during ACME-V ~~(ARM, 1993, updated daily)~~(see data set ARM, 1993, updated daily). Additionally, we grouped observations closer than 90 km to the two more continental sites Toolik and ~~Atqasuk~~ Ivotuk into a third data set (~~labeled~~ labelled TOI). During ACME-V, 156 (60%) of the 258 vertically sampled clouds were classified as liquid (see below for thresholds), showing that

liquid clouds are frequent during the summer time in Northern Alaska. Data obtained during take off and landing have been removed to avoid skewing the comparison by sampling aerosols and clouds at much lower altitudes than elsewhere.

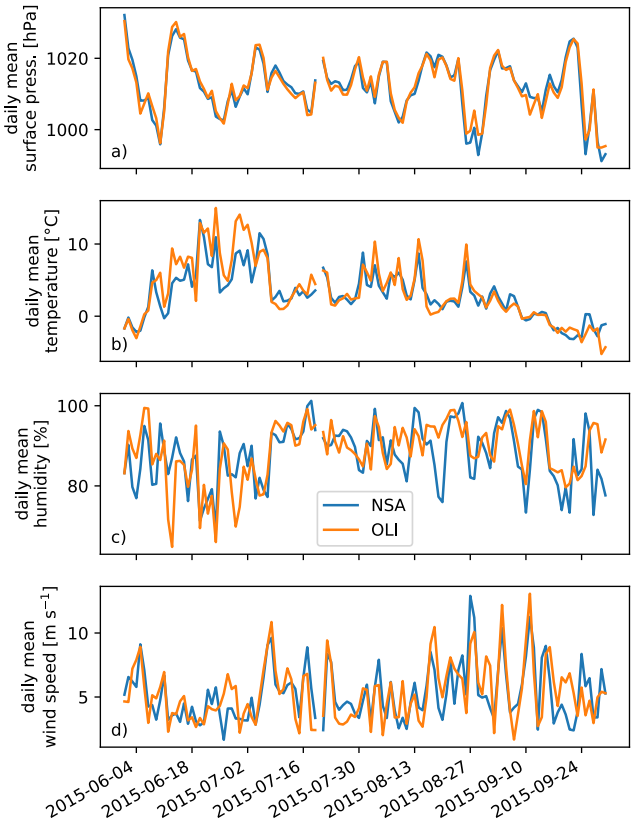


**Figure 2.** Comparison of daily mean values for a) surface pressure, b) 2 m temperature, c) 2 m humidity, and d) 10 m wind speed.

Cloud properties were observed using a combination of forward scattering ~~, optical array , and bulk~~ and optical array probes. The particle size distributions were measured using the forward scattering Cloud Droplet Probe (CDP) manufactured by Droplet Measurement Technologies (DMT), Inc.~~;~~, the Fast Cloud Droplet Probe (FCDP) from Stratton Park Engineering Company (SPEC), Inc.~~;~~, the Two Dimensional Stereo optical array ~~probes (OAPs) (2DS, Lawson et al., 2006);~~ probe (OAP) (2DS, Lawson et al., 2006), and the High Volume Precipitation Spectrometer (HVPS, Lawson et al., 1993) from SPEC, Inc. ~~The~~ For the 2DS probe, the evaluation of particle sizing and sample area determination was done following Korolev et al. (1991). The sample areas of CDP and FCDP were determined by their manufacturer using the technique described by Lance et al. (2010). The droplet size response for CDP and FCDP was calibrated weekly using glass beads in field. In addition, LWC was measured by a multi-element water content system (WCM-200) and used for evaluating the in-flight performance of the 2DS, CDP and FCDP (King et al., 1978, 1981, 1985). The raw OAP datasets were processed by the University of Illinois Optical Array Probe Processing Software (Wu and McFarquhar, 2016). In order to merge the cloud droplet size distributions, FCDP data were used for particles less than 50 $\mu$m size, the 2DS was used for particles between 50 and 605 $\mu$m, and the HVPS was used for all

particles exceeding 605 $\mu$m. In this study, particle ~~diameter~~ maximum dimension is used in general to describe the size of cloud and aerosol particles. Liquid clouds were required to have at least 10 ~~7 m~~cm$^{-3}$ droplets (Lance et al., 2011). We also evaluated the use of a lower cloud threshold (5 cm$^{-3}$ in accordance with Hobbs and Rangno (1998)), but this only increased the number of observed clouds by two, both of which were classified as impacted by forest fire (see below). In order to avoid sampling errors due to small sample sizes, we use the larger threshold (10 cm$^{-3}$) in the following. In order to remove ice clouds from the data set, the Holroyd habit classification was applied to 2DS and HVPS observations with 1 s temporal resolution, which classifies particles mainly based on a fine detail ratio $F = pd/a$, where $p$ is perimeter, $d$ is diameter and $a$ is area ~~(Holroyd, 1987)~~(Holroyd, 1987; Wu and McFarquhar, 2016). The habit classification scheme differentiates between spherical particles, tiny particles which are too small to be classified and various forms of ice crystals. Spherical particles were assumed to be liquid. Tiny particles appear only at the lower end of the 2DS ($< 105$ $\mu$m) and HVPS ($< 1125$ $\mu$m) size range. They were classified as ice only if other size ranges were not dominated by spherical particles. Otherwise, tiny particles were assumed to be liquid. Data points with more than 100 m$^{-3}$ particles larger than 400 $\mu$m (Lance et al., 2011) classified as ice were removed from the data set. This ensures that observations of spherical ice particles falsely classified as liquid, which likely occur together with larger, more complex shaped ice particles, were removed from the data set as well. Liquid water content (LWC) was obtained by integrating the merged droplet size distribution (DSD), because direct observations of LWC from the King probe (King et al., 1978) are affected by a decreasing sampling efficiency for (drizzle) drops greater than 30 $\mu$m diameter. Clouds that were observed for less than ten continuous seconds were discarded, while gaps of up to 5 s were permitted once in cloud. Considering the typical true airspeed of the G-1 of 95 m/s, ~~this corresponds~~ 10 s and 5 s correspond to 950 m and 475 m, respectively, when flying in a straight line~~, respectively~~. Additionally, only vertically sampled clouds (i.e. the aircraft was constantly ascending or descending) with a sampled vertical extent of at least 20 m were included in this evaluation to allow for comparison of in-cloud microphysical observations with below-cloud aerosol properties in sections 5 and 6. Therefore, very thin and/or small clouds might be discarded inadvertently. To make the detection of cloud boundaries more robust, the cloud probe data were smoothed using a ~~10s~~ 10 s running average. Except for the detection of the cloud boundaries, effects of the smoothing are negligible for the presented analysis. ~~For liquid clouds, aerosols of~~

Aerosol particles were sampled through an isokinetic inlet with an upper size cut of 5 $\mu$m (Zaveri et al., 2010; Dolgos and Martins, 2014). Aerosols in the size range 100 nm to 3 $\mu$m were observed with the Passive Cavity Aerosol Spectrometer (PCASP model 100X, DMT Inc.) covering most accumulation mode aerosols (Colbeck and Lazaridis, 2014). We expect particles measured by the PCASP to be mostly dry, because it was operated with an anti-ice heater. Kassianov et al. (2015) showed for the very same aircraft that this assumption leads to good agreement between calculated (using, among others, PCASP observations) and measured scattering properties. The PCASP was calibrated using both size selected ammonium sulphate particles and monodisperse polystyrene latex (PSL) spheres. The sizing accuracy was checked weekly in the field using PSL particles following Cai et al. (2013). Unfortunately, another aerosol sampler (Ultra-High Sensitivity Aerosol Sizer, UHSAS), which is able to detect aerosols below the PCASP detection threshold of 100 nm, did not operate during the majority of the ACME-V flights. Two Condensation Particle Counters (CPC, TSI, Inc. models 3025 and 3010~~, respectively~~) were used to observe total number concentrations of condensation nuclei (CN) for the size ranges 3 nm - 3 $\mu$m and 10 nm - 3 $\mu$m, respectively.

CPC calibration activities included verifying inlet flow rate with a low pressure-drop bubble flow meter, and determining the size-dependent particle counting efficiency, according to methods defined in Hermann et al. (2007) and Mordas et al. (2008). Unless otherwise stated, only the CPC 3025 featuring a size range of 3 nm - 3 $\mu$m ~~was~~ is used in this evaluation. ~~Black~~ The mass and core size of black carbon (BC), which results from incomplete combustion of biomass and fossil fuels (Schwarz et al., 2008; Bond et al., 2013; Lack et al., 2014), was measured with the Single Particle Soot Photometer (SP2, from DMT Inc.), via incandescence. Thus, only refractory black carbon (rBC) is observed by the instrument. The applied SP2 calibration methods using ambient BC and fullerene soot are described in detail by Gysel et al. (2011) and Irwin et al. (2013). The fullerene soot and PSL calibration were performed twice during this field campaign and the sensitivity of the SP2 was found to be stable to around 10% for fullerene soot particles resulting in an estimated SP2 measurement uncertainty of 10%. Concentrations of carbon monoxide (CO) were detected with a Los Gatos Research $CO/N_2O/H_2O$ Analyzer. A counter for cloud condensation nuclei (CCN) was not deployed during ACME-V. The temporal resolution of the aerosol probes is 1 s with the exception of the SP2 (10 s).

~~While Arctic Haze was not observed during ACME-V, transported~~ Transported emissions from forest fires can contribute significantly to summertime aerosol loading in the Arctic ~~(Law and Stohl, 2007)~~(Law and Stohl, 2007; Creamean et al., 2017). Therefore, we manually inspected the vertical profiles of rBC and ~~carbon monoxide (CO)~~CO, which together constitute a good tracer for biomass burning (Warneke et al., 2009, 2010). Typically, these layers are found aloft (Roiger et al., 2015), allowing us to use vertical profiles obtained by the aircraft to aid in their identification. For each spiral obtained at the two sites, elevated layers with CO $\geq$ 0.1 ppmv or rBC $\geq$ 20 ng kg$^{-1}$ were flagged as corresponding to forest fires. Local emissions, on the other hand, are expected to be found in a layer connected to the surface. Note that the data impacted by forest fires were only removed for spirals above OLI, NSA, and TOI. For clear-air observations during level flight legs between sites, it is generally impossible to determine whether a layer is connected to the surface or elevated. For ACME-V, Creamean et al. (2017) classified only four flights as impacted by long range transport from lower latitudes not related to forest fires. During these flights, only a single cloud was sampled in the vicinity of OLI or NSA which had one of the lowest aerosol concentrations measured in the whole data set. Therefore we are confident that our analysis is not strongly impacted by this kind of long range transport events.

The manual inspection was supported by aerosol dispersion simulations executed using version 4 of the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015). These simulations were forced using 1° data from the NOAA/NCEP Global Data Assimilation System (GDAS) (Kalnay et al., 1996). Five locations were included as sources (see Fig 1: (1) 62.096°N, 163.632°W, (2) 63.843°N, 159.046°W, (3) 65.294°N, 154.386°W, (4) 66.631°N, 149.023°W, and (5) 67.631°N, 144.087°W)~~and~~ . These sources were toggled on or off on a daily basis in correspondence to thermal anomaly observations in the corresponding region (see Fig. 5 of Creamean et al., 2017) from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Aqua and Terra satellites obtained using brightness temperature measurements in the 4 and 11 $\mu$m channels ~~.~~(Giglio et al., 2003; Giglio, 2013). From each fire location, particle mass concentrations were simulated for 72 h at 100-m intervals from 0 to 5,000 m above ground level (m AGL). Both dry and wet deposition were considered for particles using the default HYSPLIT ~~parametrizations~~ parametrisations (particle density 6 g cm$^{-3}$, shape factor 1.0). The particle diameter of 0.2 $\mu$m used for the simulations is based on previous observations from fossil fuel and biomass burning

sources (Brock et al., 2011; Eck et al., 1999; Rissler et al., 2006; Sakamoto et al., 2015). A dry deposition velocity of 1 x $10^{-4}$ m $s^{-1}$ was assumed according to Warneck (1999) while 4 x $10^{4}$ L $L^{-1}$ and 5 x $10^{-6}$ $s^{-1}$ were used to account for in-cloud and below-cloud wet deposition scavenging, respectively. Radioactive decay and pollutant resuspension were not considered. ~~Note that the data impacted by forest fires were only removed for spiralsabove OLI, NSA, and TOI. For clear-air observations during level flight legs between sites, it is generally impossible to distinguish aerosols~~ For the spirals, data identified as originating from forest ~~fires as compared to other sources. Therefore, data potentially impacted by forest fires have only been removed from the cloud observations which have been associated with vertical profiles (Sec. 4f), but not from the aerosol observations presented in Sec. 3.~~ fire either from manual inspection or according to HYSPLIT, were removed from subsequent analysis. With this approach, we likely removed more clouds from the analysis than required. This, however, ensures that the analysis of the remaining clouds is not biased by influences from forest fires.

## 3 Aerosol properties

The spatial ~~distributions~~ distribution of aerosol observations below 500 m MSL are presented for the CPC, the SP2, and the PCASP in Fig.3~~. The data presented~~ . As discussed above, removal of data potentially impacted by forest fires is only possible for the spirals. Therefore, the data presented in Fig. 3 are limited to observations obtained below 500 m~~in order to demonstrate the impact of local emissions and reduce the impact~~ , because transported emissions of forest fires were typically at higher altitudes during ACME-V (for a detailed study of aerosol properties during ACME-V see Creamean et al., 2017). Furthermore, aerosol data flagged as sampled in cloud using the thresholds described in the previous section ~~have been~~ were discarded in the analysis of aerosol properties due to concerns of contamination of the statistics by shattering of cloud droplets.

~~For the SP2, a~~ A clear local maximum of rBC mass concentration is visible east of OLI in the SP2 data within the 90 km radius (Fig. 3.a) where most petroleum and gas extraction facilities are located (Fig. 1). A comparison of the distributions measured within a 90 km radius around the facilities at NSA and OLI reveals that the median ~~of~~ rBC concentration is the same for both regions (4 ng $kg^{-1}$). The tail ~~of~~ and the number of outliers of the distributions towards larger concentrations, however, ~~is much~~ are greater at OLI (90th and 99.9th percentile ~~15~~ 17 ng $kg^{-1}$ and ~~42~~ 198 ng $kg^{-1}$, respectively) than at NSA (~~17~~ 15 ng $kg^{-1}$ and ~~198~~ 42 ng $kg^{-1}$, respectively). ~~This~~ rBC is a tracer for combustion emissions (Bond et al., 2004). Because the height threshold of 500 m reduces the impact of forest fires, this enhancement is most likely connected to local emissions. CN measurements from the CPC show a ~~similar pattern~~ spatial pattern similar to the SP2 even though the increased values are distributed over a larger area ~~.~~ (Fig. 3.b). For both instruments, the distributions within the 90 km circle belonging to each site are skewed towards higher concentrations and the distributions of both sites are significantly different (1% confidence interval) according to the two sample Kolmogorov–Smirnov (KS) test (Massey, 1951). Further, the difference between ~~both~~ the two CPC instruments, which ~~depends on~~ equates to the concentration of CN between 3 and 10 nm diameter, is enhanced ~~east of OLI (not shown) .~~ in the OLI region and the distribution is significantly (KS-test) different to the one at NSA (Fig. 3.c). Because this quantity is stemming from the difference in two instruments at the limit of their measurement range, the data is used here only in a qualitative way. Freshly emitted soot has been shown to be larger than ~~this (> 20 nm ), so this range is~~

15 nm (Zhang et al., 2008), so particles in the 3 to 10 nm size range are likely due to in situ nucleation of aerosol particles from gas phase precursors (i.e., formation of new particles as compared to secondary aerosol formation, where gases condense onto preexisting aerosol, Kulmala et al., 2012). Nucleated aerosols typically have sizes below 3 nm, but quickly grow via condensation and coagulation to sizes ~~>~~ > 3 nm (Colbeck and Lazaridis, 2014). This source of nucleated aerosol particles from petroleum and gas extraction activities has been reported by Kolesar et al. (2017) for emissions transported from OLI to NSA.

Unfortunately, we cannot ~~analyze~~ analyse this aerosol nucleation process in more depth given limitations with the instrumentation operated during ACME-V. rBC background concentrations appear to be similar to ~~values found by Zamora et al. (2016) (1-16 ng/m3) and Roiger et al. (2015)(median 20-30 ng /kg) for summertime transported forest fire plumes in the Arctic. Other studies (Warneke et al., 2009; Schwarz et al., 2010) found up to one order of magnitude higher rBC concentrations in the Arctic which is more similar to the maximum values we observed around OLI~~ background observations made by Zamora et al. (2016) and Roiger et al. (2015). It should be noted that emissions related to forest fires led to concentrations as high as 600 to 1000 ng $kg^{-1}$ during ACME-V (mostly at altitudes above 500 m, Creamean et al., 2017) which were also observed in other data sets (Warneke et al., 2009; Schwarz et al., 2010; Zamora et al., 2016). Consequently, the emissions from anthropogenic sources in the OLI region are about a magnitude lower. In contrast to ~~traces~~ CO concentrations sampled in air masses originating from forest fires, ~~carbon monoxide (CO ) concentrations were not found to be significantly enhanced~~ low altitude CO concentrations in the OLI region were not enhanced relative to background values (Creamean et al., 2017). The differences between CO and rBC concentrations attributed to forest fires and the concentrations measured in the OLI region ~~(not shown)~~show that our approach to use CO and rBC to separate observations impacted by forest fires is feasible.

The PCASP, which detects only particles larger than 100 nm, shows no spatial trends in the vicinity of the two sites ~~.~~ (Fig. 3.d). The comparison of the distributions around the facilities shows that the number of aerosols observed by the PCASP is on average actually slightly larger for NSA than for OLI. This is related to the fact that the median of the distribution is ~~at~~ 97 $cm^{-3}$ at NSA and ~~at~~ 76 $cm^{-3}$ at OLI. Similar to rBC, the tail of the distributions towards larger concentrations is greater at OLI (90th percentile 200 $cm^{-3}$) than at NSA (184 $cm^{-3}$) and the difference in the distributions is significant according to the KS-test with 1% confidence interval. While it is challenging to clarify the precise cause of the increased mean concentration in detail, we speculate it might be related to transported emissions, including those from forest fires, that have not been properly removed from the data set ~~.~~ because they are highly diluted. Transported forest fire aerosols are often larger than the PCASP detection threshold of 100 nm as shown by Kondo et al. (2011) and Sakamoto et al. (2015). An alternative explanation could be the fact that collision-coalescence and precipitation rates are larger at NSA than at OLI (see next Sec. 4.) resulting in more aerosol processing by precipitation ~~(e.g. Hoppel et al., 1990)~~(e.g. **?**). Cloud-based processing leads to a reduction in aerosol concentration and an increase in aerosol size through conglomoration of cloud droplets (and corresponding aerosol particles) in the drizzle formation stage and subsequent evaporation. Such effects could increase the number of aerosols within the PCASP measurement size range.

~~To investigate why there is a clear enhancement in the CPC concentrations but not in the PCASP concentrations, both instruments sets are compared for all non-cloud observations below 500 m MSL during ACME-V. Fig. ?? shows CN concentration versus PCASP mean particle size and PCASP number concentrations for the two 90 km radii around OLI and NSA. It clearly~~

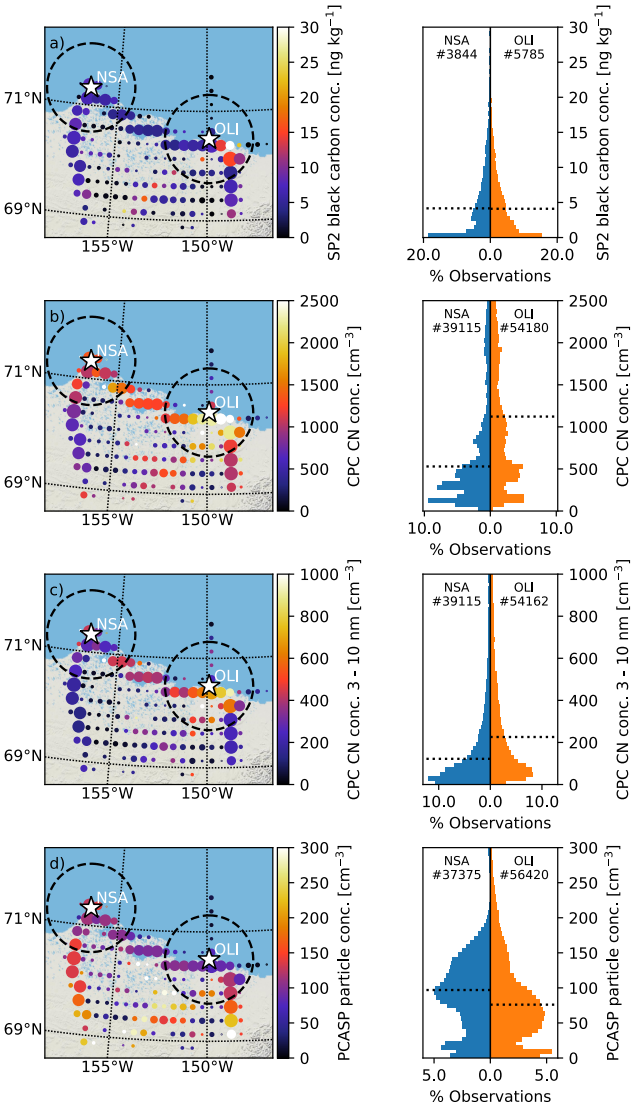


**Figure 3.** Left column: Spatial distribution of mean (a) SP2 refractory black carbon concentration, (~~a~~b) ~~,~~CPC3025 CN concentrations, (~~b~~c) difference between CPC3025 and CPC3010 CN concentration corresponding to a size range of 3 to 10 nm, and (d) PCASP aerosol concentration~~(c)~~. Only non-cloudy observations below ~~400~~ 500 m MSL have been considered. The size of the dots is proportional to the number of observations. The dashed circles correspond to a distance of 90 km. Right column: Here, the distribution of measurements within the 90 km circles are shown, the number above the distribution shows the number of observations. The horizontal dotted bar denotes the median value.

~~illustrates that elevated CPC concentrations (> 600 cm$^{-3}$) found at NSA are typically connected with increased PCASP concentrations (> 100 cm$^{-3}$). While mean particle size generally increases with decreasing CN concentration, the variability of PCASP mean size is rather low for CN concentrations > 600 cm$^{-3}$, which is consistent with the idea that particles have~~

already experienced growth. For OLI, on the other hand, variability in PCASP mean size is much larger. In addition, increased CPC CN concentrations do not necessarily correlate to increased PCASP particle concentrations at OLI. This means that for enhanced CN concentrations, which likely indicate a local source, PCASP particle concentrations can be small, but mean particle size is still significantly larger than 100 nm. The lower concentration and the higher size variability observed at OLI is consistent with the idea that particles in this region are younger. The value of 100 nm is important because it is assumed to be the size threshold between particles in Aitken mode and accumulation mode and typically only the latter can act as a CCN. (We do, however, note that smaller particles can act as CCN in very clean conditions (Leaitch et al., 2016).) In summary, aerosol particle sizes are more diverse at OLI and elevated CN concentrations are accompanied by enhanced PCASP mean particle sizes even though total PCASP particle concentrations are lower at OLI. Additional evidence for significant differences in aerosol concentrations between the sites is found in the fact that particle concentration of 600 $cm^{-3}$ is exceeded in 62% of all cases at OLI, but only 35% at NSA.

CPC3025 CN concentration versus PCASP mean particle size with PCASP particle concentration as color for (a) Utqiaġvik and (b) Oliktok Point. The black line corresponds to a CN concentration of 600 $cm^{-3}$ The dots with red edges correspond to mean sub-cloud values corresponding to the clouds discussed in this study.

## 4 Cloud properties

Here, cloud properties are compared for flights occuring occurring near NSA and near OLI. In order to evaluate a sufficiently large sample, all observations obtained closer than 90 km to NSA, OLI and the two sites comprising TOI are assigned to the corresponding site (Fig, 1). As mentioned above , Also data obtained at altitudes above 500 m is considered, but clouds impacted by forest fires have been removed based on the vertical profile as mentioned above. This limits the number of observations to 1608 996 1 s data points for OLI, 942 for NSA, and 579 514 for TOI. Cloud base varied between 178 m and 5346 m with a median of 1498 m.

When comparing 2D histograms of liquid effective radius and liquid water content for OLI and NSA (Fig. 4, a, b), a shift towards OLI values are shown to feature smaller $r_{eff}$ can be clearly seen in the measurements obtained in close proximity to OLI. This for the same LWC. The effect is most pronounced for LWC $>$ 0.1 g $m^{-3}$ (?) while distributions for LWC $<$ 0.01 g $m^{-3}$ are more similar. Note that LWC values below 0.01 g $m^{-3}$ are defined as not in-cloud by some studies (e.g., Matsui et al., 2011; Leaitch et al., 2016), but we decided to show the full data set because the in-cloud definition used here ($>$ 10 droplets $cm^{-3}$) can result in LWC as low as 0.001 g $m^{-3}$ and we wanted to make sure that all cloud data points are included in the analysis. The decrease in $r_{eff}$ supports our hypothesis that CCN concentrations are elevated in the OLI region, since the first aerosol-cloud indirect effect proposes that droplet size is reduced when more CCN are available (all else equal). While droplet $r_{eff}$ observed at NSA cover the full range from droplet nucleation to drizzle (3 to 25 $\mu$m, mean 12.2$\pm$6.9 $\mu$m), $r_{eff}$ values are typically smaller than 16 $\mu$m at OLI (mean 9.89.4$\pm$4.0 4.1 $\mu$m) and observations of drizzle-sized droplets are rare. The value of 16 $\mu$m is of special interest because it was proposed by Gerber (1996) as a minimal effective radius required to initiate collision-coalescence. Fig. 5.a reveals that this difference is statistically significant for most LWC $>$ 0.1 g

$m^{-3}$ according to a Welch's t-test (Welch, 1947) with a 5% confidence interval (Note that a 5% confidence interval is always used in this study unless noted otherwise). For comparison, data obtained in a 90 km radius around Toolik and Ivotuk (TOI) (Fig. 4, c) ~~.~~reveal that the distribution of observed $r_{eff}$ at the coastal site in OLI is ~~more similar~~ still larger to the inland sites comprising TOI (mean 7.2±3.1 $\mu$m) than to the second coastal site NSA. For TOI, the mean $r_{eff}$ is significantly different than those at NSA for LWC $> 0.02$ g $m^{-3}$. Fig. 5.b reveals that not only the mean $r_{eff}$ is reduced at OLI, but also the breadth of the distribution as shown by the standard deviation. This difference is significant for most data points with LWC $> 0.1$ g $m^{-3}$.

The Albrecht effect proposes that more polluted clouds have longer cloud lifetime due to less efficient collision-coalescence (Albrecht, 1989). It is not possible to study the cloud life cycle using aircraft in-situ observations, but the potential for impact on cloud life cycle can be estimated by calculating the collection growth rate $C$ (Long, 1974) ~~.~~and precipitation rate $R$. Even though the rate of mass removal from a cloud is an important process impacting cloud life cycle, it is important to note that modifications to $C$ and $R$ cannot be directly translated into modifications in cloud lifetime. This is because a reduction in $R$ could result in a number of feedbacks such as cloud deepening (Stevens and Feingold, 2009) or reduced evaporation just below cloud base (Feingold and Siebert, 2009) that would act to counter the extending effect of reduced precipitation rate on cloud lifetime.

$C$ describes the mass of drops collected by a unit mass in a unit volume $M$ per time interval $t$. It is the key process for converting cloud drops into precipitation and is estimated by integrating the mass collected by particles with diameter $D_1$ and mass $m_1$ over all size bins:

$$C = \frac{\mathrm{d}M}{\mathrm{d}t} = \int_{D_{\min}}^{D_{\max}} \frac{\mathrm{d}m_1}{\mathrm{d}t} N(D_1)\,\mathrm{d}D_1 \tag{1}$$

where $N(D_1)$ is the particle ~~size distribution~~ number distribution function and $D_{\min}$ and $D_{\max}$ are the bounding drop diameters as determined by the cloud probes (0.75 $\mu$m and 8.7 mm). $\frac{\mathrm{d}m_1}{\mathrm{d}t}$ is obtained by integrating the collection kernel $K$ for all smaller size bins (i.e. $D_1 > D_2$) described by the diameter of the collected drops $D_2$

$$\frac{\mathrm{d}m_1}{\mathrm{d}t} = \frac{\pi \rho_w}{6} \lim_{D_1 \to D'} \int_{D_{\min}}^{D'} K(D_1, D_2) N(D_2) D_2^3\,\mathrm{d}D_2 \tag{2}$$

where $\rho_w$ is the density of liquid water. For simplicity, here we use a simple polynomial approximation of $K$

$$K(D_1, D_2) \approx \begin{cases} 5.78 \times 10^3 (v_1 + v_2) & 20 \leq D_1 \leq 100 \mu m \\ 9.44 \times 10^9 (v_1^2 + v_2^2) & D_1 > 100 \mu m \end{cases} \tag{3}$$

where $v_i$ is the drop volume corresponding to $D_i$ (Long, 1974; Pruppacher and Klett, 2010). Typical values for $C$ range from $1 \times 10^{-16}$ kg $m^{-3}$s for LWC = 0.001 g $m^{-3}$ to $1 \times 10^{-5}$ kg $m^{-3}$s for LWC = 1 g $m^{-3}$. Note that our approximation does not consider the impact of turbulence and droplet charge on $C$. This might lead to considerable uncertainties, which have—to the authors' best knowledge—not been fully quantified. Because we are interested how $C$ is modified in the OLI region, we ~~show the difference~~ focus on the ratio of $C$ ~~between both sites in~~ determined at NSA and OLI which should

reduce the uncertainty of $C$. Fig. 6 shows the ratio between NSA and OLI of $C$ as a function of $r_{eff}$ and LWC. It can be seen that $C$ is decreased at OLI in comparison to NSA by up to one order of magnitude for constant LWC and $r_{eff}$. This is caused by reduced broadening of the drop size distribution towards large drops at OLI (~~not shown~~Fig. 5.b), consistent with ~~cloud chamber experiments (Gunn and Phillips, 1957). Interestingly, differences in $C$ are largest for $r_{eff}$~~the experiments of Gunn and Phillips (1957), who produced similar results when ingesting polluted background air into their cloud chamber. The difference between both sites is significant for most values with sufficient number of observations for both sites (see Fig. ~~smaller than 16 $\mu$m where absolute values of $C$ are small according to Gerber (1996).~~4). However, small absolute increases in $C$ for small $r_{eff}$ are also crucial for triggering the positive feedback of drop growth due to collision-coalescence. When evaluating the potential impact of reduced $C$ on cloud life cycle, one also has to consider that typical $r_{eff}$ values are reduced at OLI in comparison to NSA for the same LWC (Fig. 4, a, b). Therefore, we estimate the mean growth rate $\bar{C}$ averaged over $r_{eff}$ as a function of LWC (Fig. 6, a) red lines). Doing so reveals that, for constant LWC, $\bar{C}$ is reduced by 1 to 1.5 orders of magnitude at OLI. The offset is significant and surprisingly constant for LWC larger than 0.01 g $m^{-3}$. Differences in $C$ also translate to different rain rates $R$, which can be estimated by integrating the measured DSD and applying the fall velocity ~~parametrization of Khvorostyanov and Curry (2002)~~ parametrisation of Khvorostyanov and Curry (2002) which provides a continuous solution over the entire drop size range in dependence of the Best and Reynolds number. Like $C$, $R$ is reduced by up to one order of magnitude for constant LWC and $r_{eff}$ (Fig. 6, b). Averaging over all $r_{eff}$ enhances the effect and leads to differences of up to two orders of magnitude for $R$ as a function of LWC. This effect is statistically significant for LWC $> 0.02$ g $m^{-3}$.

Parameterizations of $C$ and $R$ are crucial in numerical models to transform cloud liquid water into rain droplets and to remove condensate from the atmosphere. Typically, numerical weather and climate models include either one (LWC, one-moment schemes) or two (LWC and drop concentration or $r_{eff}$, two-moment schemes) prognostic variables per hydrometeor species. Our comparison of $C$ and $R$ for both sites reveals, however, that these quantities vary by up to one order of magnitude for constant LWC and $r_{eff}$ (which would be equivalent to a two-moment scheme). Considering only LWC (i.e. one-moment scheme) increases the differences to 1.5 to 2 orders of magnitude. As a consequence, additional moments or the full particle size distribution need to be considered in order to accurately estimate $C$ and $R$ in these models. Otherwise, a ~~parametrization~~ parametrisation of $C$ or $R$ relying only on LWC (LWC and $r_{eff}$) might be biased up to 1.5 to 2 (1) orders of magnitude for one-moment (two-moment) schemes.

~~Even though the rate of mass removal from a cloud is an important process impacting cloud life cycle, it is important to note that modifications to $C$ and $R$ cannot be directly translated into modifications in cloud lifetime. This is because a reduction in $R$ could result in a number of feedbacks such as cloud deepening (Stevens and Feingold, 2009) or reduced evaporation just below cloud base (Jiang and Feingold, 2006; Feingold and Siebert, 2009) that would act to counter the the extending effect of reduced precipitation rate on cloud lifetime.~~

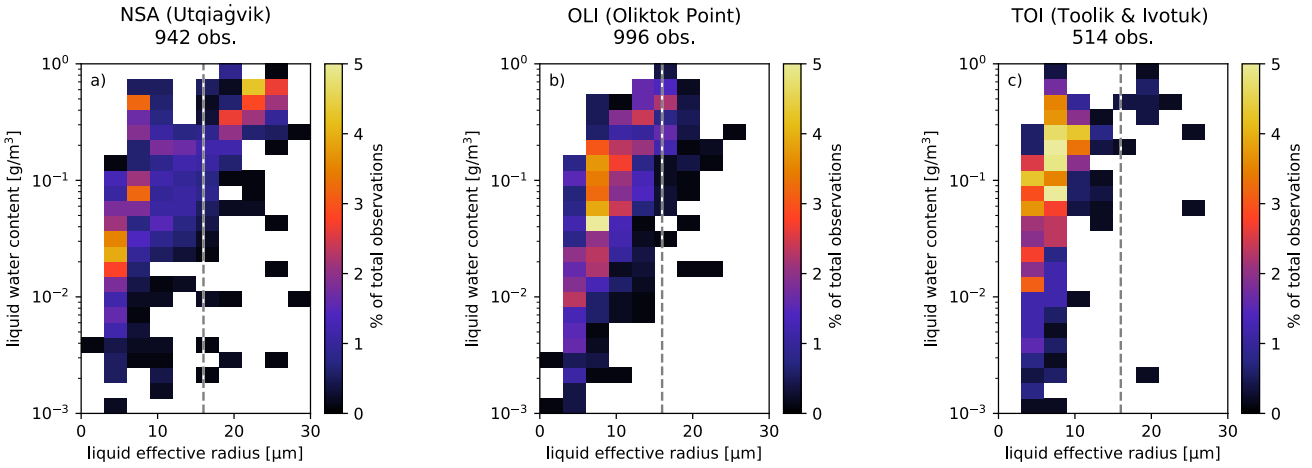


**Figure 4.** 2D-Histogram of number of observations as a function of effective radius $r_{eff}$ and liquid water content LWC for NSA (Utqiaġvik/Barrow) (a~~, 942 observations~~), OLI (Oliktok Point) (b~~, 1608 observations~~) and ~~Ivotuk/~~TOI (Toolik/Ivotuk) (c~~, 579 observations~~). The dashed line indicates a $r_{eff}$ value of 16 $\mu$m.

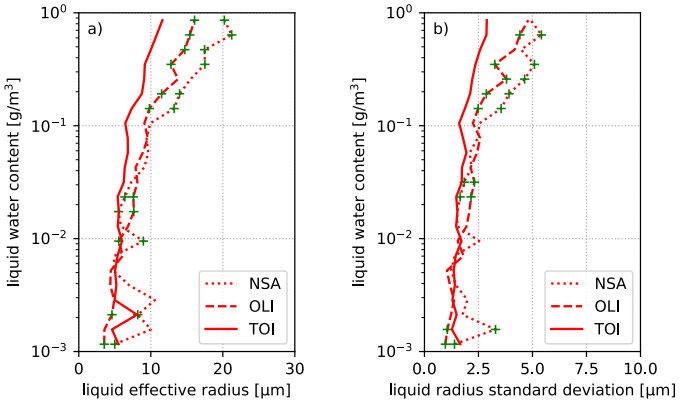


**Figure 5.** ~~As Fig. 4, but with the difference OLI - NSA of the collection kernel growth rate $C$ (~~a) Mean $r_{eff}$ and ~~the rainrate $R$ (~~b) mean standard deviation as ~~color.~~ a function of LWC for the ~~rates averaged over $r_{eff}$ are shown~~ data presented in ~~red for~~ Fig. 4. The green crosses indicate a significant difference between OLI ~~(dotted)~~ and NSA (~~dashed~~5% confidence interval).~~The green dots highlight data points with less than five observations.~~

## 5 ~~Cloud-Aerosol~~ Aerosol cloud interaction

So far, we have demonstrated that there are differences in aerosol properties and cloud properties between NSA and OLI. This is in general agreement with the findings of Hobbs and Rangno (1998) who found an increase in droplet number concentration when flying over Prudhoe Bay. In this section, we present evidence that these changes are indeed connected to local industrial activities ~~centered~~ centred around the Prudhoe Bay oil fields.

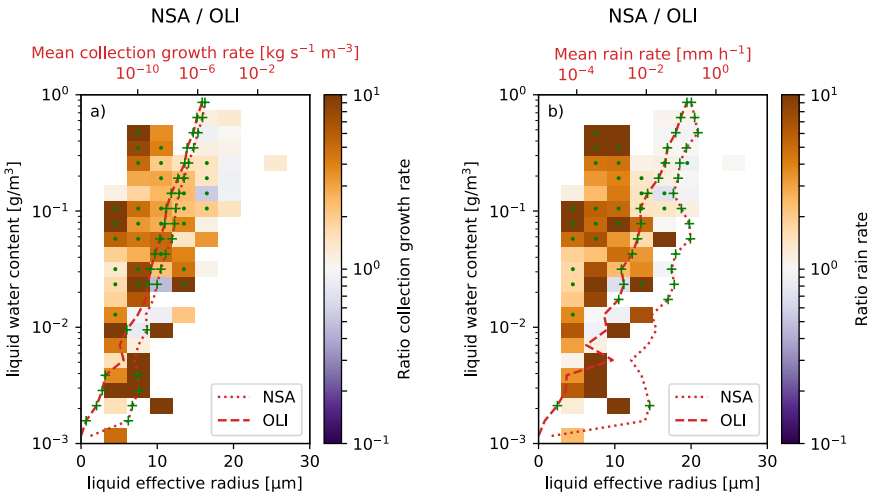


**Figure 6.** As Fig. 4, but with the coloured shading representing the site-to-site ratio (NSA/OLI) of the collection kernel growth rate $C$ (a) and the rainrate $R$ (b). The rates averaged over $r_{eff}$ are shown in red for OLI (dotted) and NSA (dashed). The green dots and crosses highlight data points with a significant difference (5% confidence interval).

In order to evaluate the likelihood that Prudhoe Bay emissions impacted different portions of the ACME-V flights, we use the HYSPLIT dispersion model. Simulations were completed using one continuously emitting source located over the Prudhoe Bay oilfields (70.2556°N, 148.3384°W), using a configuration similar to that discussed above for evaluation of wildfire emissions. Note that given the coarse resolution of the forcing model (1°) and the complexity of the Arctic boundary layer, HYSPLIT is used here only in a qualitative way, and not to select locally impacted clouds. For OLI (NSA), ~~62~~50% (16%) of all data points observed within clouds during ACME-V ~~cloud observations~~ can be traced back to surface emissions (i.e. mass concentration $> 0$ according to HYSPLIT) originating from the Prudhoe Bay oilfields. The 16% determined for NSA is roughly twice that presented in Kolesar et al. (2017). However, they studied aerosol concentration at the surface instead of aloft and used a multi-year data set, which could introduce substantial variability from the 3-month period evaluated here. The HYSPLIT simulations (Fig. 7) show that the mass concentration originating from local pollution sources ~~is on average more than two order of magnitudes~~ can be a substantially higher at OLI than at NSA. These simulations indicate that relative to NSA, the number of clouds impacted by local emission is ~~increased~~ higher at OLI and these clouds are impacted by a larger amount of aerosol particles by mass. However, an impact of local emissions on cloud properties is also possible at NSA, although less frequently than at OLI. The bin sizes in Fig. 7 were reduced in order to investigate the variability between clouds. This reveals that only a subset of clouds is associated with local pollution according to HYSPLIT. Note that HYSPLIT provides only a relative emission rate in 'mass m$^{-3}$' because the actual emission rate in the Prudhoe Bay region is unknown.

Fig. 8 relates in-cloud observations of LWC and $r_{eff}$ to below-cloud observations of rBC similar to the approach of Jackson et al. (2012). ~~This means that a~~ As a consequence, a single below-cloud aerosol value is assigned to every data point within the same cloud based on the assumption that aerosol properties are not changing on the scale of individual cloud profiles.

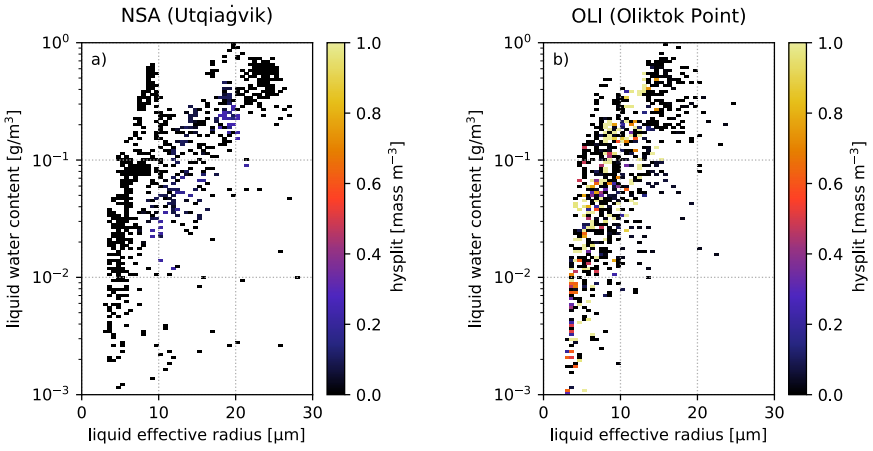


**Figure 7.** As Fig. 4, but with smaller bin size and mass concentration of local emission according to the HYSPLIT model for OLI (a) and NSA (b).~~Note that the color scales for OLI and NSA differ by a factor of 100. The red dots indicate data points with less than five observations.~~

The below-cloud values are averaged, whenever possible, over 30 seconds with a 3 second gap to the cloud base to avoid issues with time synchronisation across instruments or cloud particle contamination of aerosol probe measurements. ~~Similar to Fig.~~ It shows that the smallest $r_{eff}$ ~~8, the below-cloud CPC CN concentration is shown in~~ are connected to enhanced rBC concentrations ($>$ 10 ng kg$^{-1}$) for both sites. Note that for both sites, these high concentrations correspond in each case to a single cloud. However, for OLI there are more enhanced rBC concentrations ($>$ 4 ng kg$^{-1}$) for intermediate $r_{eff}$-values (5-10 $\mu$m) consistent with Fig. 3.a. It is interesting to note that this intermediate region is consistent with enhanced local particle concentrations according to HYSPLIT. rBC can originate from biomass burning as well as anthropogenic sources, but particle size is smaller for the latter (Schwarz et al., 2008). A comparison of rBC core size (Fig.~~10.These figures demonstrate clear relationships between cloud microphysics and rBC and CN (compare also Fig. 3) . Around OLI , below-cloud rBC and CN values are increased, more than 10 ng kg$^{-1}$ and 2000 m$^{-3}$, respectively.~~ 9) shows that black carbon particles are generally 50 to 300 nm smaller at OLI than at NSA. Together with the collocated enhanced HYSPLIT concentrations, this supports the idea that rBC measurements around OLI are associated with local emissions from Prudhoe Bay and not transported fire emissions. The coincidence of increased rBC concentrations with reduced $r_{eff}$ for OLI might indicate that the observed rBC acted as a CCN. However this would require the rBC to be coated with more hygroscopic material (e.g. ~~sulfate~~sulphate), because pure rBC does not serve as efficient CCN (Weingartner et al., 1997). ~~While many of the CN detected by the CPC~~ Note that the SP2 detects the non-coated size of the particles' rBC core, meaning the particles are larger when coated and can potentially act as a CCN despite their small core size.

Similar to Fig. 8, the below-cloud CPC CN concentration is shown in Fig. 10. This figure also indicates an impact of local emissions in the OLI data: CN-observations are enhanced (partly $>$ 1000 cm$^{-3}$) at OLI for all $r_{eff}$ even though variability is high (compare also Fig. 3.b). The CN observations are dominated by Aitken mode particles which are typically too small to

act as a CCN. This is consistent with the fact that Fig. 10 does not show a correlation between CN concentration and $r_{eff}$. Even though the CN dominating the CPC observations are likely too small to act as ~~a~~ CCN, these small particles can grow to accumulation mode quickly given sufficient gaseous precursors, potentially creating a particle population capable of acting as CCN (Jaenicke, 1980). ~~It is interesting to note that despite the notion that elevated concentrations should result in smaller $r_{eff}$, these measurement indicate that local pollution are not connected to $r_{eff}$ smaller than 9 $\mu$m. This is additionally supported by the reduced HSYPLIT mass concentrations (Fig. 7) for $r_{eff}$ below 9 $\mu$m.~~

For the PCASP (Fig. 11), the aerosol concentration ~~decreases more monotonically from small to large~~ is $> 100$ cm$^{-3}$ for small $r_{eff}$ values and $< 20$ cm$^{-3}$ for large large $r_{eff}$ values. Note that for NSA, PCASP data corresponding to some of the largest $r_{eff}$ have been flagged as invalid during quality control and are missing in the figure. The fact that the response of $r_{eff}$ to PCASP aerosol concentrations ~~is very similar~~ is ~~—for constant LWC—~~almost monotonic for both sites is likely because the PCASP covers the aerosol size range most relevant to droplet nucleation ~~. It should be noted that the monotonic decrease in PCASP concentration with increasing droplet size~~ and is consistent with the first indirect effect. ~~However, since no enhanced PCASP particle concentrations are found to be correlated to droplet sizes in the emissions-impacted 9 to 12 $\mu$m range (unlike for rBC and CN), there is no indication that local emissions are directly altering liquid clouds to have smaller $r_{eff}$~~A different behaviour would indicate that clouds react differently to the same PCASP concentration (which covers most of the accumulation mode size range, see also Sec. ~~as a result of PCASP-sized particles. Even~~ 6). However, even though similar PCASP concentrations lead to similar $r_{eff}$ for both sites, differences still exist relating to the breadth and tail of the the distributions, as can be seen from differences in $C$ and $R$ (Fig. 6).

~~Fig. ?? also includes the below-cloud concentrationsfor CN~~ Analysis of the relationship between clouds and HYSPLIT concentrations, rBC and CN shows that there are some, but not all, clouds at OLI impacted by local pollution. Because enhanced rBC and CN concentrations are expected to be good indicators of anthropogenic activity, they are used to isolate clouds impacted by anthropogenic emissions. Clouds, whose mean below-cloud rBC or CN concentration is above the median concentrations shown in Fig. 3 (4.1 ng kg$^{-1}$ and ~~PCASP assigned to the clouds presented in this study (Note that—in contrast to the aerosols—clouds from all altitudes are shown). While for NSA, most clouds correspond to small CN concentrations, for OLI there are also clouds present corresponding to high CN values (i.e. potentially fresh emissions) most of which also feature high PCASP concentrations~~ 1122 cm$^{-3}$, respectively), are identified as potentially impacted. When using this criterion, 10 of 24 (3 of 16) clouds at OLI (NSA) are identified as potentially locally influenced (Fig. 12). For NSA, two of the three clouds corresponded to either extremely low CN ($~~>~~ 200 ~~-~~ < 20$ cm$^{-3}$) or rBC ($< 1$ ng kg$^{-1}$) values, making a connection to anthropogenic activities unlikely. But, the clouds classified as anthropogenic at OLI correspond mostly to enhanced concentrations of rBC (Fig. 8) and CN (Fig. 10) and mid-sized $r_{eff}$ (5 - 15 $\mu$m). Fig. 12 shows how the PCASP concentrations of the potential locally affected clouds compares to the clouds classified as affected by forest fires (these clouds are removed in all other Figures except Fig. 14) and the remaining, non-classified clouds referred to as 'other'. Note that forest fire emissions were also present in the vicinity of NSA, but cloud measurements from these time periods did not pass the quality control measures implemented (continuously ascending or descending profiles). It is striking that the clouds classified as associated with forest fire have a significant (t-test), 6-times larger linear mean PCASP concentration than the clouds classified

as locally affected at OLI (510 vs 80 $cm^{-3}$). Despite this big difference, clouds classified as locally affected still feature PCASP concentrations significantly larger than the clouds classified as other (35 $cm^{-3}$)~~with mean sizes > 200 nm. This indicates that even though CN measurements are dominated by Aitken mode particles, there are frequently sufficient CCN concentrations present to allow cloud formation. Because small particles can grow quickly into~~ . We conclude that CN and rBC particles, which were used to classify local clouds, have the potential to grow to accumulation mode particles ~~, these CCN might also originate from local sources.~~ measured by the PCASP. For NSA, however, the mean PCASP concentration for clouds classified as other is not significantly (t-test) different from the clouds classified as locally affected at OLI. This is also true when including the three clouds classified as locally affected (from which only one is potentially local as discussed above). This is consistent with the findings of Fig. 3 which shows that the general PCASP concentration background is enhanced at NSA in comparison to OLI.

For the clouds classified as locally affected at OLI, the difference in rBC, CN and PCASP particle concentration above and below the cloud is presented in Fig. 13. This figure confirms that clouds impacted by local emissions feature higher aerosol concentrations below the cloud than above. This also supports our assumption that below-cloud aerosol properties are most relevant for clouds impacted by anthropogenic emissions, which is also true for the remaining anthropogenically influenced cloud at NSA (not shown).

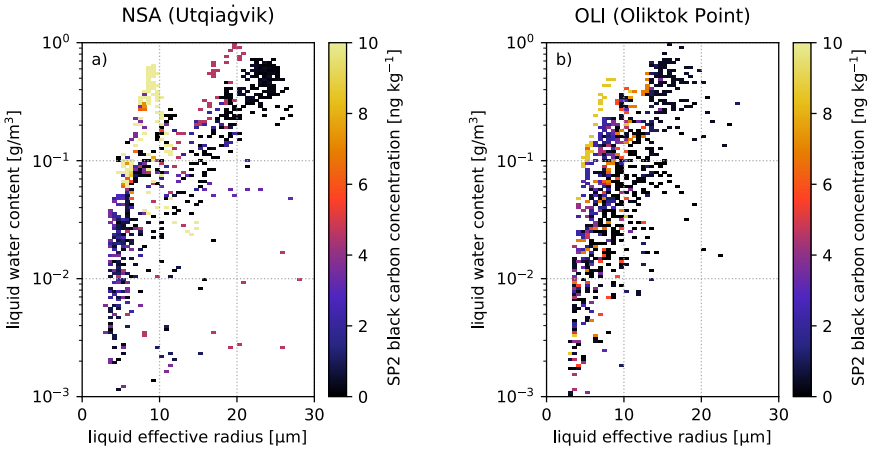


**Figure 8.** As Fig. 4, but with absolute values for SP2 refractory black carbon rBC concentration for (a) OLI and (b) NSA

~~Finally, comparison of rBC size (Fig. 9) shows that black carbon particles are generally 50 to 300 nm smaller at OLI than at NSA. This is consistent with aging of rBC during atmospheric transport, and supports the idea that rBC measurements around OLI are associated with local emissions from Prudhoe Bay and not transported fire emissions. At NSA, this pattern of larger SP2 sizes is only interrupted for a small range of droplet $r_{eff}$ between 9 and 12 $\mu$m, where both sites have similar mean sizes. This might be related to sampling issues, because this range coincides with very low rBC concentrations (Fig. 8) and low number of observations at NSA.~~

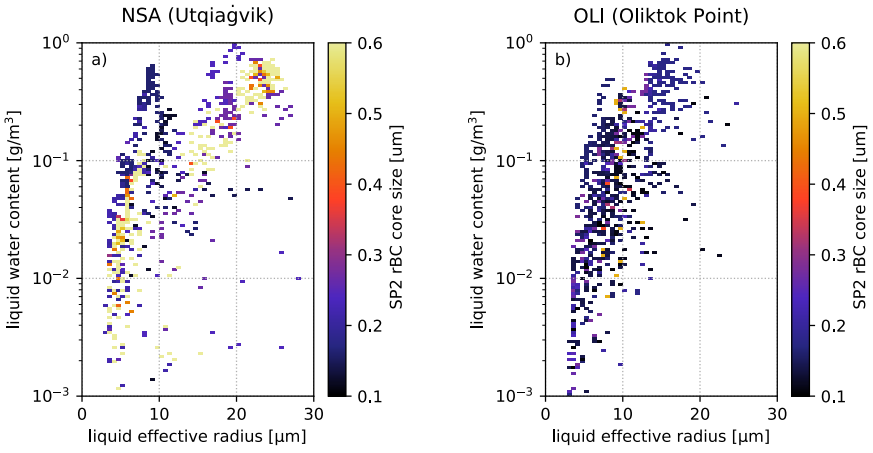


**Figure 9.** As Fig. 8, but with the mean size of refractory black carbon rBC measured below cloud.

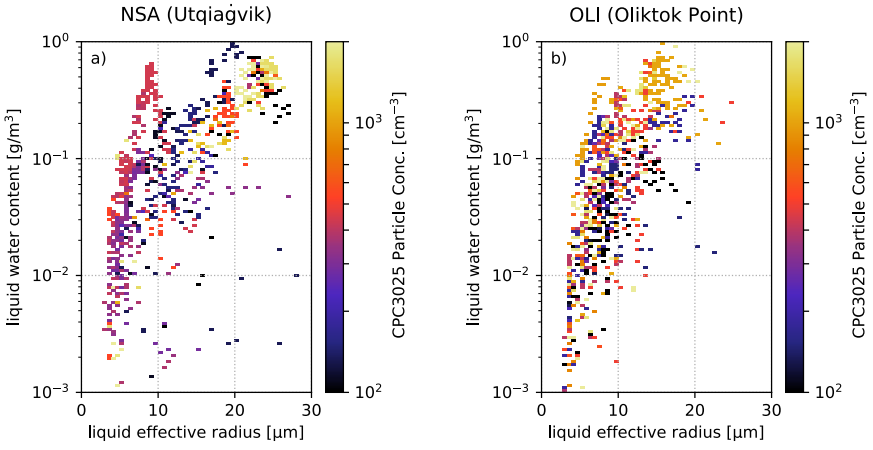


**Figure 10.** As Fig. 8, but with absolute values for CPC3025 condensation nuclei CN concentration.

## 6 Quantification of ~~cloud~~ aerosol cloud interaction

Various attempts have been carried out to quantify aerosol cloud interaction (ACI) in Arctic regions (Coopman et al., 2016; Zamora et al., 2016) and its impact on radiation (Earle et al., 2011; Tietze et al., 2011). One common definition used for quantification purposes is:

$$ACI = \frac{1}{3}\frac{d\ln N_{\mathrm{tot}}}{d\ln N_{\mathrm{a}}}\frac{\mathrm{d}\log_{10} N_{\mathrm{tot}}}{\mathrm{d}\log_{10} N_{\mathrm{a}}} \quad (4)$$

with $N_{tot}$ the number concentration of cloud droplets and $N_a$ the number concentration of aerosols (Feingold et al., 2001; McComiskey et al., 2009). For observations, ACI is obtained using a linear regression of ~~ln~~ $\log_{10} N_{tot}$ and ~~ln~~ $\log_{10} N_a$. We prefer defining ACI using $N_{tot}$ instead of $r_{eff}$, because the latter varies stronger vertically and would require to classify

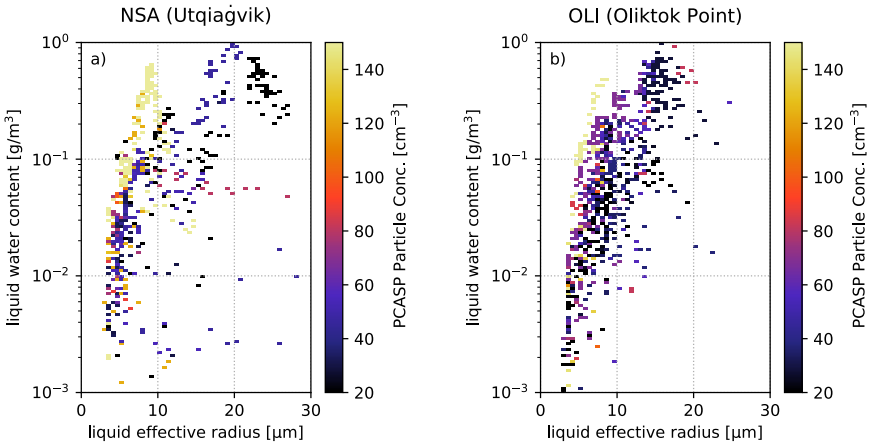


**Figure 11.** As Fig. 8, but with absolute values for PCASP particle concentration.

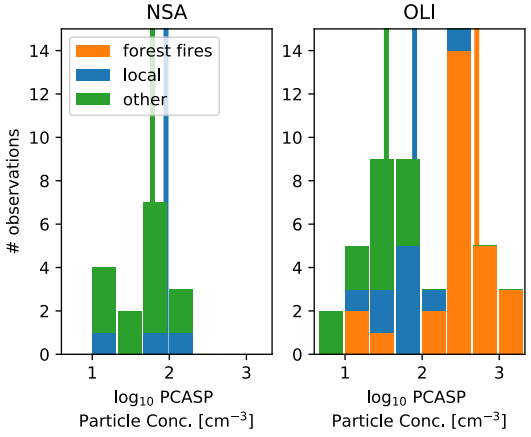


**Figure 12.** ~~As Fig. 8~~Stacked histograms of PCASP particle concentration for a) NSA and b) OLI for clouds classified as forest fire (orange), ~~but with~~ local (blue) and the residual (green). The vertical lines are for the corresponding linear mean ~~size of refractory black carbon rBC measured below cloud~~values.

the clouds by ~~LWP~~liquid water path, significantly reducing the size of the data set. Fig. 14 shows $N_{tot}$ and $N_a$ for both sites. $N_a$ is obtained ~~form~~ from the PCASP because it covers the size range of active accumulation mode aerosols best. The ACI value for ~~all clouds is 0.13~~ clouds at both sites is 0.14±0.04 with $R^2$ = ~~0.23~~0.30. Even though $R^2$ is small, the ACI value found here is similar to Zamora et al. (2016) who found ACI values of 0.15 for the PCASP using a multi-campaign data set focused on biomass burning. McComiskey and Feingold (2012) found that the choice of platform and observational scales can have a significant impact on the estimation of ACI making comparisons between data sets challenging. Zamora et al. (2016), however, also used cloud-averaged in-situ aircraft observations and as a consequence we expect them to be comparable. When applying the linear regression to the data sets corresponding to the two sites separately, the obtained ACI values differ (Table

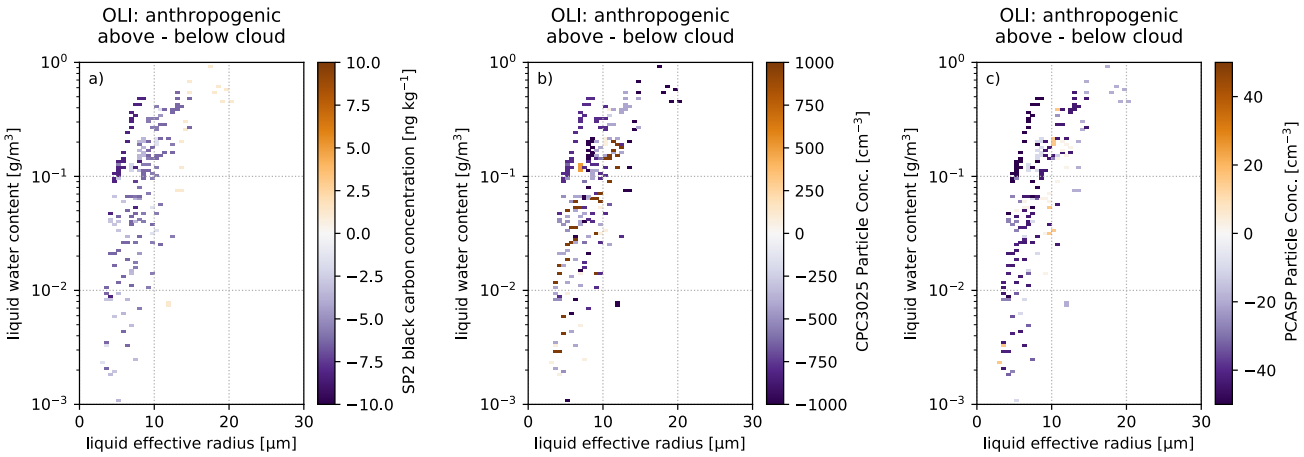


**Figure 13.** As Figs. 8, 10, and 11 for OLI, but showing the difference of rBC (a), CN (b), and PCASP concentration (C) between observations above and below the cloud.

1), with OLI having a lower ACI value (~~0.1~~0.12±0.05) than NSA (~~0.2~~0.20±0.07). Given the small sample size (~~35~~ 24 and 16 cases for OLI and NSA~~, respectively)~~ ) which was caused by the PCASP data being quality-flagged for some cases, it is not possible to ~~answer the question of~~ determine whether this is ~~related to~~ caused by a difference in nucleation efficiency between aerosols at the two sites ~~,~~ or a random effect. In addition, given the small sample size, we did not estimate ACI for local clouds only. The lower $R^2$ value for OLI (~~0.18~~0.24) in comparison to NSA (0.40) could indicate that the assumption that PCASP particle concentrations represent a good approximation for CCN concentrations is partly violated at OLI. This could result from those particles being less aged and consequently less coated by ~~sulfates~~ sulphates and organics in comparison to those observed around NSA. In addition, some data points lie above the 1:1 line which might indicate that particles smaller than the PCASP size range (i.e. $<$ 100 nm) are acting as CCN (Leaitch et al., 2016). Further, the assumption that the below-cloud aerosol properties govern the cloud microphysical properties might not be true for all clouds depending on sub-cloud vertical mixing. Therefore, we identified all clouds where the above-cloud PCASP concentration is larger than below-cloud (red dots in Fig 14), and indeed half of these clouds are above the 1:1 line. When using the above-cloud concentration for these clouds, only two of these clouds are above the 1:1 line. However, there are still 11 more clouds above the 1:1 line. Since these clouds generally feature PCASP concentrations $<$ 50 $cm^{-3}$, the fact that they are above the 1:1 line could be related to increasing sampling errors for small concentrations. They may also confirm the finding of Leaitch et al. (2016) that aerosols below 100 nm can act as CCN for thin clouds.

For comparison, we also evaluate ACI calculated ~~using~~ including data points associated with forest fires. Based on the flight patterns executed, all of the cloud measurements associated with forest fire emissions were sampled in the vicinity of OLI ~~. While fire emissions were also advected to the area surrounding NSA, cloud measurements from these time periods did not pass the quality control measures implemented (continuously ascending or descending profiles).~~ as discussed above. Based on the measurements collected, aerosols associated with forest fires generally feature higher ~~accumulation mode~~ PCASP concentra-

**Table 1.** ACI values for the subsets presented in Fig. 14

| Data set | ACI | $R^2$ | # clouds |
|---|---|---|---|
| ~~all data~~ both sites | ~~0.13~~ 0.14±0.04 | ~~0.23~~ 0.30 | ~~51~~ 40 |
| ~~Oliktok Point~~ OLI | ~~0.10~~ 0.12±0.05 | ~~0.18~~ 0.24 | ~~35~~ 24 |
| ~~Utqiaġvik/Barrow~~ NSA | 0.20±0.07 | 0.40 | 16 |
| ~~all data~~ both sites (with fires) | 0.14±0.02 | ~~0.43~~ 0.47 | ~~75~~67 |
| ~~Oliktok Point~~ OLI (with fires) | 0.14±0.02 | ~~0.44~~ 0.48 | ~~59~~51 |

tions (and in consequence smaller $r_{eff}$ ), which is consistent with ~~aging~~ ageing of these particles during transport, and in contrast to the freshly emitted particles generally found around OLI. As already discussed, clouds associated with local emissions have lower PCASP (and likely accumulation mode) concentrations than forest fires, but still have larger concentrations than for the other clouds. When including cases associated with forest fire emissions, ACI is found to be 0.14±0.02 for both OLI and the complete data set, and is similar to results obtained when omitting forest fire influenced cases. Therefore, we conclude that a ~~significant difference of~~ difference in ACI between local emissions and forest fires cannot be found, given the limited data set. This refers only to the mechanisms through which aerosols change cloud properties, and does not imply that local emissions do not change cloud properties.

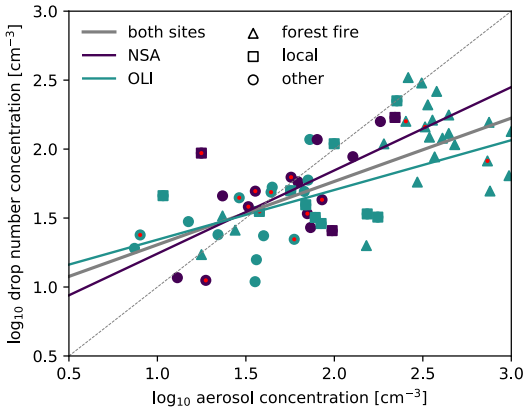


**Figure 14.** Aerosol indirect effect defined using cloud averaged cloud drop concentration $N_{tot}$ and PCASP aerosol concentration $N_a$ obtained below cloud. The ~~color is to differentiate~~ colour differentiates between OLI (green) and NSA (purple). Clouds classified as anthropogenically impacted, related to forest fires, and remaining other cloud are marked with ~~a '+'~~square, triangle, and circle, respectively. Red dots indicate clouds where the aerosol concentration above the cloud is higher than below the cloud. The trend lines indicate the linear regressions to obtain ACI (excluding forest fires) for the complete data set (~~gray~~grey), NSA (purple) and OLI (green), the doted line is the 1:1 line.

## 7 Conclusions

The impact of local emissions from industrial activities in northern Alaska on liquid clouds has been investigated based on vertical profiles of aerosol and cloud properties during the ACME-V aircraft campaign, together with measurements from the ARM sites in Northern Alaska: Oliktok Point (OLI) and Utqiaġvik (formerly known as Barrow or ARM's North Slope of Alaska site, NSA). Our main findings can be ~~summarized~~ summarised as follows:

1. Concentrations of condensation nuclei (CN) and refractory black carbon (rBC) are higher in the OLI area (Fig. 3). This is related to emissions associated with local oil and natural gas extraction activities. In contrast, ~~concentrations of larger (diameter >~~ PCASP particle concentrations (diameter > 100 nm~~) particles~~ , mostly accumulation mode) are not elevated around OLI when compared to NSA.

2. In addition, we found (Fig. 4) that liquid clouds generally feature significantly smaller $r_{eff}$ at OLI ~~(mean 9.8±4.0 $\mu$m)~~ when compared with NSA ~~(12.2±6.9~~ for LWC > 0.1 g kg$^{-1}$. Clouds with $r_{eff}$ > 18 $\mu$m ~~)~~are only rarely observed at OLI. Furthermore, collision-coalescence and precipitation rates are reduced by up to two orders of magnitude around OLI (Fig. 6). Only half of this reduction can be explained by the reduced $r_{eff}$. As a consequence, the breadth of the size distribution of liquid droplets ~~has to be~~ is smaller at OLI ~~.~~ as was observed (Fig. 5.b). The reduction of $r_{eff}$ at OLI occurs despite the larger background of PCASP concentrations at NSA (Fig. 3.c)

3. Multiple lines of evidence connect these changes in cloud properties to the observed local emissions. First, HYSPLIT simulations show that ~~62~~50% of all cloud observations around OLI can be traced back to local emission sources (Fig. 7). Second, ~~reduced~~ some clouds with mid-size $r_{eff}$ (between 9 and 12 $\mu$m) ~~of OLI clouds~~ at OLI correspond to increased CN and rBC concentrations (Figs. 8, 10). Third, the mean size of cloud-associated rBC particles is smaller at OLI which is consistent with ~~freshly emitted, less aged particles~~ the assumption of anthropogenic sources (Fig. 9). Finally, ~~while no enhanced concentrations of larger accumulation mode particles were observed for OLI~~ the clouds identified as most likely influenced by anthropogenic activities have significantly higher PCASP concentrations for OLI than for the remaining clouds (Fig.~~3), clouds there were found to be frequently connected to enhanced CN and accumulation mode concentrations~~ 12). However, the PCASP concentration of local clouds is not significantly higher than at NSA which might be related to a higher background of PCASP particle concentrations at NSA.

4. Given the limited data set, we found ten of 24 clouds at OLI, but only one of 16 clouds at NSA which might be impacted by anthropogenic emissions.

5. The PCASP concentration of clouds associated with forest fires is on average six times larger than for locally impacted clouds (Fig.~~??).~~ 12). Consequently, the impact of local emissions on cloud properties is small compared to the influence of forest fires (Fig. 14).

6. Quantification of aerosol cloud interaction (ACI) is challenging due to the small data set. Having said this, ~~the results from this study,~~ based on evaluation of clouds impacted by both local emissions and forest fires, the results are consistent

with previous studies of ACI in the Arctic environment (Fig. 14). While forest fire cases have typically higher ~~aerosol~~ PCASP concentrations and consequently droplet concentrations, their inclusion into the estimation of ACI does not substantially alter the found relationship.

Because ~~only~~ liquid clouds were observed ~~during the majority of the flights~~most often (60%), the impact of local pollution on mixed phase and pure ice clouds is not covered here. Moreover, the question as to what percentage of clouds at OLI (and NSA) is impacted by local emissions and whether the industrial activities at the North Slope of Alaska also lead to a change in local climate (e.g. due to cloud radiative forcing, precipitation impacts, or cloud life cycle), cannot be answered with in-situ aircraft measurements alone. These questions can likely better be answered using ground- and satellite-based remote sensing data from OLI and NSA by identifying differences between the sites in cloud cover, liquid water path, emissivity, effective droplet size, and precipitation occurrence. Nevertheless, based on this limited in-situ data set we can conclude that local emissions form industrial facilities in Alaska do influence local cloud properties while the overall spatial extent of these influences has yet to be evaluated. Given the observed cloud modifications, the effects of anthropogenic pollution on local climate should be considered when developing industrial infrastructure in an already fragile and warming Arctic environment.

## 8 Data availability

Data were obtained from the Atmospheric Radiation Measurement (ARM) climate research facility, a U.S. Department of Energy Office of Science user facility sponsored by the Office of Biological and Environmental Research. The surface observations from OLI and NSA as well as the ACME-V data set are available at the ARM archive www.arm.gov/data ~~(ARM, 1993, updated daily, 20~~ the phase classification of the cloud probes is available ~~form~~ from the corresponding author on request.

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