Review of:
The observed influence of local anthropogenic pollution on northern Alaskan cloud properties
Authors: Maximilian Maahn et al.
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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?
- 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.?
- 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.)

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.

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.

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

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.

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* r_{eff}." 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. 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* r_{eff} *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.

P10l11-12: "$\bar{C}$ *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?

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* r_{eff} *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.

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.?

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?

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).

b) *"Reduced* r_{eff} *(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).

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?

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.

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?
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.
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?

6) Fig1: Maybe include the MODIS fire locations?
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.
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 1-10^3 ng/m^3 (average appears on the order of 10^2 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.
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.
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?
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.
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.
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.

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?
15) P10l9: "*typical r_{eff} 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.
16) P10l13: more info on this droplet fall velocity parameterization would be helpful.
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.
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).
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?
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?

Technical corrections:

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

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.

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?).

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)?

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.

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."

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.

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.

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

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

P17l19: from not form.

References

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