Responses to the Reviewers

We would like to thank all three reviewers for their thorough comments on this manuscript, which helped to improve the paper. Our responses to general and specific comments are below.

The comments of the reviewers are printed in bold. All line numbers in bold refer to the original manuscript, all others to the revised version.

Reviewer 3

We thank the reviewer for their comments.

Reviewer: As a description of a flight campaign this manuscript includes a lot of information and the authors should have credit for trying to limit what is probably much more than enough behind the scene. Just as using cloud probes as a tool to stratify cloudy or non-cloudy measurement I see no problem of using differences between instruments as indicators for NPF (despite any measurement problems). The absolute numbers are really not followed up in the work. Hence, I will not dwell on measurement details.

Response: We thank the reviewer for their perspectives on the measurement approaches used in the study. We also believe also that there is merit to using the difference between the numbers of particles between 5 and 20 nm, i.e. $N_{5\text{-}20}$, to study the nature of aerosol processes as a function of location and height in this high Arctic regime. In our response to Reviewer 2, we discuss further the merits of this approach.

Reviewer: What I am missing is a Reader's Digest for modelers. Much of what is presented was already observed during previous campaigns, but the wealth of data could be presented in a summary nicely arranged with pertinent chemical and thermodynamically properties.

Response: This is a very good point, and so we have re-written the abstract of the paper and tightened the language in the conclusions section. Overall, we believe that the main points ("Readers Digest") of the study that a modeler should take away are that: 1) new particle formation occurs readily in the Canadian high Arctic boundary layer, a region dominated by marine and coastal regions, 2) particle growth also occurs in these regions under specific environments, 3) the highest levels of ultrafine particles were associated with above-cloud conditions influenced by marine air, and 4) ultrafine particle formation occurs much less frequently in the free troposphere under these conditions. Modeling efforts would ideally represent such behavior but are currently limited by our knowledge of marine aerosol precursor emissions.

That all said, we actually disagree that much of what we have observed has been seen before. In particular, this is the first systematic altitude-resolved study of the nature of ultrafine particles in mid-summer in the high Arctic. As we responded to Reviewer 2, what is important here is that relative to the cloud-free observations over ice and water, the UFP over cloud are common and the associated concentrations are higher (Figure 8). Also, in both the cloud and open water cases, the highest UFP concentrations are found at the lowest measurement levels, implying that the

surface (water or cloud) is critical to the NPF process. It is information of this type that is needed for comparisons against model output, to test the validity of the model representations of aerosol processes.

Reviewer: These cases could then be tried and tested using models. The aim in the beginning of the manuscript states a focus on UFP and this is ok, but quantifying their potential impact requires a model. The processes are very complex, and any changes in cloud base height for instance will over compensate any aerosol effect. Again, a model is needed.

Response: While the impacts of the UPFs are certainly interesting and have motivated this study to a large degree, it was beyond the scope of this observational paper to include the impacts that can only be evaluated with a model. However, we make reference to the work of Leaitch et al. (2016) that has pointed out that particles as small as 20 nm become activated into cloud droplets in this environment, motivating potential impacts and the needed to understand the processes that lead to their formation. Also, we now refer to Croft et al. (2016) that models one significant impact of NPF on Arctic radiative forcing.

Reviewer: I'm not convinced the CCN chapter of the manuscript is required for the NPF focus. In my opinion, the papers stands well as a description of the campaign, but I would prefer that the paper takes the understanding further than that of Shaw, Atmospheric Environment Vol. 23, No. 12, pp. 284-2846, 1989. What extra knowledge stands out form these flights besides, low mixing, low surface area, high insolation? A summary of this specifically would be a nice contribution. I don't contest that it is in the manuscript, but it could be summarized in a nice form.

Response: We agree that Shaw nicely illustrated that particle nucleation may occur in clean atmospheric environments, such as those in polar regions that have experienced recent scavenging. So, in that context, we fully agree that there are no new conceptual findings in our work compared to this work by Shaw, and other, earlier studies. However, what is new in this work are actual measurements of ultrafine particles in the high Arctic summer, especially in an altitude-resolved manner. These have not been documented so clearly before and such information is needed to compare against model output. Further, the work contrasts NPF over three different surfaces (ice, water and top of low cloud) in the same environment, which has never been done before and is important. We don't have sufficient statistics to conclude that the low cloud presence enhances the NPF relative to open water, but that certainly is the indication.

While we also agree that the CCN measurements are in some sense disconnected from the focus on the paper on the UFPs, we prefer to leave them in the paper as an illustration of the numbers of particles that may be arising, in part, from the growth of the UFPs that were measured. In order to improve the connection between the UFP observations and the CCN, we have added Figure 12 that shows correlations among the smaller particle sizes and the CCN. We also emphasise that the CCN measured here are larger than the average size of particle found by Leaitch et al. (2016) to participate in cloud droplet nucleation.

Reviewer: Orography is a source for concern at Svalbard, what about the conditions at the flight campaign?

Response: This is an interesting point re. orography. It is true that the nucleation and growth event documented in Willis et al. occurred in air that had resided over Devon Island (maximum altitude 2000 m) before descending through katabatic flow to the Lancaster Sound, and the same is evident in the event documented in Figure 8 of the present paper. Such air may have been cleaned by passing through this higher elevation location, lowering its condensation sink. However, aside from whatever reduced the condensation sink, the surfaces appear to be the sources of the particle precursors.

Reviewer: Ström et al. 2009 fig 11 Tellus would be nice to compare directly with the supplement figure 1. The fact that Aitken mode particles are not observed right at the surface could be an instrument detection issue I guess. Particles must grow to detectable size. On the source of particle near the surface, have a look at: Lampert et al., Inclined Lidar Observations of Boundary Layer Aerosol Particles above the Kongsfjord, Svalbard As an example of ocean source. Acta Geophysica 60(5), October 2012.

Response: This is a very valuable point, that it is possible that particles nucleate at the surface but require time to growth to sizes that are detectable. However, as referred to in our response to Reviewer 2, we apologize because we have now improved our profile averaging approach. We have revised Figure 8, which originally showed that the maximum in the UFP concentration over open water was above the lowest sampling level. That was due in part to a bias associated with the averaging time of the SMS, which has been removed. To Figure 8, we have now added N_{tot}, which was sampled every second and shows the increase in particles over open water is also associated with the lowest sampling level. That observation is consistent with the results of Willis et al. (2016) as well as past observations related to polynyas (Leaitch et al. 1983 and 1994).

References

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