**Response to comments of referee #2**

This manuscript describes an interesting and valuable set of measurements made aboard Polarstern in the region near Svalbard, during May to July 2017. The authors report measurements of neutral and charged clusters particle size (together covering a size range on <1 - 800nm), particle hygroscopicity and cloud condensation nuclei. Measurements of nucleation mode particle size and charged/neutral cluster abundance have been rarely made in Arctic regions, making this a unique and valuable data set that indirectly furthers our knowledge on the identity of species responsible for Arctic nucleation and initial growth of particles to Aitken mode sizes. The manuscript is well written and largely well organized. The following comments are intended to improve an already very good manuscript.

Reply:

We thank to referee #2 for understanding the value of such studies. We also appreciate referee for his/her time, comments, and suggestions, which are intended to improve the manuscript. In the following, we would like to address the comments step-by-step.

**General Comments**

1. L65-73: This is an important section of the introduction, and at present is missing several of the relatively small amount of studies that exist. Since so few data sets exist, it is reasonable to cite them even if they are not all discussed in detail. Some that should be included are listed here: Freud et al, doi:10.5194/acp-17-8101-2017; Nguyen et al, doi: 10.5194/acp-16-11319-2016; Dall’Osto et al, doi: 10.5194/acp-19-7377-2019; Burkart et al, doi: 10.5194/acp-17-5515-2017; Burkart et al, doi: 10.1002/2017gl075671; Collins et al., doi: 10.5194/acp-17-13119-2017; Leaitch et al., doi: 10.12952/jour-nal.elementa.000017; Tremblay et al., doi: doi.org/10.5194/acp-19-5589-2019; These and others were recently reviewed in doi: 10.1029/2018rg000602.

Reply:

Thank you for the comment. We have included the suggested references to manuscript. We have also re-wrote introduction to briefly overview the main findings of mentioned studies. Changes in introduction can be seen in yellow in a new manuscript version.

1. The discussion of ship deck observations in Section 2.1.1 may serve the reader better if they were incorporated into the larger discussion of NPF events in Section 3.1

Reply:

Thank you for your suggestion. We merged Section 2.1.1 into Section 3.1. We agree that ship deck observations serve the reader better if they are incorporated into the discussion. Also, we noticed that some of the information is redundant. Changes are highlighted in yellow.

### 3.1.1 NPF 1: 1 June

The first NPF event with a subsequent particle growth was observed from around 6 am onwards on 1 June, 2017. The RV Polarstern reached the marginal ice zone at 11 am and entered the pack ice at around 3 pm on 31 May, 2017 (note that all times in this study are given in UTC). This can be seen from the air and water temperature profiles (Fig. 2). The temperature of air and water decreased from approx. +5 °C to -5 °C (air) and -2 °C (water). In this area, the ice was broken up by leads, which facilitated the passage of the vessel towards the north. Around 8 pm a region with more densely packed ice was reached, which obstructed the movement of the ship (Nicolaus, 2018). On these occasions, due to frequent reverse-forward ship movement, pollution highly affected the measurements on-board (see PNSDs in Fig. 1). On 1 June, the vessel could once again pass through open leads in the pack ice, allowing for contamination-free scans for the time period from 4 am to 8 pm. During this time, RV Polarstern moved 26 km (from 80.39°N 7.58°E to 80.62°N 7.94°E) in mostly cloud-free conditions. From 6 pm to 8 pm, a thin ice cloud was present in over 8 km altitude. Also, over a short period from 2 to 3 pm, intermittent low-level liquid clouds were present, which however did not decrease the global radiation significantly. For a more detailed description of local and associated large scale weather patterns during PS106 refer to Knudsen et al. (2018).

Before the NPF event, the average particle number concentration in a size range from 10 to 50 nm (PNC10-50), was 50 particles cm-3.

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### 3.1.2 NPF 2: 18 June

On 17 June, the ship was moving southward through packed ice area, breaking floes and navigating through polynyas (Nicolaus, 2018). Over the complete day of 17 June, low-level stratocumulus clouds were present, which were broken up occasionally between 7 am and 1 pm, and 4 to 10 pm. Between 11 pm on 17 June and 1 am on 18 June, measured visibility decreased, accompanied by an increase in relative humidity (RH), indicating fog. This low-level cloud layer was present until approximately 8 am on 18 June, when RV Polarstern left the packed ice entering the marginal ice zone. This resulted in water and air temperature increase from -1.9 °C to approx. 2 and 0.5 °C above zero, respectively. At the same time, local wind speed decreased from 5 to 2 m s-1. During the following hours, until 6 pm, no clouds were present except for a very thin, high ice cloud at 8 km from approximately 11:30 am to 12 pm. This period of high incident radiation was only briefly interrupted by a short fog event from 3 to 3:30 pm. During this whole time, RV Polarstern moved through open water, but was always surrounded by floating ice. Starting at 6 pm, a thin low-level cloud layer was present above the ship, which decreased the global radiation significantly. This cloud layer was present until the next day, 19 June, at approximately 12 pm. During 19 June, RV Polarstern was moving through open water and ice along the west coast of Spitsbergen Island (Fig. 1). From approximately 12:30 pm to 3 pm another short cloud-free period led to high global radiation. At 4 pm at approx. 3 km altitude a cloud moved in decreasing the global radiation once again.

The PNC10-50 and PNC100-800 from 17 June prior to the NPF event were rather stable, with an average value of approx. 30 cm-3.

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### 3.1.3 NPF 3: 26 June

The third, least intensive NPF event occurred during the second leg of the expedition, 26 June, when RV Polarstern was at the marginal ice zone, around 200 km east of Svalbard, moving towards the North. Areas dominated by open water were passed by the vessel, as well as ice-covered water (Nicolaus, 2018). However, the ice was never very densely packed and the transit of the ship did not require breaking the ice. Low-level clouds and fog were present during all of 25 June to 27 June; on 28 of June a short period of cloud-free conditions was observed from around 4 to 6 am. There were two short floe stations, one on 25 June from around 5 pm until midnight and the other on 27 June from around midnight to 3 am.

The formation and growth of particles was already observed on both 24 and 25 June during less pronounced NPF events (not shown), when the ship was approx. 100 km South of Svalbard coast.

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### 3.1.4 NPF 4: 2 July

From midnight of 1 July to 4 July, RV Polarstern was moving northwards from 81.64°N 32.62°E to 82.16°N 32.87°E. This region was mostly ice-covered with some open leads through which the vessel could pass without having to break the ice. At this time of the expedition, melt ponds were observed frequently on the ice floes. On 1 July, there was a thick (up to 3 km altitude) low-level cloud layer present until 2 pm associated with some snow fall. After 1 pm, the cloud bottom height increased steadily; however, some intermittent fog was still present at sea level. A single fogbow was observed between 6:20 and 7 pm. The fog dissolved at midnight on 2 July. Almost throughout the entire day of 2 July, no clouds were present except for optically thin cirrus clouds, allowing for high solar irradiation.

On 2 July, the RV Polarstern ventured further into the Arctic ice, more than 300 km from the coasts of Svalbard and Prince George Land (81.51°N, 32.97°E).

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1. A detailed description of how pollution influence from the ship was reproducibly removed from the data set needs to be included in the Methods section. Was the influence quantified with a specific measurement (e.g., rapid variability in particle concentrations in a certain size range?)? Or, was a specific wind sector removed from the data? “Abrupt and short increases in particle number concentration recorded by a CPC” as cited in L286 as the method for filtering ship stack pollution; this should be elaborated in the methods section and detail is needed on how the authors determined that “most of the cases when the particle number increased tenfold” could be attributed to NPF. I strongly discourage the authors from including periods of ship pollution in Figure 2 (noted in L374-375). I disagree that “better representation of particle growth” (L1031-1032) warrants this.

Reply:

Thank you for the comment. We would like to split the answer to referee #2 into several parts.

1. *Was the influence quantified with a specific measurement (e.g., rapid variability in particle concentrations in a certain size range?)?...“Abrupt and short increases in particle number concentration recorded by a CPC” as cited in L286 as the method for filtering ship stack pollution; this should be elaborated in the methods section and detail is needed on how the authors determined that “most of the cases when the particle number increased tenfold” could be attributed to NPF.*

Reply:

Indeed, the pollution influence from the ship was quantified with a rapid variability in total particle number concentration. A separate condensation particle counter (CPC) was used to measure total particle number concentration (PNC) with time resolution of 2 seconds (please note that in the text, chapter 3, time resolution was said to be 1 s, which is not correct). An exemplary data of PNC is presented in Fig. R1. It can be clearly seen that ship pollution heavily affects the momentary PNC. Even during NPF event, the maximum particle number concentration did not exceed 5000 particles cm-3. Meanwhile, ship pollution resulted in order of magnitude higher concentrations. It is obvious that such increase must be related to ship exhaust pollution. Moreover, we have also compared PNC increase with a signal from Single Particle Soot Photometer (SP2, operated inside the aerosol container by scientist from Alfred Wegener Institute. Data is not presented in the manuscript). After comparing the results from both SP2 and CPC, we came to conclusion that CPC recorded sharp increase in PNC is a good indicator for ship-related pollution. Therefore, for online measurements only CPC and SMPS data was used for contamination inspection.

Different methods exist to clean ambient background aerosol measurements from pollution plumes. For example, Kivekäs et al. (2014) used rolling window percentile to extract background concentrations from particle number size distributions effected by ship plumes. In our previous works, we used rolling window minimum method (see Kecorius et al., 2017). Nevertheless, in this work, we cleaned the online measurements by hand keeping 2 second time resolution CPC data as a reference. This way we are 100% certain that our analyzed results are ship contamination free. Software used to clean up the data was “Dafit” by Ries (2013).

1. *Or, was a specific wind sector removed from the data?*

Reply:

Automatic system (to measure relative wind direction) was installed together with high volume sampler to stop the pumps when wind came from a specific direction (related to ship exhaust). Similar approach was used by Huang et al. (2018), who performed measurements inside same TROPOS measurement container onboard RV Polarstern. Berner impactor, on the other hand, did not have such system to prevent samples from contamination. However, we only used those samples, which had same order of OC with Digitel PM1 samples. More detailed discussion regarding OC measurements using Digitel and Berner samplers will be provided in separate work. In the text, we addressed this limitation and added the following in chapter 3.4:

It must also be noted that there was no action taken (e.g. sampling interrupt dependent on specific wind sector) to reduce ship contamination for the size-segregated aerosol particle measurements. Thus, the contamination from the ship exhaust cannot be ruled out completely. However, the high concentrations of biogenic compounds like MSA and the presence of sodium on the aerosol particles suggested a strong marine influence to the particle composition.

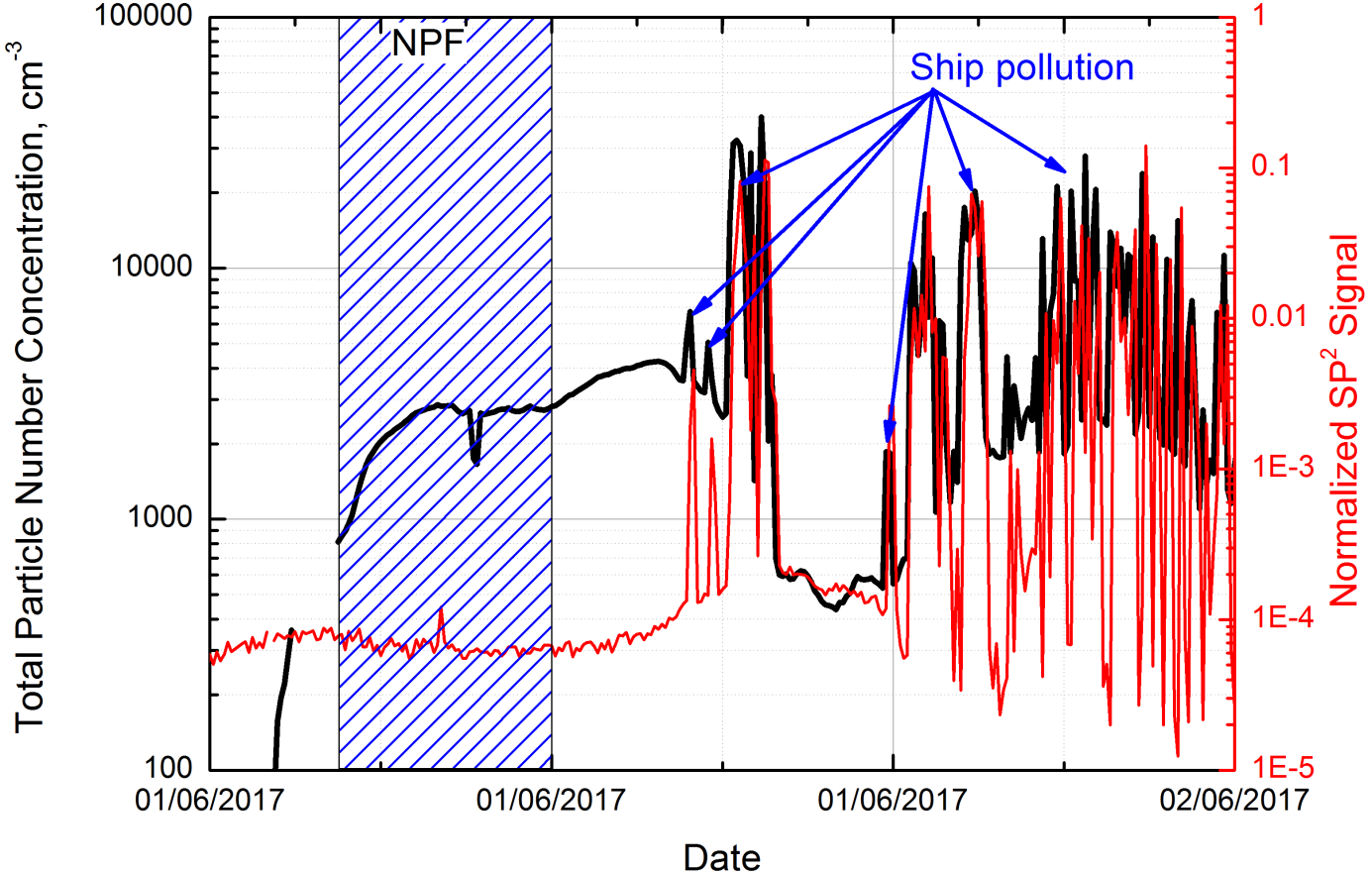


Fig. R1. Total particle number concentration during new particle formation event and contamination from ship.

1. *I strongly discourage the authors from including periods of ship pollution in Figure 2 (noted in L374-375). I disagree that “better representation of particle growth” (L1031-1032) warrants this.*

Reply:

Ship pollution was included only for visual representation. The data discussed in the manuscript is pollution free. Moreover, by removing data from Fig. 2 that is influenced by ship pollution, we would introduce blank gaps in contour plots. This would greatly diminish the visibility of particle new formation and subsequent growth, which is extended over time period of several days. The inclusion of pollution episodes in Fig. 2 does not obstruct the reader from seeing the processes happening during measurements.

To note the ship pollution influence onto our results, we have included a new section, 2.5 Contamination from ship exhaust, into the manuscript.

Huang, S., Wu, Z., Poulain, L., van Pinxteren, M., Merkel, M., Assmann, D., Herrmann, H., and Wiedensohler, A.: Source apportionment of the organic aerosol over the Atlantic Ocean from 53° N to 53° S: significant contributions from marine emissions and long-range transport, Atmos. Chem. Phys., 18, 18043–18062, https://doi.org/10.5194/acp-18-18043-2018, 2018.

Kecorius, S., Madueño, L., Vallar, E., Alas, H., Betito, G., Birmili, W., Cambaliza, M.O., Catipay, G., Gonzaga-Cayetano, M., Galvez, M.C. and Lorenzo, G., 2017. Aerosol particle mixing state, refractory particle number size distributions and emission factors in a polluted urban environment: Case study of Metro Manila, Philippines. Atmospheric environment, 170, pp.169-183.

Kivekäs, N., Massling, A., Grythe, H., Lange, R., Rusnak, V., Carreno, S., Skov, H., Swietlicki, E., Nguyen, Q.T., Glasius, M. and Kristensson, A., 2014. Contribution of ship traffic to aerosol particle concentrations downwind of a major shipping lane. Atmospheric Chemistry and Physics, 14(16), pp.8255-8267.

Ries, L.C., 2013. Dafit-a new work flow oriented approach for time efficient data preparation, validation and flagging of time series data from environmental monitoring. In EnviroInfo (pp. 651-656).

1. Further to comment (3) above, for the offline chemical analysis the sampling time was 72-144 hours (L223); how was sampling pollution from the ship minimized or avoided (e.g., by a wind switch)? The presence or absence of any such precautions should be stated, and if they were absent the possible implications should be discussed.

Reply:

Thank you for the comment. As stated above, to note the ship pollution influence onto our results, we have included a new section, 2.5 Contamination from ship exhaust, into the manuscript.

1. A slightly more descriptive title might be helpful. For example, “during PS106 cruise” could be replaced by a few words describing the region of measurements. This would be helpful for readers not familiar with the region covered by the cruise.

Reply:

Thank you for the suggestion. We agree with referee #2 that more descriptive title would benefit the reader. We have updated the title to “New particle formation and its effect on CCN abundance in the summer Arctic: a case study in Fram Strait and Barents Sea”.

**Specific Comments**

1. L44: This statement might be best attributed to a paper such as Croft et al., Nature Communications, 2016, that makes a more direct connection to radiative forcing.

Reply:

Thank you for the comment. We have included mentioned citation to manuscript.

1. L60-62: This statement is more attributable to Willis et al., Reviews of Geophysics, 2018 (https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2018RG000602) and Abbatt et al., ACP, 2019 (<https://www.atmos-chem-phys.net/19/2527/2019/acp-19-2527-2019.html>).

Reply:

Thank you for the comment. We have included mentioned citation to manuscript.

1. L63: Dall’Osto et al., Scientific Reports, 2018 corroborates these results for different regions and multiple stations (https://www.nature.com/articles/s41598-017-17343-9 and <https://www.nature.com/articles/s41598-018-24426-8>)

Reply:

Thank you for the comment. We have included mentioned citation to manuscript.

1. L206: I assume that the CCN measurements were made on poly disperse aerosol, but this is not explicitly stated.

Reply:

Thank you for the comment. Yes, CCN measurements were made only for poly disperse aerosol. Now we have included this statement in the manuscript.

1. L218-219: A large fraction of the organic aerosol may be semivolatile (e.g., Burkart et al, GRL, 2017). How might the semivolatile fraction be impacted by both heating of the inlet and long sampling times? Possibly biases introduced into these measurements should be discussed.

Reply:

Thank you for the comment. Aerosol inlet for online measurements was heated to 30 °C to prevent ice formation and blockage of the inlet. Only very tip of the inlet was heated. Here, the residence time of aerosol in this heated area was 0.06 s. That is considerably shorter than aerosol spends inside instruments itself before being measured. For offline measurements, on the other hand, residence time is much higher – approx. 1.2 s (for Berner sampler). However, as stated in the text (Lines 219-220): The temperature difference between the ambient air at the impactor inlet and the sampled air after the conditioning unit did not exceed a value of 9 K. Taking into account the low outside temperatures, we do not expect a significant loss of semi volatiles due to this conditioning process (the maximum temperature that tube was heated to was 7 °C). The aerosol particles on the aluminum foils were not exposed to heating and sampled at the (cold) outside temperatures. We added this information in the manuscript (Line 193):

The losses due to evaporation of semi volatile compounds are expected to be minimal.

Furthermore, during sample transportation, samples were stored at -20 °C, thus, we do not expect losses during this stage.

1. L390-391: This is a very interesting observation and is in agreement with Tremblay et al., 2018 (doi above), could these data be included in the Supplement?

Reply:

Thank you for the comment. We do not possess the observational data neither from Villum Research Station nor Zeppelin mountain Observatory. Nevertheless, for comparison purposes, particle number size distributions can be freely obtained from EBAS, developed and operated by the Norwegian Institute for Air Research (NILU, <http://ebas.nilu.no/>).

1. L425-429: Burkart et al., 2017 (doi above) came to similar conclusions.

Reply:

Thank you for the comment. We have included mentioned citation.

1. L431-441 and Figure 4: Rather than referring to the stage number here I suggest the authors refer to the corresponding size range.

Reply:

Thank you for the comment. We have updates the text and Fig. 4 as suggested by referee #2.

1. Related to comment (8), are any accumulation mode particle present during growth events? Do these larger modes also grow during NPF? Is sea salt present in accumulation mode sizes? Collins et al., 2017 (doi above) and Burkart et al., 2017 (doi above) observed growth of multiple modes at different rates showing that the species responsible for growing the larger mode was more semi volatile. If this is the case can the composition of the larger modes be connected to what is growing the smallest particles?

Reply:

Thank you for the questions, which we would like to answer step-by-step:

1. *are any accumulation mode particle present during growth events?*

From figure 2 (in manuscript) and figure R2 (see below), it can be seen that indeed there is an accumulation mode particles present during both particle formation and growth events.

1. *Do these larger modes also grow during NPF?*

In three of four analyzed NPF events, we did not observe accumulation particle growth. Some accumulation particle growth could be seen during NPF event on 18 June. On 17 June, geometric mean diameter of accumulation mode particles was approx. 100 nm. In 32 hours (from 17 June 08:00 to 18 June 16:00), accumulation mode particle geometric mean diameter grew to 120 nm, that is 0.6 nm h-1. Burkart et al. (2017) observed Aitken mode particle growth to sizes above 100 nm in parallel with nucleation particles. The growth rate was estimated to be 3.4 nm h-1.

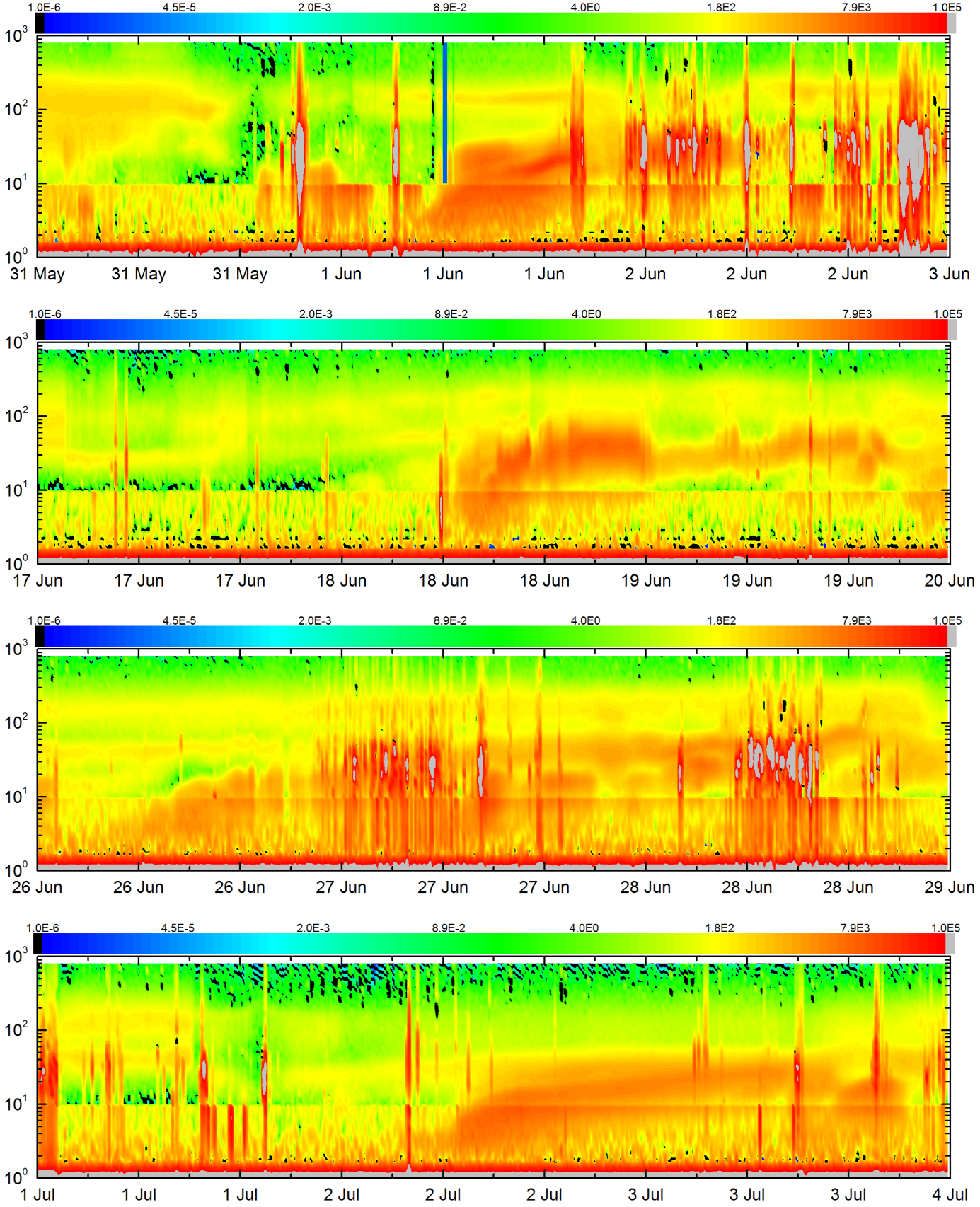


Fig. R2. Particle number size distribution during the new particle formation and growth events. Y-axis – mobility diameter (in nm), X-axis – time, color represents normalized particle concentration (dN/dlogDp, cm-3).

References:

Burkart, J., Hodshire, A.L., Mungall, E.L., Pierce, J.R., Collins, D.B., Ladino, L.A., Lee, A.K., Irish, V., Wentzell, J.J., Liggio, J. and Papakyriakou, T., 2017. Organic condensation and particle growth to CCN sizes in the summertime marine Arctic is driven by materials more semivolatile than at continental sites. *Geophysical Research Letters*, *44*(20), pp.10-725.

1. *Is sea salt present in accumulation mode sizes?*

Unfortunately, we were not able to investigate chemical composition of aerosol particles for NPF event on 18 June. This is because the high volume and Berner samplers did not overlap with the measurement period, during which NPF was observed.

1. *Collins et al., 2017 (doi above) and Burkart et al., 2017 (doi above) observed growth of multiple modes at different rates showing that the species responsible for growing the larger mode was more semi volatile. If this is the case can the composition of the larger modes be connected to what is growing the smallest particles?*

As mentioned above, on most of the cases we did not observe the accumulation mode particle growth during NPF events. The exception is 18 June, when a slight accumulation mode particle growth can be noticed. The particles grew from approx. 100 to 120 nm (geometric mean diameter) with a rate of 0.6 nm h-1. Because chemical composition of aerosol particles was not possible to determine, we cannot neither confirm nor deny the resemblance of such observation with previous studies.

1. L444-446: This sentence described a very unique aspect of this study which could be highlighted more, for example in the abstract or at the end of the introduction.

Reply:

Thank you for the comment. The methodology to observe new particle formation events is well defined in scientific literature, as well as frequently used in various environments (including Antarctica). We do not consider the used instrumentation to be unique in those terms. Therefore, although there are not many studies with same instruments in the Arctic region, we prefer not to highlight the uniqueness of the instruments, but rather focus on the scientific conclusions. Instruments are only the tools.

1. L450-460: Comparison to other Arctic studies that report growth rates might be more appropriate here, though I do not dispute the value of comparing to Antarctic studies. Collins et al., 2017 (doi above) report growth rates for events in the Canadian Arctic during two summers. Nieminen et al., ACP 2018 (doi: 10.5194/acp-18-14737-2018) include growth rates from Alert. Available observations were reviewed by Willis et al., 2018 (doi above).

Reply:

Thank you for the comment. We have compared our results to the studies suggested by referee #2. The comparison was included into manuscript as well.

1. L567-569: Describing the experiments (i.e., the information in brackets) might be more useful than using experiment numbers for those readers not closely familiar with the CLOUD experiments.

Reply:

Thank you for the comment. We have updated the manuscript as suggested by referee #2.

1. L572-573: If the measured Aitken mode particles were all organic, how much OM mass would you expect and how does that compare with the measured masses? Also, if the material is largely semivolatile how much do you expect that losses during sampling would impact this assessment?

Reply:

Thank you for the question. To answer it, requires a mass closure between physical and chemical measurements of the atmospheric aerosol particles. Such an exercise is a great idea and could easily result in a publication itself; however, it is beyond the scope of this study. Multiple assumptions (e.g. Aitken mode particle density and shape; losses in Berner sampling lines; sampling efficiency; etc.) must be made to convert mobility particle diameter to volume equivalent particle diameter. Moreover, as can be seen from contour plots of particle number size distribution (Fig. 2; Fig. R2), pollution had a noticeable impact on measured particle number concentration. This may have effect on estimating particle shape and density, thus, affecting the final closure results (as Berner samplers were not stopped during pollution events). We could certainly provide a number here, however, without an extensive investigation and discussion on possible errors it would be meaningless.

To answer second part of the question: as stated above, we cannot account for losses of semivolatile organic matter during the sampling procedure. However, the samples were not exposed to heating (in fact they were cooled and experienced outside temperature) and they were immediately frozen after the sampling. We don´t expect significant losses of OM.

1. L619-620: The authors of Willis et al., 2016 provide reasonable evidence for a marine source. For example, that the organic and MSA driven growth was only observed in a shallow marine inversion layer and not aloft. Burkart et al., ACP, 2017 corroborate a marine source of NPF precursors.

Reply:

Thank you for the comment. We have rephrased the text in manuscript to account for evidence or marine sources in NPF.

1. L634-636: The cited observations, as well as Collins et al., 2017, do demonstrate some growth into sizes above 50nm. The same work also demonstrates frequent simultaneous growth of multiple modes, and a resulting strong impact on CCN.

Reply:

Thank you for the comment. In this work we were able to show that newly formed and slightly grown particles do not need to grow beyond 50 nm to act as CCN. This, however, does not mean that particles do not/cannot grow beyond these sizes. The increase in CCN number concentration due to none-NPF related particle growth was not the scope of this study.

1. L660-670: This is a useful analysis, and I don’t suggest that the authors make substantial changes here. However, I do wonder if updraft the most appropriate way to assess this for summer Arctic low level clouds directly impacted by marine sources? Advection from warmer to colder surfaces in a shallow boundary layer might be another mechanism for CCN to active in low altitude clouds, suggested by Leaitch et al., 2016. Marine influence was significantly less for the upper level clouds observed in Leaitch 2016 (see Bozem et al., ACPD, 2019 doi: 10.5194/acp-2019-70).

Reply:

Thank you for the comment. The referee #2 is right wondering if updraft is the most appropriate way to assess NPF influence onto Arctic low level clouds. We do not state that this method is the most appropriate way to do so. We used the zero-dimensional, adiabatic cloud parcel model due to its availability and ease of use. More sophisticated model accounting for advection from warmer to colder surfaces (as suggested by referee #2) as well as topography influence may be a good starting point to answer this question in the future.

1. L706: Will these data be made publicly available in the future?

Reply:

Thank you for the question. Making data publicly available requires it to be submitted to specific data base, which in turn necessitates the data to be in a specific format (e.g. NASA- Ames). At the moment, the data was analyzed, formatted and reformatted multiple times to serve the manuscript preparation. It is thus much easier to make data available on request. If referee #2 has an interest in specific data set, we may be able to provide a temporal data download link. This way the reviewer anonymity can be sustained.

**Technical Corrections**

1. L144: “0 am” typo?

Reply:

Thank you for the notification. The typo was corrected.

1. L471: “is” to “are”

Reply:

Thank you for the notification. The typo was corrected.

1. Supplement Figure S1: typo in the y-axis label? [m] to [nm]?

Reply:

Thank you for the notification. The typo was corrected.