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New particle formation and its effect on cloud condensation nuclei abundance in the summer Arctic: a case study in the Fram Strait and Barents Sea

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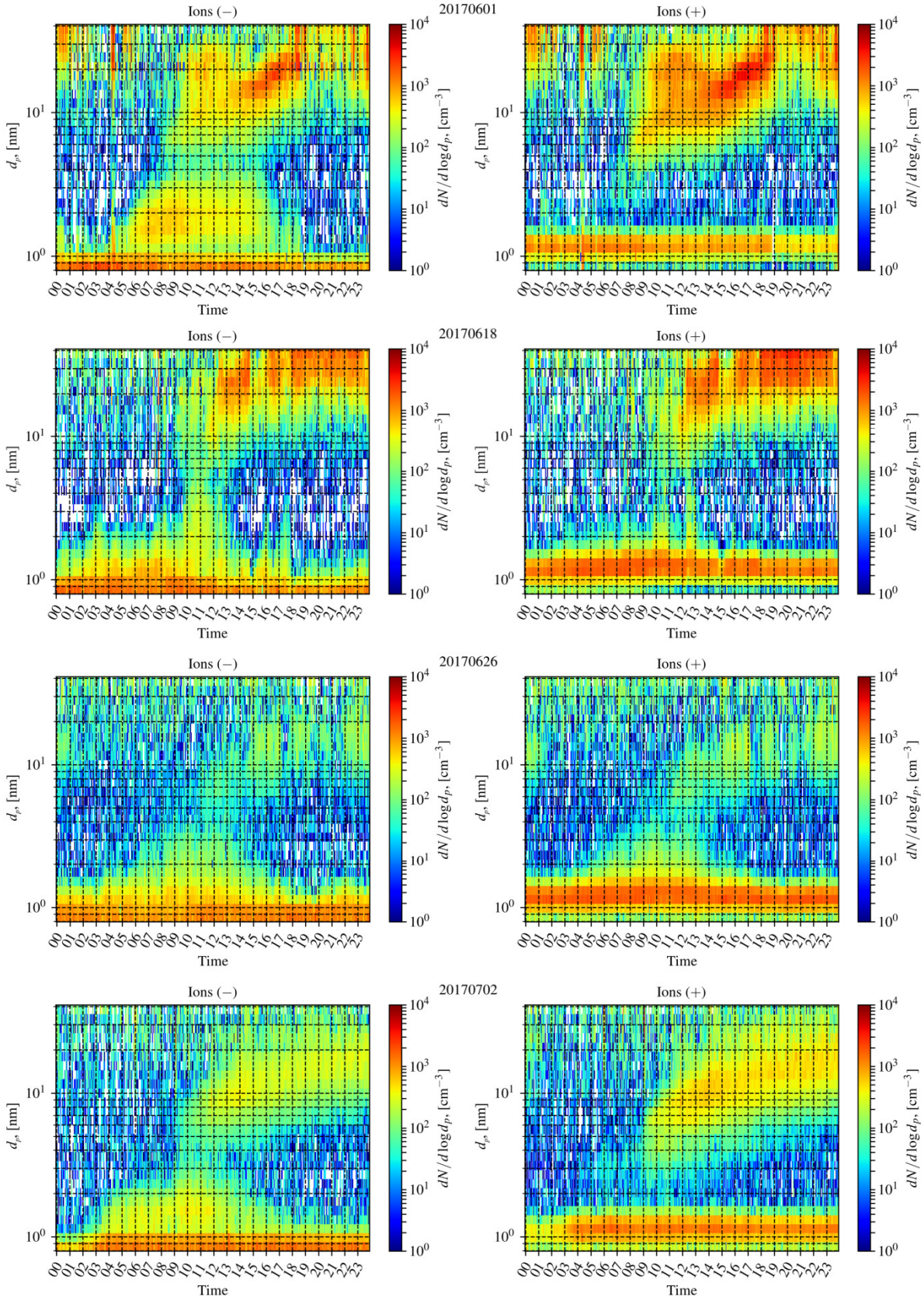
Supplementary material



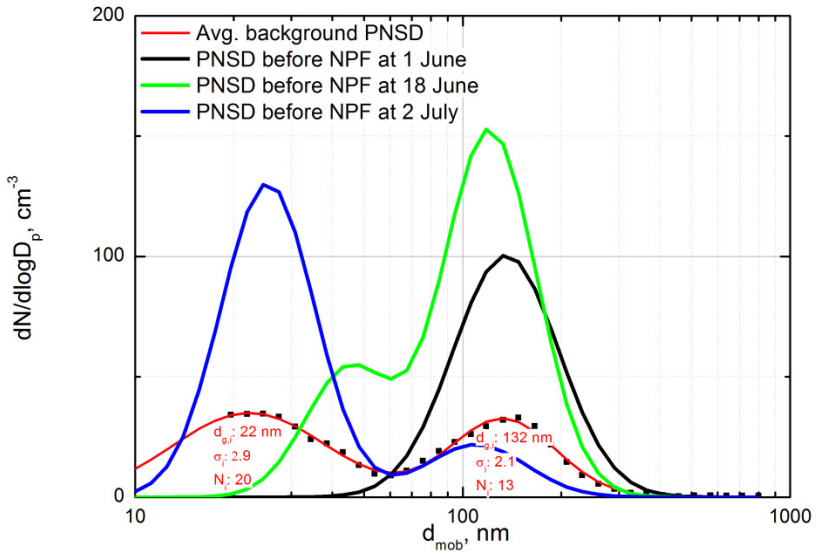
S1. Negative and positive ions during new particle formation events observed during RV Polarstern cruise PS106. Ion number size distributions were measured using Neutral cluster and Air Ion Spectrometer (NAIS). The color scale represents ion number concentration as dN/dlogD_p. The presence of corona charger ions (<2 nm) can also be seen in NAIS data. This artefact was excluded from data analysis.

The CCN number concentration increase calculation.

In the following, we would like to clarify the line of thought when calculating the CCN increase due to NPF:

1. We calculate CCN number concentration resulting from parcel model using input values presented in Table 3. That is, we use those PNSDs, which is a result of new particle formation and subsequent growth. Model then outputs the CCN number concentration resulting from each mode (smaller particle mode at 13 to 44 nm; and larger particle mode at 101 to 194 nm). Total CCN number concentration is then a sum of CCN number from each mode. Please note that we calculate CCN number concentration resulting from 2 modes.
2. We then take total CCN number concentration (resulting from bi-modal size distribution, influenced by NPF) and divide it by the number of CCN resulting only from uni-modal distribution of >100 nm particles (from a different parcel model run, including only accumulation mode particles). We called these (>100 nm) particles as background aerosol, because from our observations it seems that it is always present in the Arctic atmosphere. In figure S2, one can see that average PNSD (PNSDs were taken from the cleanest episodes observed onboard RV Polarstern: 31 May 15:00 to 1 June 06:00; 14 June 15:00 to 15 June 15:00; 1 July 15:00 to 2 July 03:00; 5 July 21:00 to 6 July 12:00; 8 July 00:00 to 8 July 12:00) is indeed composed of two modes with geometric mean diameters at 22 and 132 nm.

   It might sound reasonable to calculate the increase in CCN number concentration with the reference to average background PNSD (which includes two modes). However, if looked at overall campaign PNSDs (not shown in the manuscript; available at request), in many cases the smaller particle mode is a result of new particle formation that happened either in the past days, or different location (and were transported to measurement site). Moreover, during NPF at 1 June, mode at <50 nm was not present at all. Another way would be to use PNSDs recorded just before each NPF event (SP-2). However, in this case, <50 nm particle mode is also frequently a result of the past NPFs. Therefore, it would be not entirely correct to include <50 nm mode if we want to estimate the increase in CCN number concentration due to NPF.



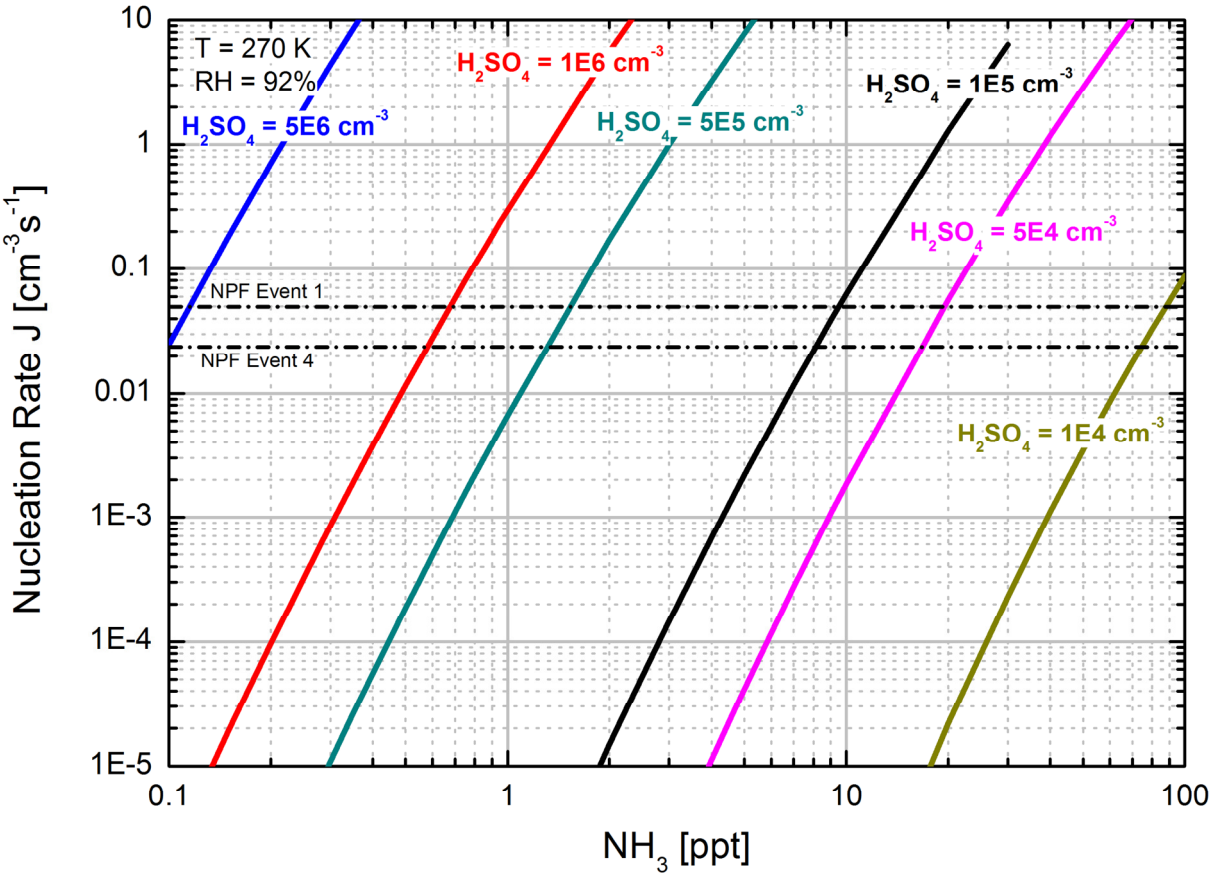
S2. Particle number size distributions (PNSD) measured prior new particle formation and campaign average PNSD (derived from the clean episodes). Please note that the mode at geometric mean diameter > 100 nm is present in all the cases, which cannot be said about ultrafine particle mode (PNSD before NPF at 1 June).

That being said, we have re-analyzed the increase in CCN number concentration in two different ways: 1) using the CCN number concentration resulting from campaign average PNSD (PNSD only from clean episodes); and b) using the CCN number concentration resulting from PNSD measured just before the NPF. Because we did not measure the hygroscopicity parameter kappa of 20 nm particles continuously, we used kappa value of 0.28 (average kappa value of 16-25 nm particles, measured during NPF). For accumulation mode particles, we assumed kappa of 0.33 (campaign average). In case 1, the number concentration of CCN (resulting from campaign average background PNSD) was 17 and 28 cm^{-3}, for updraft wind speeds of 0.1 and 3.2 m s^{-1}, respectively. This gave us the CCN number increase by 2 to 11 fold (versus 2 to 5 if old calculation from manuscript is used) for updraft wind speed of 0.1 m s^{-1}. For updraft wind speed of 3.2 m s^{-1}, the increase in CCN number was even higher – from 8 to 51 fold (versus 4 to 32 if old calculation from manuscript is used)

For case 2, the number concentration of CCN (resulting from PNSD just before NPF) depends on a specific PNSD, which was measured before every NPF event. Let us start with 1 June. The CCN number concentration, resulted from PNSD measured during 1 June (SP-2, black line) was 35 (for 0.1 m s^{-1} updraft wind) and 40 (for 3.2 m s^{-1} updraft wind) cm^{-3}. For lower updraft wind speed, parcel model did not show any CCN being created during NPF event. However, for higher updraft wind, the CCN increase was 29 fold (versus 10 fold if old calculation method is used). For 18 June, the CCN increase was from 2 to 4 fold (same as in our old calculations) and from 6 to 20 fold (versus 6 to 32 fold in our old calculations) for updraft wind speeds of 0.1 and 3.2 m s^{-1}, respectively. And finally for 2 July, the CCN concentration increase was 5 fold in both updraft wind speed cases (same as in our old calculation). Please note that we did not include here calculations for 26 June NPF, because PNSD prior NPF was strongly affected by NPF on 24 and 25 June.

The main conclusions from such an exercise could be as follows:

1. Aerosol particle mode at geometric mean diameter of >100 nm was always present during the measurement campaign.
2. This cannot be said about <50 nm particle mode (see SP-2, PNSD before 1 June NPF).
3. It may seem sensible to use campaign averaged PNSD from clean episodes to define background PNSD, however, from our measurements, we noticed that in most cases the smaller particle mode is a result of NPF (either past days or transported from different location). Thus, it is not very precise to include nucleation mode particles in background aerosol definition when we try to estimate the increase in CCN number due to NPF.
4. Another approach could be using PNSD before NPF event to define background aerosol (same problem as in point 3).
5. We have calculated the increase in CCN concentration using two different PNSDs: for case one, campaign clean episode average PNSD; and PNSD recorded just before NPF event (point 3 and point 4 of these conclusions). We found that in case campaign average PNSD is assumed as background aerosol, the CCN increase is even higher (up to 11 fold (versus 5 fold; for updraft wind speed of 0.1 m s^{-1}). In case the PNSD prior NPF event was assumed as background aerosol, there was almost no difference compared to the results obtained using the methodology presented in the manuscript.
6. In both cases, we were able to show the CCN increase due to NPF.



S3. Nucleation rate as a function of ammonia mixing ratio at T= 270 K and RH= 92% (according to parametrization by Napari et al. (2002). Total sulfuric acid concentration (in molecules per cm^3) is indicated for each curve. Dash-dot lines show the observed formation rate limits.