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Interactive comment on "Summertime observations of ultrafine particles and cloud condensation nuclei from the boundary layer to the free troposphere in the Arctic" *by* Julia Burkart et al.

Anonymous Referee #2

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This paper investigates ultrafine particle (UFP) concentrations in the Canadian Arctic through aircraft measurements. The topic is of interest and important given the rapid changes expected for this region. Overall, I think an attempt at a more quantitative analysis, a more in-depth discussion of the limitations of the approach used in this work, (given that this has been grappled with and addressed in the past literature) and a more complete citation of the existing publications on this topic is needed.

A stated goal is to understand new particle formation (NPF) events (ie, the source of UFP) and how they evolve (grow) to affect radiative forcing through acting as CCN.

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Discussion paper

The paper really mainly focuses on the NPF events. A number or instruments were deployed, most important for addressing this question are measurements of all particle number concentrations larger than 5nm and number size distributions of particles larger than 20 nm. By difference, the authors determine the number between 5 and 20 nm to assess NPF. There is a substantial body of published literature on the the subject of investigating NFP using difference measurements, ie, typically this was done by subtracting data from a UFCPC (eg, TSI 3025) and CPC (eg, TSI 3010 or similar), to obtain concentrations of particle in the 3 to 10 nm range, and the limitations with this type of difference approach. In this study, these limitations are more severe as UFPs are defined over a larger size range, 5 to 20 nm, and smallest size is higher, 5 nm, further away from the size of the newly nucleated particle ($\sim$1 to 1.5 nm). Furthermore, there are no measurements of possible nucleation gas-phase precursors, such as sulfuric acid, (or SO2 and estimated OH could work, but not as well), ammonia and possible low volatile VOCs. Since new stable particles have sizes near 1 or so nm, and in pristine environments particle growth rates can be slow, it can take considerable time for the NPF event to be observable with the the deployed instrumentation. This make interpretation of this data very challenging for investigating nucleation conditions. Thus, essentially, the authors instrumentation is not ideal for addressing the scientific aim of defining NPF conditions in the Arctic.

First, it is recommended that the authors present a thorough assessment of the limitations with their method. This should include: 1) An analysis of the LOD of the difference method used to determine N(5-20) is needed. This will likely show that only significant concentrations of N(5-20) particles can be detected with certainty, potentially leading to a view biased by focusing only on larger nucleation events, which should be discussed. (This should likely include uncertainty in counting statistics, integrated size distributions to get total N of the measured distribution, N20, both instrument flow uncertainties, errors associated with subtracting two large numbers of small difference, etc. 2) The uncertainty in conditions that actually existed at time of the NPF event versus what existed at the time of the measurement due to expected times between the

two (ie, if possible, estimate the delay from nucleation to detection based on estimated or assumed growth rates of nanoparticles).

Without measurements of possible new particle precursor gases and lack of particle concentrations near the critical nucleation diameter it is very difficult to discuss conditions leading to NPF with certainty. For example, it is noted that often an inverse relation is seen between N(5-20) and larger particle concentrations. This could be due to formation of these smaller particles in cleaner air (ie, nucleation precursor vapors can build up due to lack of vapor condensational sink), or the new particles in the cleaner region just have a longer life span compared to particles that may have been formed in the other regions that had higher concentrations of pre-existing particles, which depleted the N(5-20) particles faster. Thus, how can one conclude that the only particle formation events are in the clean regions (possible confirmation bias).

I think the authors have provided evidence that UF particles are generated in the Arctic BL and it can occur under clean conditions, but what happens in more polluted air masses is not really known. Specifically, how (by what process), under what conditions (where), is not definitively known. More analysis, involving calculations (estimations) of lifetimes, more discussion of possible differences in measured total particle surface areas between regions, may allow more definitive conclusions. Some data is clearly not available to make more quantitative assessments, but maybe typical values reported in clean similar environments can set a bound.

Finally, it is not clear what is special about the clouds that leads to NPF in their vicinity. Since this is one major finding of this paper, more details are warranted, including a more comprehensive analysis of the literature, eg; [Clarke et al., 1999a; Clarke et al., 1999b; Hegg, 1991; Hegg et al., 1990; Hoppel et al., 1994; Mauldin et al., 1997; Perry and Hobbs, 1994; Radke and Hobbs, 1991; Raes, 1995; Weber et al., 2001; Wiedensohler et al., 1997]

Specifics.

Lines 73 to 76. These statements are unsupported and possibly not accurate. It all depends on the conditions (thus the statement must be qualified) and also depends on what is defined as ultrafine particles.

Should explicitly calculate estimated losses associated with sampling 5 nm particles (ie, this is the worst case scenario).

For given AMS LODs, what was the AMS sampling rate.

Was N(tot) every defined? Ie, what is the smallest size.

Line 339, what exactly does a burst of N(5-20) mean, was it a burst because the aircraft flow through a small region of high concentrations, or is it being implied that there was a burst in NPF? If the latter, how is that known? In fact, the use of "burst" throughout the paper is confusing since what it really means is isolated regions of high UF particles were encountered, not the typical meaning of a large nucleation event occurring over a short period of time (which I believe is what is implied). It is not clear that the latter can be determined from this sampling approach.

Lines 350... on possible reasons for high fractions of UF particles; this could also include, very slow growth rates, biased aircraft sampling (ie, you went looking for them).

Lines 462-465. This sentence seems to exactly confirm the limitations with the analysis used in the paper; the presence of UFP may have mostly to do with residence time in the BL and not mechanism that formed them.

Line 497-498; This conclusion is not the only possible one, it is also possible that they just are not scavenged and so have a longer life time (as noted already). Without measuring the nucleation precursor species, it is hard to tell. Calculations of life times of particles of 5 to 20nm due to coagulation, based on given size distributions may be insightful. Growth out of the 5 to 20 nm size range due to condensation is likely more important. Maybe if some assumptions are made on typical concentrations of H2SO4 or low volatile VOCS one could also estimate this, or use reported growth rates in other

clean regions?

Lines 573-575 and 594-595. Not clear what the all the ideal conditions for NPF were, is it just high OA levels? More details would be good, see references (eg, precipitating clouds scavenge accumulation mode aerosols, reducing surface area, but nucleation precursor gases can pass through (eg, SO2) forming H2SO4 in the high OH (high actinic flux and RH) in the cloud outflow, plus high RH leads to NPF. . ..

Lines 647-648: I think this sentence should be clarified. It cannot be said that the open water is the source of the observed particles, it maybe true, but it could also be that stable new particles of sub 5 nm size were formed elsewhere and just grew to your detectable size due to ocean emissions.

References:

Clarke, A. D., V. N. Kapustin, F. L. Eisele, R. J. Weber, and P. H. McMurry (1999a), Particle production near marine clouds: sulfuric acid and predictions from classical binary nucleation, Geophys. Res. Lett., 26, 2425-2428.

Clarke, A. D., F. Eisele, V. N. Kapustin, K. Moore, D. Tanner, L. Mauldin, M. Litchy, B. Lienert, M. A. Carroll, and G. Albercook (1999b), Nucleation in the equatorial free troposphere: Favorable environments during PEM-Tropics, J. Geophys. Res., 104, 5735-5744.

Hegg, D. A. (1991), Particle production in clouds, Geophys. Res. Let., 18, 995-998.

Hegg, D. A., L. F. Radke, and P. V. Hobbs (1990), Particle production associated with marine clouds, J. Geophys. Res., 95, 13917-13926.

Hoppel, W. A., G. M. Frick, J. W. Fitzgerald, and R. E. Larson (1994), Marine boundary layer measurements of new particle formation and the effects nonprecipitating clouds have on aerosol size distributions, J. Geophys. Res., 99, 14,443-414,459.

Mauldin, R. L., S. Madronich, S. J. Flocke, F. L. Eisele, G. J. Frost, and A. S. H. Prevot

(1997), New insights on OH: Measurements around and in clouds, Geophys. Res. Lett., 24(No 23), 3033-3036.

Perry, K. D., and P. V. Hobbs (1994), Further evidence for particle nucleation in clear air adjacent to marine cumulus clouds, J. Geophys. Res., 99, 22803-22818.

Radke, L. F., and P. V. Hobbs (1991), Humidity and particle fields around some small cumulus clouds, J. Atmos. Sci., 48, 1190-1193.

Raes, F. (1995), Entrainment of free tropospheric aerosols as a regulating mechanism for cloud condensation nuclei in the remote marine boundary layer, J. Geophys. Res., 100, 2893-2903.

Weber, R. J., G. Chen, D. D. Davis, R. L. Mauldin, D. J. Tanner, F. L. Eisele, A. D. Clarke, D. C. Thornton, and A. R. Bandy (2001), Measurements of enhanced H2SO4 and 3-4 nm particles near a frontal cloud during ACE 1, J. Geophys. Res., 106, 24,107-124,117.

Wiedensohler, A., H.-C. Hansson, D. Orsini, M. Wendisch, F. Wagner, K. N. Bower, T. W. Chourlarton, M. Wells, M. Parkin, K. Acker, W. Wieprecht, M. C. Facchini, J. A. Lind, S. Fuzzi, B. G. Arends, and M. Kulmala (1997), Night-time formation and occurrence of new particles associated with orographic clouds, Atmos. Environ., 31, 2545-2559.

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