

Review: "Is the summer aerosol over the Arctic controlled by regional atmospheric circulation or ice conditions? Trends and Future Implications"  
Leck et al.

This paper combines in situ shipboard measurements of high Arctic aerosol size distributions spanning the years 1991-2018 with reanalysis and back-trajectories to relate summer synoptic process and sea ice conditions on changes to the size distributions. The authors find, using Self Organizing Maps, that three synoptic regimes were most common for August-September and likely have the greatest effect on the measured distributions. Namely, the largest differences were between freeze-up and melting conditions, such that new particle formation from frost flowers were likely attributable to size distribution changes. Using the observed regime shift in open water fraction and temperature during the record, the authors posit that future conditions will become more favorable for new particle formation in the high Arctic, however, diverse impacts from different biogenic aerosol sources at the air-sea interface due to these changes confounds predicting exact changes.

Overall, this paper provides interesting insights on environmental and meteorological conditions that likely impact size distributions and the implications for these effects in the future, however several areas of the discussion lack clarity, depth, and support from the available measurements and the analysis conducted. The authors should make a concerted effort to carefully revise and edit the paper for clarity and continuity between passages and sections. I believe this paper is suitable for this journal, but it is necessary to consider and address the following, major comments and technical concerns before being acceptable for publication.

#### Major Comments:

- There are several passages in this paper that speak at depth to the biological and physical characteristics at the ocean surface and its composition, but many of these are ancillary to the key points made in the paper and their relation to the overall findings. In the proceeding major/technical comments I identify where some of these sections are and how they lead to a digression in the focus and should thus be reduced or revised to more directly emphasize their relevance.
- Introduction: I believe this section should be significantly revised. The first couple of paragraphs in the introduction can be reduced for brevity. Too much superfluous detail is included here that does not exactly connect to the main motivation or research questions that are meant to be answered in the paper. It would be much more appropriate for the authors to include discussion on relevant Arctic topics including air mass characterizations, the seasonality of aerosol and their sources, and potential impacts of synoptic circulation and regional processes. Less attention should be focused on jet/film/gels etc. It is necessary that the authors provide support and context that justifies the paper's motivation. What are the current limitations and uncertainties and research questions that motivate carrying out this work?
- SOM Clustering: How can the authors justify that the use of only August-September in the SOM sufficiently captures synoptic variability in the Arctic? I believe limiting the

scope to these months will characterize the synoptic circulation most relevant and common to those months, but likely not the extensive annual and interannual variability throughout the season that could occur. Was the full annual cycle tested for comparison? I feel that completing such an analysis will justify this choice and make the results more robust.

- More detail should be provided on the lognormal fitting of the aerosol size distributions. Was this an automated process? How were the number of modes determined and the robustness of fit evaluated? The authors should provide statistics on the mode fits. What was done for periods in which an extension of the size range with the spectrometer wasn't included and how can those periods be justifiably compared to when only the DMA was available? All of these points must be discussed because the results of fitting of the lognormal modes is not discussed anywhere in the paper.

#### Minor Comments and Technical Corrections:

- Line 88: The more canonical term is “aerosol-cloud interactions”, rather than “aerosol-to-cloud interactions”. I suggest this be revised.
- Line 88-89: The authors should clarify how aerosol-cloud interactions counteract warming in the Arctic as relevant to this claim and citation.
- Line 95: Revise “most significant” to “largest”.
- Lines 130-131: It should be specified here that the August-September input data were used because they coincided with when shipboard measurements were most often collected.
- Lines 140-141: Can the authors please be more specific when they say, “...sufficiently represent the range of large-scale circulation patterns in the Arctic.”? There is no context for Arctic circulation patterns (in the introduction or elsewhere) prior to this description and no citation.
- Line 144-149: The citations that describe the ERA5 reanalysis and the description of its data and assimilation should be moved to the first mention of the product in Lines 125-126. In its current location it is a digression.
- Lines 150-152: Did the authors intend to say that means (averages) of the variables listed in these lines “were separately calculated” for each MSLP regime? If that is the case, I believe that this sentence can be rewritten for clarity to say that means of variables including [...] were calculated for each MSLP regime.
- Line 156: Please clarify why the latitude band 78N-82N was used separately to 85N. What distinguishes these regions?

- Lines 163-165: The authors should specify what kind of differential mobility analyzers (and CPCs) were used to measure the aerosol size distributions. What were the size ranges of the DMAs?
- Line 166: What type of “contamination”? Contamination from the ship? Please specify and provide more detail (at least as a brief summary) of how and why this was done.
- Line 168: The authors should specify what kind of aerosol spectrometer was used and what was its size range.
- Line 169-171: For the “harmonized” size distribution interpolation, was any merging done? If so, please describe how and if there was diameter overlap between the different measurements. Please include detail on how considerations were made to combine distributions from mobility and/or optical diameters. An assessment of the merging should also be detailed.
- Line 189: What is meant by the sentence “Vertical layers one through 25 hPa separates five layers”?
- Figure 6: The size distributions in these plots show values extending down to 1 nm, while it was described in the methods that the distributions were interpolated to a range of 3.37-900 nm. Can the authors please clarify this discrepancy?
- Line 301-302: A citation or evidence should be provided here to justify claiming the source of the high nucleation mode as new particle formation in this region. The authors should also clarify what is meant by “new particle formation”. I believe the authors mean the formation of particles from gaseous precursors. If so, this should be specified.
- Line 302: revise “dealt with” to “discussed”.
- Figure 6: What is meant by the y-label in this figure? Are these size distributions showing the number concentration in each bin not the normalized number concentration per  $d\log D_p$  bin? If it is the former, why is that presentation of the distribution shown rather than the  $dN/d\log D_p$  (the concentration normalized by the differential log diameter which is more commonly used)?
- Section 4: Can the authors provide some quantitative statistical information on the modal properties of the size distributions between nodes and melting/freeze-up? Specifically, the authors should quantify the mean diameters and number concentrations of the modes between nodes and freeze-up and melt. How do these properties compare to previous literature if available for synoptic regime differences in the size distribution? What do these results suggest for changes due to aerosol sources, synoptic circulation, and sea ice conditions?

- Line 323: Clarify what is meant by “outlier size distributions of node 2 in Fig. 6b.” I believe the authors are indicating the high concentration nucleation mode remaining in the melt group.
- Lines 325-326: Specify that the description “Figure 8c showed an average...” is pertaining to the average of nodes 1,3,4,5, and 6.
- Line 326: Revise to “the [sea] ice”.
- Lines 372-373: Please clarify what are the “segment ratios”.
- Line 379-380: The final sentence in this passage is very confusing and vague. Please clarify/revise or remove.
- Lines 410-411: “systematic trends”, what is meant by this? Were the trends found to be statistically significant?
- Lines 435-437: In these lines, the differences in the size distributions should be further quantified and compared using statistical information from the modes.
- Lines 437-439: Can the authors not attribute any of the differences in the size distributions between periods to transport? No evidence in this paper is given to justify claiming these differences are solely driven by new particle formation. A further consideration of these differences needs should be done here.
- Line 451-470: I believe this entire passage should be significantly reduced to remain within context and on topic of the discussion in this section. As it stands, this passage is a substantial digression. Key findings from the works and discussion provided here would be more useful to put into context for the proceeding paragraphs.
- Lines 471-502: Like my previous comment, there appears to be more superfluous content here that digresses from the main point of this section. The authors should make a more concerted effort to relate brief key findings from the many cited works in these passages within the context of their effect on aerosol size distribution changes in the long-term in freeze-up and melt conditions. Only a brief discussion from Lines 498-502 does this, but it is inadequate and should thus be expanded. One point of expansion could be discussing the size-dependent effect of these changes. For example, the authors discuss effects on sea spray sources and emission and cloud processing, which will likely have dissimilar effects to biogenic impact on sub-Aitken-mode particles and other modes.
- Lines 527-528: I don’t think “aerosol-cloud-climate connections would *[require]* model simulations”. Aircraft, ground-based, and remotely sensed cloud microphysical properties can be retrieved for this purpose by relating them to the observed aerosol measurements of this study and has been done in previous work that should be cited. This should also be mentioned in this concluding discussion.