Response to Review

Frequent Ultrafine Particle Formation and Growth in Canadian Arctic Marine and Coastal Environments

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Referee comments are reproduced in blue. Author responses are provided in black.

Anonymous Referee #1:

This manuscript summarizes measurements conducted in 2014 and 2016 in the Canadian Arctic aboard the CCGS Amundsen. Ultrafine particle formation and growth were frequently encountered especially in 2016. In addition to summarizing the particle events the authors look at various meteorological and oceanographic data that might explain the differences in particle events between 2014 and 2016. Unfortunately the differences in atmospheric and oceanic conditions were small and there was no smoking gun that clearly explains the differences in particle events between the two years. The manuscript is well written and easy to read. I feel the manuscript is appropriate for ACP and should be published with minor revisions.

The authors appreciate the assessment and accurate summary of the main points of the manuscript.

1. I think it should be clearly pointed out that these are coastal measurements and not the open Arctic Ocean. I think "Coastal" should be added to the title before Canadian.

The authors agree with the reviewer's point that the environment sampled aboard CCGS *Amundsen* during both expeditions is often proximal to a coastline. While air mass histories that consider the 0-10 km column height show appreciable residence over the central Arctic Ocean, the spatial component of near surface air mass history is restricted to a small region (c.f. Item 3 from Referee #1) which is often overlapping a coastline. Indeed, coastal impacts are of possible importance to ultrafine particle (UFP) formation and growth phenomena in this region, as discussed in this manuscript (e.g., p10 starting at line 24) and the relevant references cited therein. The authors feel that the study is both coastal and marine, so both terms now appear in the revised title:

"Frequent Ultrafine Particle Formation and Growth in Canadian Arctic Marine and Coastal Environments"

2. Page 10, line 15. What VOCs?

The reference to Mungall et al. (2017) is directed at recent measurements of oxygenated volatile organic compounds (VOCs) conducted on the CCGS *Amundsen* expedition in 2014 using acetate chemical ionization mass spectrometry. Factor analysis of the data revealed a unique contribution of an ocean source

to formic acid, isocyanic acid, and organic oxo-acids of varying carbon number. A larger array of compounds may have been present and co-emitted with compounds attributed to the 'Ocean Factor' in that study but do not have a greater proton affinity than acetate, and so cannot be measured by that technique. For this reason, it is prudent to retain a general characterization of the possible compounds, as little is known about the profile of VOCs emitted from the ocean and their dependence on biological and/or chemical interactions at the air-sea interface.

The manuscript now specifies the VOCs as oxygenated: (p10, line 14-15) "…and evidence of an ocean source for oxygenated volatile organic compounds…".

3. Figure 5, e-h. The Flexpart PES for 0-200m are very hard to read. Can these figures be expanded to better show the data? This really points to the local source of the particles.

The referee's comment reflects a conceptual decision on how the figure was designed. The 8-panel figure was intended to illustrate the history of relevant air masses over the Arctic Ocean in addition to the finding that the relevant air masses were in contact with the boundary layer/Earth's surface only within a limited spatial extent around the model 'release' point (ship location). The referee's recommendation to expand the spatial scale of Figure 5(e-h) has been incorporated in the revised manuscript with a specific note added to the caption highlighting the vastly different spatial scales used in the top versus the bottom row of panels. In order to achieve a readable map with a finer spatial scale, a slightly different projection was used for the new maps.

4. Page 17 line 15. Saying these factors "led to a greater frequency of UFP formation" is really speculation. Perhaps "could have individually or collectively contributed to a greater frequency..."

The measured language suggested by the referee has been adopted in the revised manuscript. The excerpt on p17 line 15-16 is now: "…and the differences in the biological activity in the local marine environment may have individually or collectively contributed to a greater frequency in UFP formation and growth in 2016." Indeed this study was not able to unequivocally identify a driving force for UFP formation and growth, but we hope that future chemical transport model studies that are constrained by the data reported here may help elucidate factors that lead to the observations.

References

Mungall, E. L., Abbatt, J. P. D., Wentzell, J. J. B., Lee, A. K. Y., Thomas, J. L., Blais, M., Gosselin, M., Miller, L. A., Papakyriakou, T., Willis, M. D. and Liggio, J.: A novel source of oxygenated volatile organic compounds in the summertime marine Arctic boundary layer, Proc. Natl. Acad. Sci. U.S.A., 114(24), 6203–6208, doi:10.1073/pnas.1620571114, 2017.