Ice nucleating particles in Canadian Arctic sea-surface microlayer and bulk seawater.

Victoria E. Irish, Sarah J. Hanna, Megan D. Willis, Swarup China, Jennie L. Thomas, Jeremy J. B. Wentzell, Ana Cirisan, Meng Si, W. Richard Leaitch, Jennifer G. Murphy, Jonathan P. D. Abbatt, Alexander Laskin, Eric Girard, Allan K. Bertram

https://doi.org/10.5194/acp-2018-735

Summary

This study researched the quantity, chemistry, and potential sources of immersion mode ice nucleating particles in the Canadian Arctic marine boundary layer during the summer of 2014. Aerosol samples were collected at 28 locations on a MOSSI impactor and then analyzed using a droplet freezing technique to quantify the concentration of INPs as a function of temperature. The ratio of mineral dust to sea spray particle surface area was quantified using EDX for three samples. These surface area ratios were then converted to active site density ratios, which revealed that mineral dust was the dominant INP type in the considered samples. Finally, the authors use the FLEXPART particle dispersion model to correlate INP concentration data with air mass back trajectories. This analysis suggests the source of INPs in the artic marine boundary layer are relatively local mineral or soil dusts.

General Comments

This study makes a substantial contribution to the field by presenting an impressive amount of immersion mode INP concentration data for the Arctic. The locality of the field work is very timely, and the additional analyses on particle composition and back trajectories nicely support the authors' main goal of quantifying immersion mode INP concentrations. The paper is well suited for publication in ACP. Below, I provide a few questions and comments to strengthen the paper and clarify its results.

Note on INP Nomenclature:

Please try to be clearer throughout the text that you considered the immersion freezing mode of ice nucleation. This is especially important given your results that mineral dust was an important source of particulate matter in your samples: mineral dust can activate in the deposition freezing mode at the temperatures considered here (e.g. Hoose and Mohler, 2012), but the mode of freezing investigated in this study isn't even included in the abstract.

Specific Comments

1. **Page 2, Line 9:** Please clarify here that sea spray particles actually vary widely in composition, depending on mechanism of formation. For instance, film rupture particles are organic enriched compared to jet droplet particles (e.g. Wang et al. 2017). This will highlight the importance of considering both microlayer and bulk seawater samples, as you suggest in the following sentence.
2. **Page 2, Line 11:** For completeness, the authors may also consider citing Huang et al. 2018 here. This paper follows up on Burrows et al. 2013 by discussing the sensitivity of the relative contribution of sea spray to INP concentrations at high latitudes to a model's choice of active site density parameterizations.

3. **Page 3, Line 15:** Were the metrological data collected with on-ship sensors? If so, can some estimate of the height of these sensors above sea level be given?
4. **Page 4, Line 8:** Has the transmission efficiency of the louvered total suspended particulate (TSP) inlet been quantified before? For example, it sounds similar to an inlet characterized by Kenny et al. 2005. If the exact loss percentages have not been measured, that's okay; but the authors should note that larger particles (like mineral dust) may be more prone to impaction and thus be undersampled on the MOSSI slides.
5. **Page 6, Line 7:** Was the height of the chemical ionization mass spectrometer inlet at a similar height to the inlet of the MOSSI?
6. **Page 7, Line 17:** The authors state that only particles at the edge of the spots were analyzed with EDX, but how are they sure these particles are representative of the bulk of particles directly below the micro-orifices? For example, the authors mention the particle rebound effect (Page 4, Line 12). Can the authors discuss if perhaps mineral dust is more prone to rebound than the deliquesced salty particles, and therefore may be more likely to end up at the edge of the spot where EDX was performed?
7. **Page 7, Line 22:** "First, the atomic percentages of each particle were determined from EDX spectra." Can the authors clarify if only one discrete spot of each particle was examined, or if the EDX data represents an average signal from a raster scan?
8. **Page 7, Line 23:** Do the authors have any way to estimate what fraction of particles were internally mixed; i.e. mineral dust coated with sea salt?
9. **Page 8, Line 4:** How were blanks prepared and treated? Were they fresh slides out of the package? Or were they treated the same way as the sampled slides, i.e. placed within the MOSSI only without turning on the pump? The latter would better account for contamination in the sample handling and preparation process; but in either case, more information is needed here.
10. **Page 8, Line 17:** "The two samples corresponding to high [INP(T)] were collected on July 21st and 25th." As I read Figure 2, these are among the highest but not the highest INP concentration days. Can the authors provide any more reasoning behind why these particular days were chosen for compositional analysis? How typical was the meteorology on these days? Or perhaps they were chosen to be evenly spaced in time and location? Or was the choice random?
11. **Page 8, Line 14:** The jump to biological particles is a bit of a non-sequitur. You might elaborate on how biological activity can influence sea spray particle composition, which in turn effects INP activity.
12. **Page 8, Conclusions:** Can the authors discuss their findings that mineral dust immersion INPs dominated over sea spray INPs in the context of Irish et al. 2017, which found immersion mode INPs to be abundant in the seawater from this region? E.g. perhaps the INPs were present in the seawater but were never aerosolized because it wasn't windy enough to generated sea spray. This also ties in with Reviewer 1's comment that time spent over open ocean may not correlate with sea spray INP concentration if sea spray particles are not being formed. Then, include a discussion on whether these conditions are typical: how do your wind speeds compare with average (intra- and interannual) wind speeds in the region? Briefly delving into a reanalysis dataset (e.g. ECMWF ERA-Interim) might help you to explore this question in depth, or you could look for historical observational data. This will require a little bit more work on your end, but it would strengthen your analysis immensely by allowing you to hypothesize whether your findings are typical.

13. **Page 20, Table 1:** Please include in the table caption the length of the back-trajectory (7 days).
14. **Page 21, Figure 1:** Can you also denote in this figure where the samples for chemical/EDX analysis were collected? That will help us visualize the potential sources of these particles in light of the data in Figure 5. Otherwise, this figure looks great!
15. **Page 23, Figure 3:** Please also show the min/max INP concentration at each temperature (in addition to the average and standard deviation you've already shown) to provide more information on the degree of variability. Are the data from DeMott et al. 2015 averages, or discrete measurements?
16. **Page 25, Figure 5:** Please increase the font size in this figure to something legible.
17. **Supplement, Page 9, Figure S6:** Please include units (L^{-1}) for INP concentrations either in the axis label or the figure caption.

References:

Hoose, C. and Möhler, O., 2012. Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, Atmospheric Chemistry and Physics, 12, 9817–9854.

Huang, W.T.K., Ickes, L., Tegen, I., Rinaldi, M., Ceburnis, D. and Lohmann, U., 2018. Global relevance of marine organic aerosol as ice nucleating particles. *Atmospheric Chemistry & Physics*, *18*(15).

Kenny, L., Beaumont, G., Gudmundsson, A., Thorpe, A. and Koch, W., 2005. Aspiration and sampling efficiencies of the TSP and louvered particulate matter inlets. *Journal of Environmental Monitoring*, *7*(5), pp.481-487.

Wang, X., Deane, G.B., Moore, K.A., Ryder, O.S., Stokes, M.D., Beall, C.M., Collins, D.B., Santander, M.V., Burrows, S.M., Sultana, C.M. and Prather, K.A., 2017. The role of jet and film drops in controlling the mixing state of submicron sea spray aerosol particles. *Proceedings of the National Academy of Sciences*, *114*(27), pp.6978-6983.