Title: *Ice nucleating particles in Canadian Arctic sea-surface microlayer and bulk seawater.*
Authors: Irish *et al.* 2017
DOI: 10.5194/acp-2017-384
Journal: Atmospheric Chemistry and Physics (ACP)

Summary

This study evaluates the abundance and characteristics of ice nucleating particles (INPs) in Canadian Arctic waters during the summer of 2014. Nucleation in the immersion freezing mode was quantified for microlayer and bulk (subsurface) seawater samples. Analysis of samples from eight process stations reveal that both bulk and microlayer samples contained elevated INP concentrations compared to an ultrapure water control, as well as station blanks (sample passed through a 0.02 μm filter). Contrary to previous experiments (Wilson *et al.* 2015), the results do not indicate an enrichment of INPs in the microlayer relative to the bulk samples. The concentration of INPs varies considerably between the eight geographically diverse process stations. The authors correlate INP concentrations with an array of 6 other variables (DMS concentration, bacterial and phytoplankton cell counts, temperature, pH, and salinity), finding that salinity provided the strongest and only statistically significant relationship with INP concentration. Finally, filtration and heating experiments suggest such INPs were between 0.02 μm and 0.2 μm in diameter and thermolabile, suggesting INPs were organic in composition and may have consisted of femtoplankton, cell fragments, or cell exudates/lysates.

General Comments

In all, this paper makes an important contribution to the field by helping to quantify the range of variability in INP concentration sourced from Arctic marine waters. The authors' effort to compare their results with other recent measurements is particularly laudable, and highlights the need for subsequent studies that contrast how oceanic variables, laboratory protocol, and sampling techniques ultimately affect measured INP concentrations. As such, this paper sets the stage nicely for further developments in the field, and therefore is well qualified for publication within ACP. Below, I offer a few questions and comments to strengthen the paper and clarify the results:

Scientific Comments

1. Page 3, Lines 8 – 9: The authors report that the Niskin bottles were washed with large amounts of seawater before sampling at 0.5 meters below the surface commenced. At the same time, the results suggest that the microlayer samples did exhibit significant INP enrichment relative to the bulk seawater, in contrast to Wilson *et al.* 2015. Were the Niskin bottles rinsed on-site in the zodiac? If so, is it possible that the organic-enriched microlayer was disturbed in the rinsing process, mixing INPs to subsurface waters and muddling the distinction between the two samples that would ordinarily exist?

2. Page 3, Lines 33 – 35: Heating seawater to high temperatures (100 ˚C) and gradual cooling can cause salts, especially carbonates, to precipitate from solution (e.g. Anderson, 2005; Jones 1967; Harrison et al. 1980). Were precipitates observed during the

thermodegradation heating tests? If so, have the calculations for Corrections for Freezing Temperature Depression (Section 2.4.4) taken this change in salinity/alkalinity into account? If precipitates did form and were unnoticed, then the corrected temperatures reported in Figure 4 may in fact be lower limits.

3. Page 5, Lines 15 – 17: A strong anticorrelation between salinity and ice nucleation efficacy was observed across the 8 process stations' samples. Studies have found that ice rafting and melting spurs cell growth (e.g. iron fertilization in the Southern Ocean (Duprat et al. 2016) and possibly phosphorus addition in the Arctic (Perrette et al. 2011)). Although there was only a moderate correlation between cell count and T10 value (or better in the microlayer – Table S2), was there a correlation between salinity and cell count? This would suggest that nutrient addition from melt water might be spurring cell growth and possibly INP production, with interesting implications for future Arctic and Greenland ice loss.

Clarification and Technical Comments

1. Page 1, Line 22 – 23: "INPs were ubiquitous in the microlayer and bulk seawater with freezing temperatures *in the immersion mode* as high as -14 °C," or something similar to indicate mode of activation.

2. Page 2, Line 10 – 11: "Modeling studies have also suggested that marine INPs may offset the magnitude of anthropogenic aerosol forcing by influencing cloud formation (Yun and Penner, 2013)." This is vaguely worded. How specifically do marine INPs reduce the negative anthropogenic forcing?

3. Page 2, Line 23 – 25: The original (Vali 1971) notation makes it more clear that INP concentration is a function of temperature: use [INP(T)] instead of [INP]. This notation more clearly denotes that INP activation is temperature-dependent.

4. Page 16, Figure 2: What are the temperature uncertainties on a typical data point?

5. Page 18, Figure 4: Since the data from Schnell (1975) and Schnell and Vali (1975) are so sparse, they would be easier to see if they were plotted over the other data.

6. Page 18, Figure 4: How are the reported uncertainties in [INP(T)] calculated?

6. Supplement Pages 4-6, Figures S1–S3: It would be helpful if the x and y axes sizes and marks (latitude and longitude labels) were consistent between the sample-site plots and the chlorophyll a plots. That way, chlorophyll concentrations at sampling sites could more easily be determined.

Works Cited

Anderson, R.A. (ed.), Algal Culturing Techniques, Elsevier Academic Press, Burlington, Massachusetts, 578 pp, 2005.

Duprat, L.P., Bigg, G.R. and Wilton, D.J., Enhanced Southern Ocean marine productivity due to fertilization by giant icebergs, Nature Geoscience, 2016.

Harrison, P.J., Waters, R.E. and Taylor, F.J.R., A broad spectrum artificial sea water medium for coastal and open ocean phytoplankton. Journal of Phycology, 16(1), pp.28-35, 1980.

Jones, G.: Precipitates from Autoclaved Seawater, Limnology and Oceanography, 12(1), 165–167, 1967.
Perrette, M., Yool, A., Quartly, G.D. and Popova, E.E., Near-ubiquity of ice-edge blooms in the Arctic, Biogeosciences, 8(2), p.515, 2011.