Review for “Revisiting properties and concentrations of ice nucleating particles in the sea surface microlayer and bulk seawater in the Canadian Arctic during summer” by Victoria E. Irish et al.

Anonymous Referee

The revised submission of ”Revisiting properties and concentrations of ice nucleating particles in the sea surface microlayer and bulk seawater in the Canadian Arctic during summer” by Irish et al. has improved compared to the initial submission, but this is very little to bring it up to the standards of the journal Atmospheric Chemistry and Physics. My previous comments in summary, were that there was insufficient scientific advancement in physics or chemistry to warrant publication. In this submission there are two new extensions beyond what was published in Irish *et al.*[1] which are i) oxygen isotopic fractionation and ii) a quantitative analysis of ice nucleating particles in air from a marine source. The isotope measurements adds to the study that terrestrial runoff and precipitation are correlated with the freezing temperature at which 10% of droplets froze, T_{10}. This correlation was better than for melting sea ice or seawater. To calculate the concentration of ice nucleating particles (INPs) in air, mass concentrations of sodium in ambient aerosol were used to scale their results. In total, the new findings compared to Irish *et al.*[1] were that INP concentrations were higher in 2016 than 2014 which are likely due to volume sampling differences, a correlation between calculated meteoric and sea ice melt water fractions and T_{10}, and back of the envelope calculations for INP concentrations in air. These extensions unfortunately do not apply any theory or give fundamental understanding in physics and chemistry and so I cannot recommend publication in the journal Atmospheric Chemistry and Physics which stresses exactly this. I warn the authors that if the editor allows for resubmission, much more work must be done in this regard to significantly shorten discussions that are already made in Irish et al. while emphasizing any new discussion. Calculating correlations and up scaling data to the atmosphere is good but not sufficient for greater physical and chemical understanding.

Certainly, the measurements done on board a research vessel are very difficult, and there are now small extensions beyond the previous submission. These should be published, but I recommend elsewhere in literature. I would concede if the authors were restricted in time and submitted some months after the research cruise was over, then the benefit of the doubt would be given to publish exciting results as soon as possible. Was there some limitation in time or some issue with the data or paper that I should be unaware of when re-evaluating this manuscript?

Major Comments

There remains an absence of testing any theory. This includes any chemistry, physics or thermodynamics. Free energy calculation for ice nucleation or critical ice embryo size is not calculated. Nucleation theories are not applied or tested. There is no evaluation on the transfer of particles from the bulk to the microlayer or into the air that uses physics or chemical transformation. Measurement of biological tracers are done, but only correlation is made without any other hypothesis testing.

The authors did not need to make more clear that they observed enhanced INP numbers in microlayer layer more in 2016 than in 2014 on l. 27-29. They needed to explain and give a physical-chemical reason as to why. Instead they only claim that ocean variability was the cause, or more likely than not it was an artifact of sampling a factor of 3 less in layer thickness 2016. This means that the microlayer concentrations in 2014 were simply diluted. It is true that the authors data make a comparison quantifying how the properties and concentrations of INPs have remained the same or have varied between these years, however, it does not answer the question of why. In general, the authors have not extended their manuscript enough and should choose a different journal that stresses measurements and data more.

The authors state that much of their results and data are consistent with Irish *et al.*[1]. I had previously made the comment that the manuscript was too similar to their previous work, being about 30% identical to Irish *et al.*[1] and other material they published based on the iThenticate.com Similarity Report. Although the addition of oxygen stable isotopes and calculation of airborne INPs will make this less similar, not enough was done to reword the rest of the manuscript. Therefore, my previous major comment that this manuscript it too similar to their previous is still warranted.

Minor Comments

- p.1, l.17 - The word choice is too negative. The way it was in the first version using the word "limited" better states that good work has been done and there is a need for more.
- p.4, l.14-15 - The freezing temperature is not determined visually. The freezing is determined

visualy and the temperature is measured by an instrument at the same time it freezed. Please reword this sentence.

- Please indicate in one sentence or so in section 2.2.1 how temperature was calibrated.

- There is a section 2.1.1 but no section 2.1.2. There is no need to separate here. Please have only section 2.1.

- Description of blanks for the lab and field for different filtering are in different places, p.12 l.30 - p.13 l.2, p.13 l.14 - 16, p.17 l.3-7. Field blanks are discussed many times but found it hard when reading through the paper, where to locate their description. I recommend the authors dedicate a new short section to describe all the blanks one after another. This will help the reader refer back to the definition of the blanks.

- Another point about the field blanks. I understand that when seawater is filtered, freezing temperatures are much lower than field blanks. The procedure to make a field blank is first, to rinse all glassware and tubing for some time then second, sample and freeze drops of pure water that rinsed and flushed all glassware and tubing after the first rinse. Therefore, is it safe to say the purpose for field blanks is to evaluate the ability to reuse the same glass plate sampler and tubing to not cross contaminate between different stations? I think this is the case. It should be directly stated in the manuscript.

- The short sentence on p.6 l.18 should be removed as it is a repeat of the previous.

- The phrase *in situ* was not used in the previous manuscript, but it is used in the revised version. However, an *in situ* chlorophyll measurement was not performed because the authors did not measure in water that remained in the ocean. Water was removed from the ocean. Samples of water were used for chlorophyll concentrations measurements. Please correct this.

- p.9 l.29 - The correlation coefficient of -0.83 and p value of 0.001 is exactly the same for both T_{10} and T_{50} in Tables 2 and S2. Is the a typo or coincidence?

- p.10 l.4-8 - Deviation in freezing temperatures from those of constant Δa_{w} was observed only for ammonium containing solutes[2]. Ammonia concentration in seawater should be on the order of micromolar and therefore should not affect freezing temperature in this way. This authors may wish to include this.

- p.10 l.12-14 Terrestrial runoff can also contain nutrients to grow marine microorganisms. After these nutrients are used up, cells can lyse, sink or their exudate can remain in surface waters. Then the source of INP may still be marine organisms. These sentences imply that terrestrial organisms in fresh water/lower salinity water are the major INP source, but this is only one possibility. The authors should include both.

- What does "the upper end of the average values" mean on p.11 l.13? I have never heard of this measure before. Should the authors simply use the average of these 6 values?

- In Fig. 10, there are many conclusions missing that I hope the author would reconsider. First is that similar INP values per volume of air to previous literature is only seen for 2 or 3 stations, at temperatures for -10 to -5 C and more for microlayer samples than seawater samples. Could the authors state that a seawater source of ambient INP should be more important at warmer temperatures than for colder temperatures? At colder temperatures, there may be insignificant contribution of primary emission of INP from seawater. Would their other measurements such as filtering and heat treatment allow for the suggestion that these warm temperature INPs in ambient air may be from primary emission and also biogenic? Can the authors claim any evidence for a known aerosolized biogenic particle in the size range of $0.02 - 0.2$ μm? Is algal and bacterial exudate this size?

References

[1] V. E. Irish, P. Elizondo, J. Chen, C. Chou, J. Charette, M. Lizotte, L. A. Ladino, T. W. Wilson, M. Gosselin, B. J. Murray, E. Polishchuk, J. P. D. Abbatt, L. A. Miller and A. K. Bertram, *Atmos. Chem. Phys.*, 2017, **17**, 10583–10595.

[2] A. Kumar, C. Marcolli, B. Luo and T. Peter, *Atmospheric Chemistry and Physics*, 2018, **18**, 7057–7079.