

Supplemental information

S1 Non-colligative effects of seawater on freezing temperatures

Two ml of a microlayer sample stored at -80 °C for less than 6 months (station 4) was mixed with 2 ml of a solution of Instant Ocean salt in ultrapure water. Instant Ocean is a commercially available sea salt replica used regularly by aquariums to mimic ocean composition (Atkinson and Bingman, 1997). The microlayer sample was mixed with three different concentrations of the Instant Ocean solution (26, 55, and 77 g of salt per kg of water) to make three salinity-altered microlayer samples. The freezing temperatures of the three salinity altered microlayer samples and the freezing temperatures of the three Instant Ocean solutions were measured and are shown in Figure S6.

Station	Photos	Notes	Station	Photos	Notes
1		Foggy. Rippled sea surface with a small swell. Beaufort scale (BS) approx. 3-4. Wind speed 3.8 m/s.	7		A lot of ice and icebergs. Starting to spit rain. Overcast. Polar bears in vicinity. Wind speed 0.3 m/s.
2		Sunny. Next to ice island. Icebergs approx. 100m away. BS approx. 2. Wind speed 6.1 m/s.	8		Overcast. Ice bits around. BS ~1. No polar bears...yet. Wind speed 3.5 m/s.
3		Partly sunny. Light winds. A few icebergs approx. 200m away. Can see land. Wind speed 6.6 m/s.	9		Sunny, ice bits, 1/10 th ice. BS 0. Clouds 1/9 th cirrus. Wind speed 1.3 m/s.
4		Can see Greenland. Overcast, rippled sea surface and a bit of a swell. BS ~2. Wind speed 0.7 m/s.	10		BS ~4, sunrise, clear skies, most wavy conditions yet. Wind speed 6.9 m/s.
5		Sunny, slight swell, rippled surface. BS ~3/4. Wind speed 2.8 m/s.	11		BS ~3, sunrise, mostly cloudy. Wind speed 5.6 m/s.
6		Sunny, rippled waves. Icebergs more than 2miles away. BS 2. Wind speed 4.4 m/s.			

Table S1. Conditions at sampling stations.

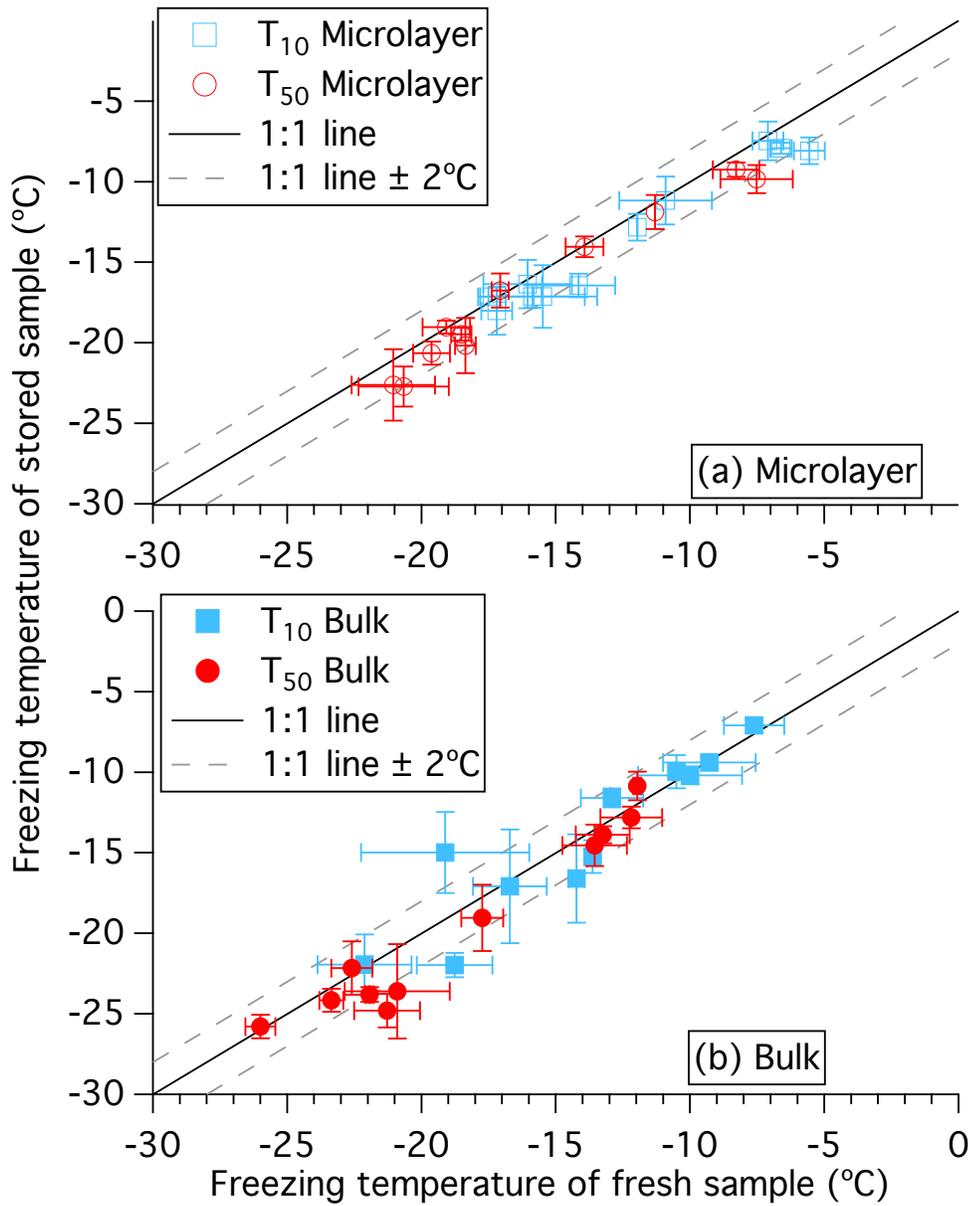


Figure S1. Plots comparing the T_{50} -values and T_{10} -values for duplicate samples not frozen or stored (i.e. fresh) and samples stored at -80°C for up to six months. (a) microlayer samples and (b) bulk seawater samples. Data points are the average T_{50} -values and T_{10} -values from three repeat experiments. Error bars are the 95% confidence interval for three repeat experiments. With experimental variability, all microlayer and bulk seawater samples lie within the $1:1 \text{ line} \pm 2^\circ\text{C}$.

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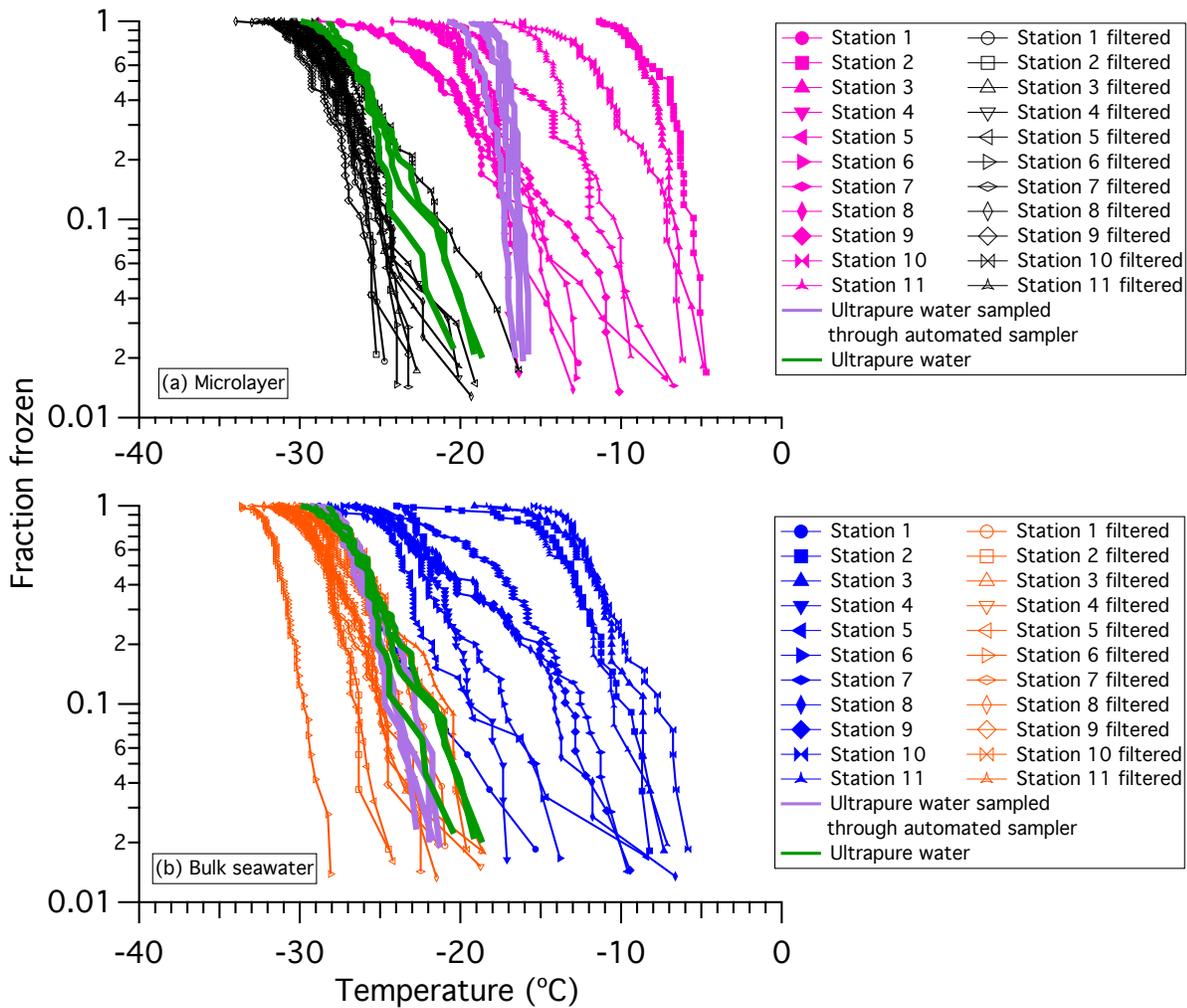


Figure S2. Fraction of droplets frozen (in the immersion mode) versus temperature. (a) and (b) correspond to the microlayer and bulk seawater, respectively. Each set of line and markers represents the results for 3 repeat experiments of a sample or a sample passed through a 0.02 μm filter, adding up to a total of between 45 to 60 freezing events in each set. Each data point corresponds to a single freezing event in the experiments. Also included are the results for ultrapure water and for ultrapure water sampled through the automated sampler (procedural blanks). All microlayer and bulk seawater freezing points have been corrected for freezing point depression to account for dissolved salts in seawater (Section 2.2.4). The uncertainty in temperature is not shown but is ± 0.3 $^{\circ}\text{C}$. The freezing properties of the samples after 0.02 μm filtration have the caveat that they may still contain some small particles (< 0.02 μm in diameter) that can act as INPs (Dreischmeier et al., 2017; O'Sullivan et al., 2015).

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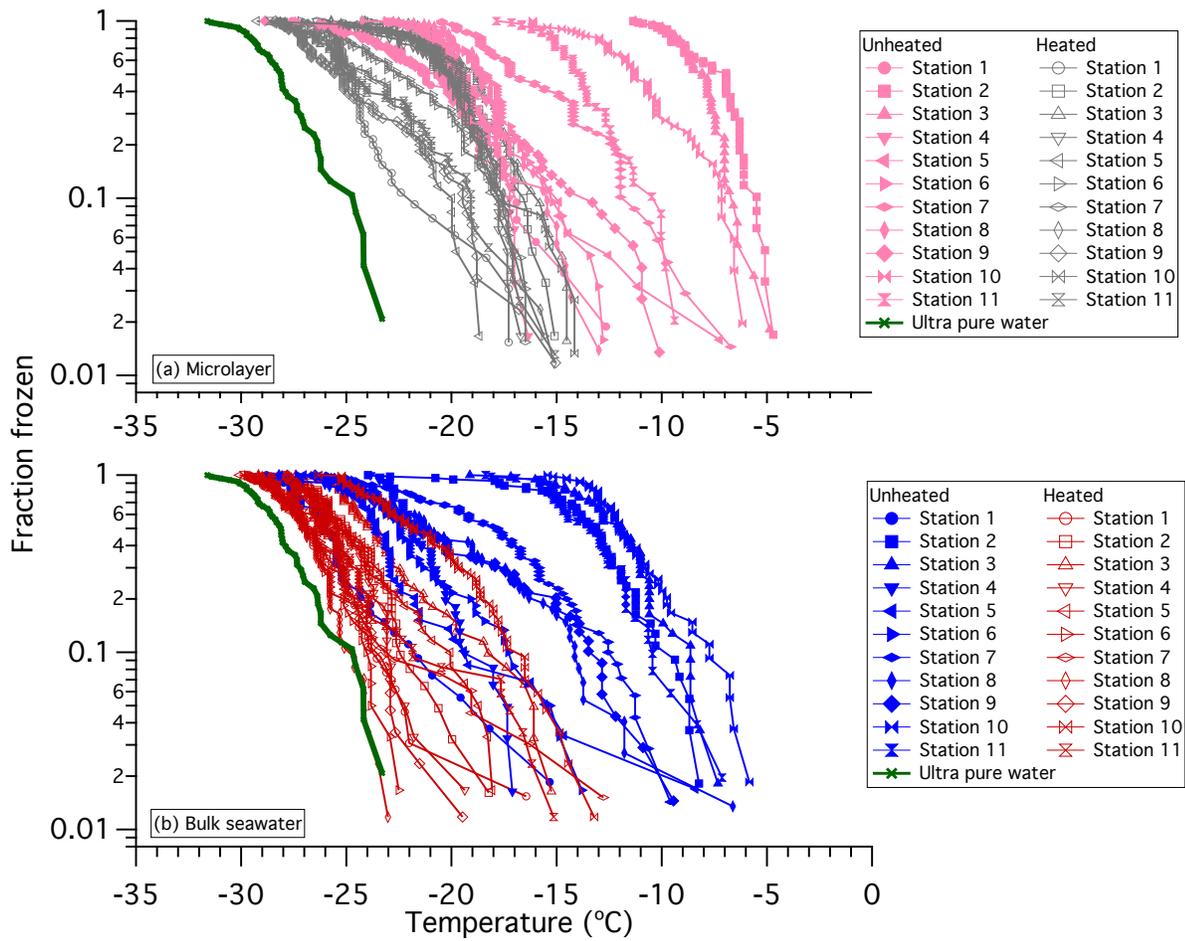


Figure S3. Effect of heating on the fraction frozen curves for unfiltered samples from (a) the microlayer and (b) bulk seawater. Each data point corresponds to one droplet freezing event. All data have been corrected for freezing point depression.

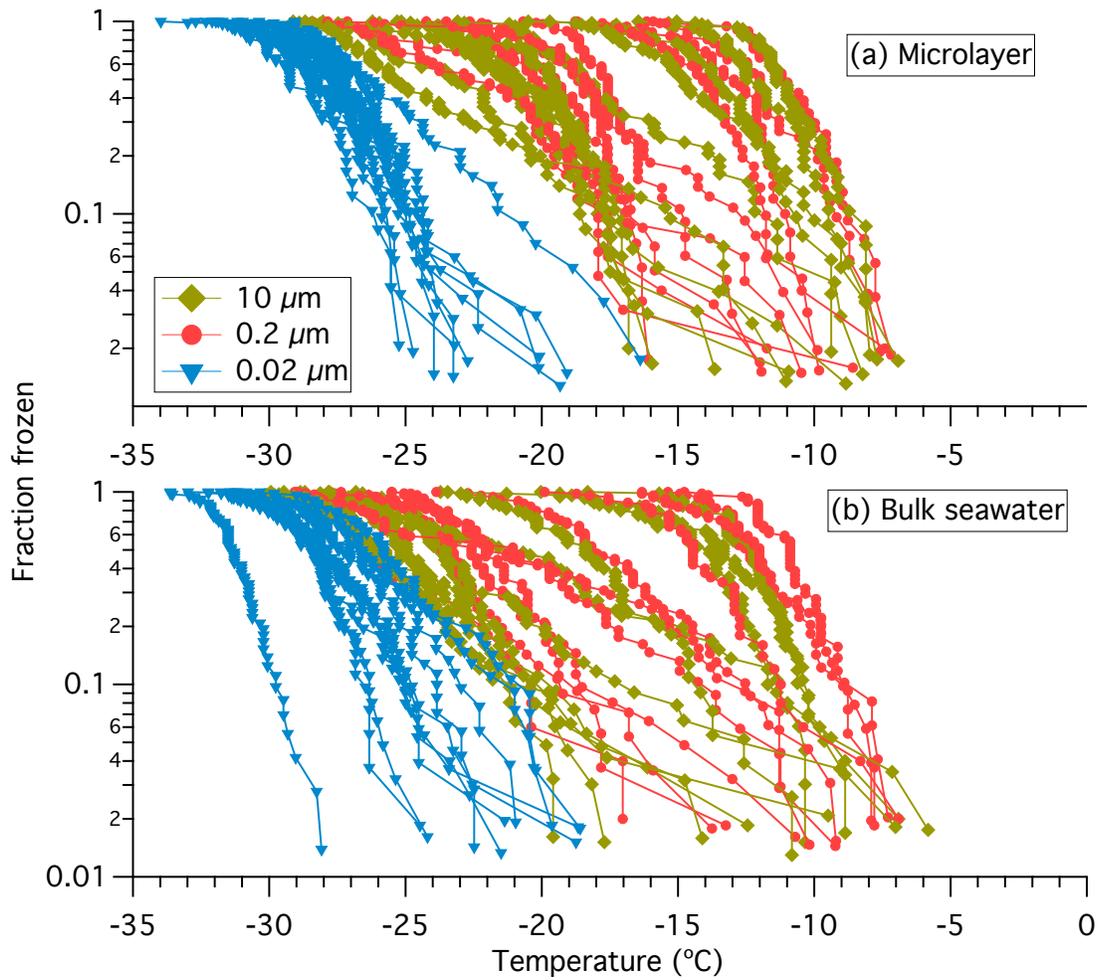


Figure S4. Fraction of droplets frozen as a function of filter pore size in (a) microlayer samples and (b) bulk seawater samples. Filter pore sizes used in the experiments were 10 μm (yellow diamonds), 0.2 μm (red circles) and 0.02 μm (blue triangles). All data have been corrected for freezing point depression.

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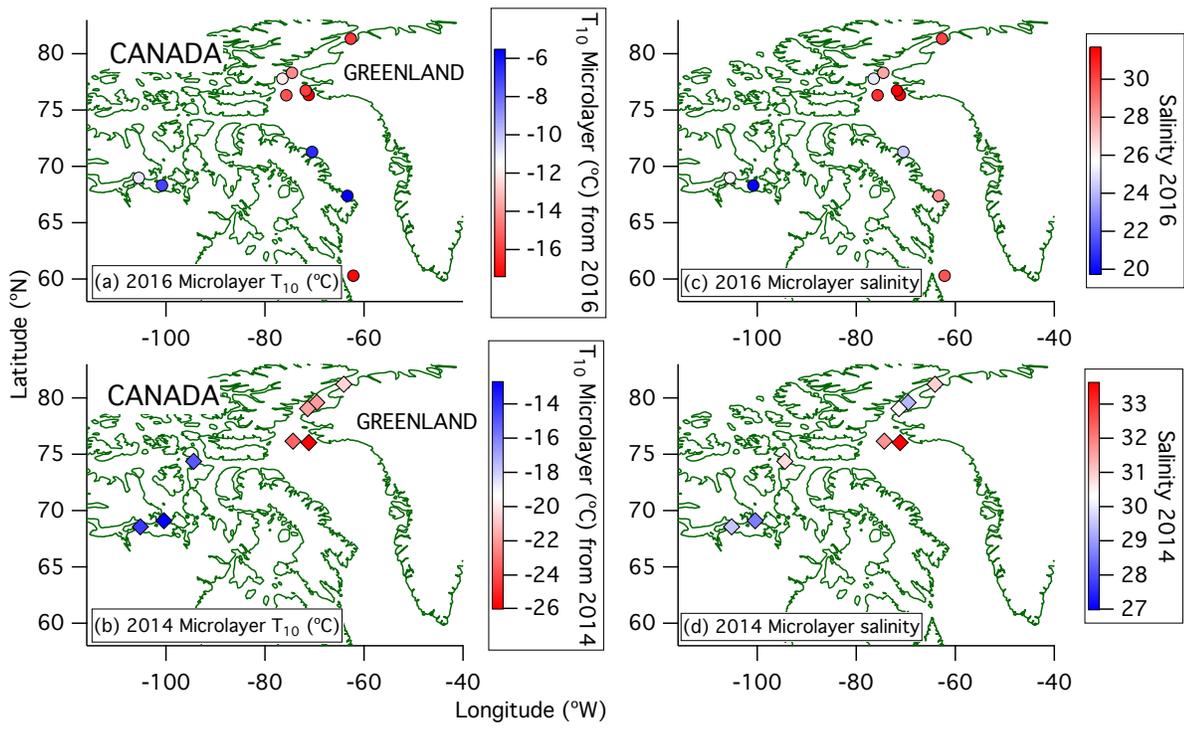


Figure S5. Spatial patterns of (a, b) T_{10} -values and (c, d) salinities in (a, c) 2016 and (b, d) 2014 for the microlayer.

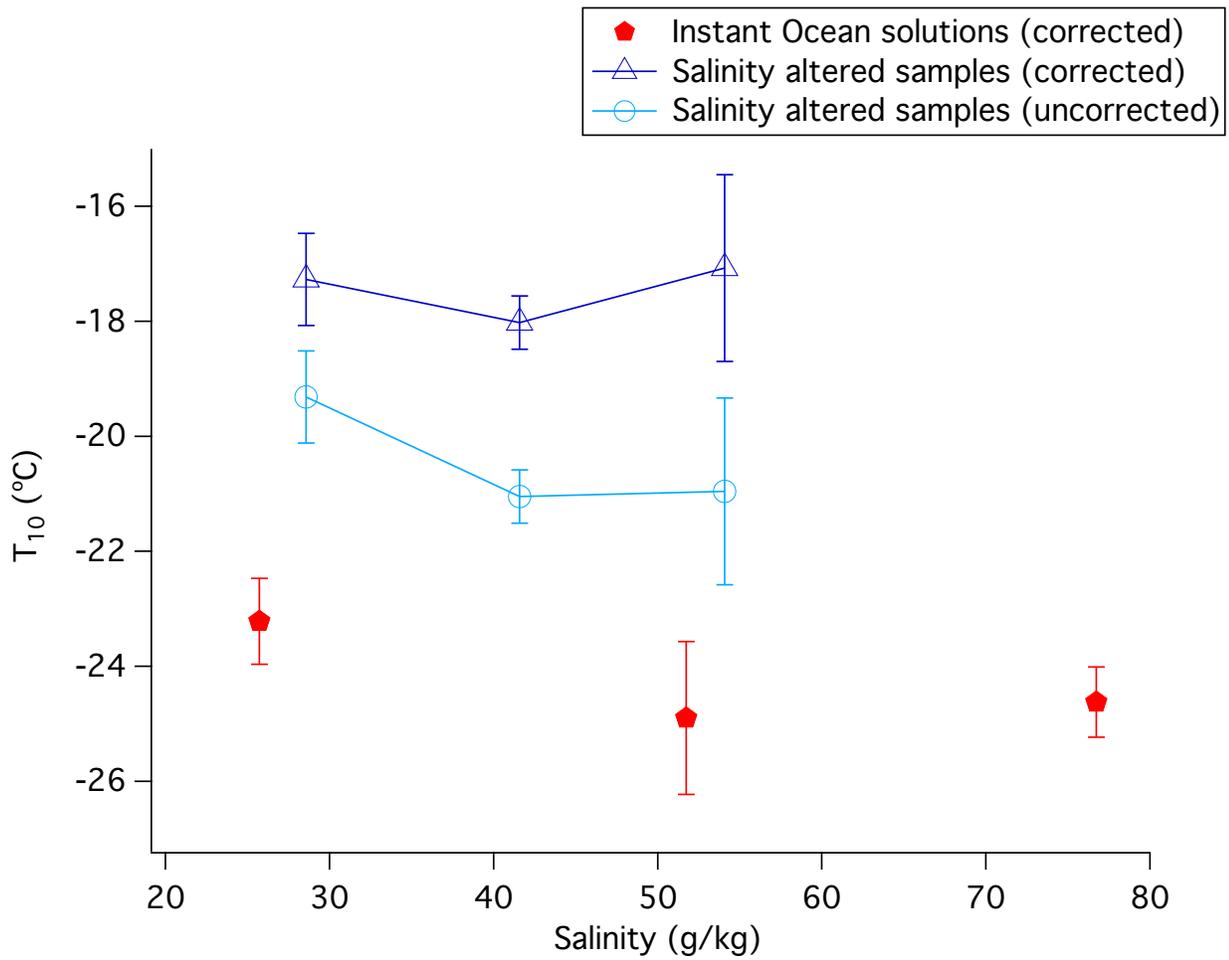


Figure S6. Plot of the T_{10} -values of microlayer sample from station 4 as a function of salinity. The salinity was adjusted by adding solutions of Instant Ocean in ultrapure water to the samples. Data points represent averages and error bars represent the 95 % confidence intervals of 3 repeat experiments. Light blue circles represent T_{10} -values uncorrected for freezing point depression. Dark blue triangles represent T_{10} -values after correcting for freezing point depression by the presence of salts as described in Section 2.3.3. Red pentagons represent T_{10} -values for solutions of Instant Ocean in ultrapure water, after correcting for freezing point depression.

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References

- Atkinson, M. J. and Bingman, C.: Elemental composition of commercial seasalts, *J. Aquaric. Aquat. Sci.*, 8, 39–43, 1997.
- Dreischmeier, K., Budke, C., Wiehemeier, L., Kottke, T. and Koop, T.: Boreal pollen contain ice-nucleating as well as ice-binding ‘antifreeze’ polysaccharides, *Sci. Rep.*, 7, 41890, doi:10.1038/srep41890, 2017.
- 5 O’Sullivan, D., Murray, B. J., Ross, J. F., Whale, T. F., Price, H. C., Atkinson, J. D., Umo, N. S. and Webb, M. E.: The relevance of nanoscale biological fragments for ice nucleation in clouds, *Sci. Rep.*, 5, 8082, doi:10.1038/srep08082, 2015.