Response to Reviewer 1:

Thank you to this anonymous reviewer that made helpful suggestions for strengthening this paper. As requested, we have performed further analyses, adding an additional sample to the heat labile INP section, as well as clarifications in the text and tables. We have made the changes and have hopefully addressed all concerns. Below, we provide a line by line response and detail all the changes (line numbers refer to the tracked changes version). Black text indicates the exact reviewer comment, green is our response, and blue is relevant text changes that appear in the manuscript.

Comments:

1. Elaborate on how exactly INPs affect the surface energy budget via the Arctic Clouds

   The surface energy budget over the Arctic is impacted strongly by cloud properties, such as phase and particle size, and INPs affect these cloud properties. For example, liquid clouds over the Arctic strongly contribute to a positive cloud forcing, a warming effect, and so the level of INPs will change that balance.

   We now added a line:

   Lines 42-45: "They can alter the surface energy budget by impacting the cloud phase and optical thickness, as Arctic liquid clouds strongly contribute to a positive cloud forcing (Shupe & Intrieri, 2004). Replacement of ice with liquid in clouds has been shown to strengthen Arctic amplification, which is the enhanced regional warming due to phenomena such as the ice-albedo feedback (Tan & Storelvmo, 2019)."

   Shupe, M. D., & Intrieri, J. M. (2004). Cloud Radiative Forcing of the Arctic Surface: The Influence of Cloud Properties, Surface Albedo, and Solar Zenith Angle. Journal of Climate, 17(3), 616–628. https://doi.org/10.1175/1520-0442(2004)017<0616:CRFOTA>2.0.CO;2

   Tan, I., & Storelvmo, T. (2019). Evidence of Strong Contributions From Mixed-Phase Clouds to Arctic Climate Change. Geophysical Research Letters.

2. The sampling height at some sites was 1.5m and for some 10m. Are both representative of the surface? How did you compare the two heights?

   This higher sampling height was chosen to get above roads and other buildings at the fixed site (photo below), in order to get an accurate picture of the INPs in the general Arctic boundary layer. For all of the field measurements, we were able to sample closer to the ground to get an idea of what the INPs were like from some of the potential periodic local sources (e.g., downwind of a thermokarst lake). Also, for transportability, having a 10 m sampling height in some of the remote sites would not be feasible. We believe the

measurements are both representative of the surface since they are in the boundary layer, as well as the averages for some types are similar: at -15 °C, the average INP concentration from thermokarst lakes was 0.02 L^{-1} (1.5m), from the ocean was 0.04 L^{-1}, and from the DOE site (10m) was 0.01 L^{-1}. However, the INPs from the lagoon air (1.5m) were higher with an average of 0.16 L^{-1}.

3. In sample analysis section, please close the bracket for 20-fold dilutions (250 microL sample…

   Thank you, Line 117 now reads "...(250 µL sample and 4750 µL 0.1-µm-filtered DI water)...".

4. Why is there data gap for 3,4, 12 and 16 September? The sampling period is very small!

   The 4th, 12th, and 16th were days when only DOE measurements were collected. We added the dates of DOE sampling for completeness and to limit gaps (below). We were not able to sample in the tundra every day due to planning logistics, weather conditions, and availability of polar bear guards that accompanied us.

Date (2021)	*Name*	*Latitude (°)*	*Longitude (°)*	*Environment/ Collection type*	*Samples analyzed*
1-Sep	Emaiksoun Lake	71.25057	-156.77317	Thermokarst lake (TKL)	DA, P, S, TW
2-Sep	Untitled Lake 1	71.23529	-156.30406	TKL: upwind and downwind	DA, UA, AL, P, I, S, TW, V
4-Sep	DOE	71.32272	-156.61506	Fixed	A
5-Sep	Point Barrow	71.38535	-156.46100	Ocean and lagoon	A, LW, SW
6-Sep	Nunavak Bay	71.25240	-156.87332	TKL (brackish)	DA, AL, P, I, S, TW, V
7-Sep	Elson Lagoon	71.29581	-156.26890	Lagoon	A, LW
8-Sep	Will Rogers/ Wiley Post Monument	71.15311	-157.06609	Ocean: onshore	A, AL, P, S, TW, V
9-Sep	Elson Lagoon DOE	71.25556 71.32272	-156.01580 -156.61506	Lagoon Fixed	A, LW, SW A
11-Sep	Untitled Lake 2	71.23806	-156.60472	TKL: upwind and downwind	DA, UA, AL, P, S, TW, V
12-Sep	DOE	71.32272	-156.61506	Fixed	A
13-Sep	Mayoeak River	71.25915	-156.44528	TKL (brackish)	DA, P, I, S, TW, V
14-Sep	Will Rogers/ Wiley Post Monument	71.15312	-157.06609	Ocean: offshore	A, AL, P, I, S
15-Sep	Elson Lagoon DOE	71.31522 71.32272	-156.29845 -156.61506	Lagoon Fixed	A, LW, SW A
17-Sep	Emaiksoun Lake	71.23097	-156.77237	TKL: upwind and downwind	DA, UA, AL, P, I, S, TW, V
18-Sep	DOE	71.32272	-156.61506	Fixed	A

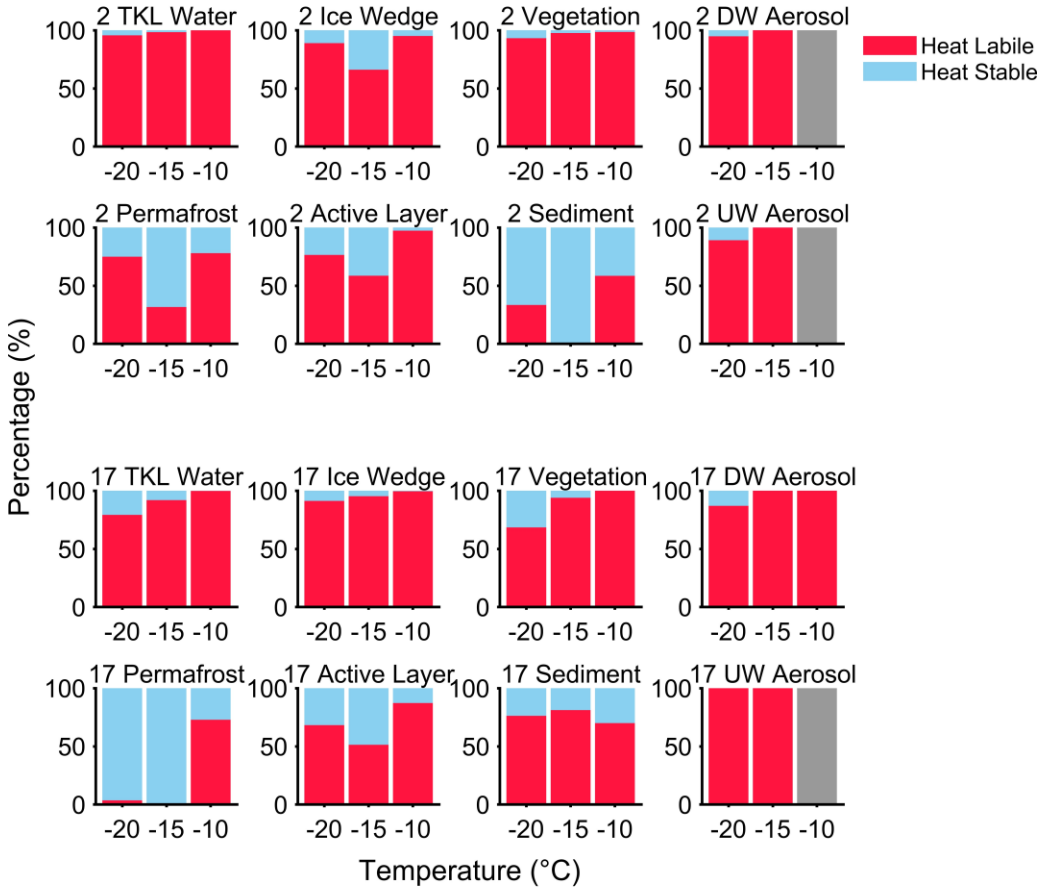
5. Make a table listing all the sources and their concentration of INPs

Thanks for the suggestion, we have now made a Table S1.

Source	*Mean INP Concentration (-15 °C)*
Aerosol	$4.4*10^{-2}$ L^{-1}
TKL	$1.2*10^{5}$ mL^{-1}
Lagoon	$3.1*10^{4}$ mL^{-1}
Seawater	$1.7*10^{4}$ mL^{-1}
Active Layer	$3.6*10^{8}$ g^{-1}
Permafrost	$1.1*10^{8}$ g^{-1}
Sediment	$3.2*10^{7}$ g^{-1}
Vegetation	$2.0*10^{6}$ g^{-1}
Ice Wedge	$1.7*10^{5}$ g^{-1}

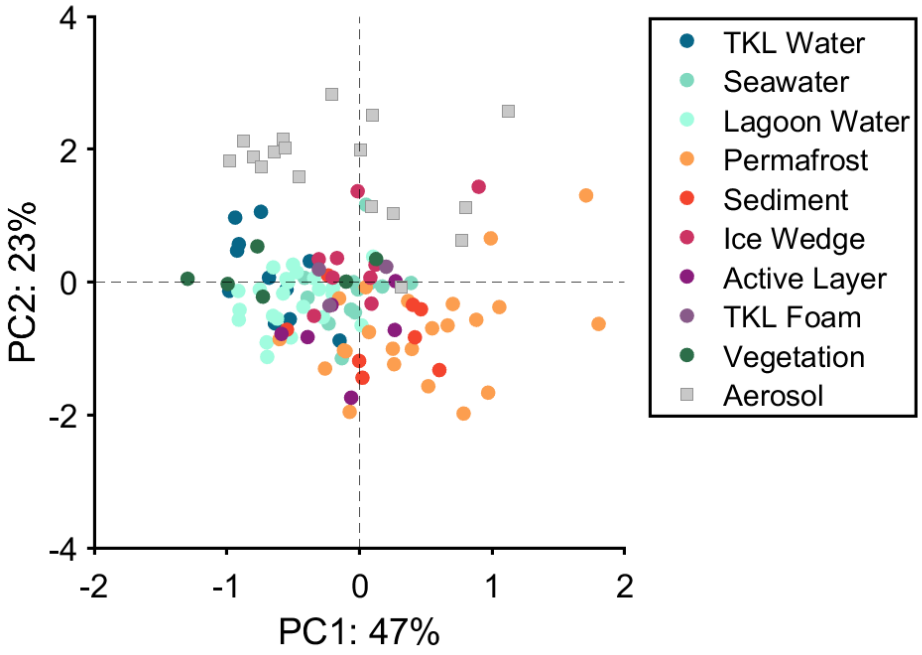
6. To divide INPs into heat labile and stable fractions, why only one day data was used? What was the rationale behind this?

   To strengthen the general conclusions, we analyzed another day and now added that day to the text, with the updated figure shown below.



7. Figure 7: choose different colour scale

   We now updated the color scheme, shown below.



8. Why is correlation weaker with increased homogeneity? Please elaborate.

We believe that the correlation is weaker due to both the INPs and TOC being similar and less variable from the relatively homogeneous lagoon, unlike the thermokarst lakes which cover a larger range due to being sometimes isolated and sometimes connected to the ocean via riverine transport, as well as varying depth and fetch. The ocean itself can have a large range with weather systems enabling waves to kick up sediment, while the lagoon is more sheltered.

9. INPs in the water are predominantly organic….are you referring to heat labile or stable organics?

We only tested the heat labile fraction thus far, so we can only say that. They are strongly heat-labile through sensitivity to 95 °C and so we will add that in the sentence for clarification.

Lines 321-323:

"Based on heating to 95 °C (Fig. 8), the INPs in the water were predominantly heat-labile (presumably organic), and therefore might correlate with TOC."