

Sanchez-Marroquin et al. present airborne measurements of INP concentrations and size-resolved, single-particle aerosol composition over coastal regions of the Pacific Arctic sector in March 2018. Main conclusions include aerosol composition dominated by sea spray and mineral dust. Further, the authors concluded that long-range transported fertile soil dust was likely the main contributor to the INP population during four cases. Reporting these measurements is important, given the limited observations of INPs in the Arctic, and especially observations above the ground. However, the manuscript has several major issues that should be addressed prior to publication in ACP.

### **General comments:**

The introduction is too short and does not include enough relevant background. For instance, there is only a very brief introduction to Arctic mixed-phase clouds here. Some more effort should go into describing AMPCs, INP observations, and how limited they are relative to other latitudes. For the INP aspect, since this is an Arctic manuscript, the introduction material on INPs should focus more on what has been observed in that region. There are some missing key references and studies of Arctic INPs. See my specific comments below on these and what should be included. In general, *some* of the relevant literature is included, but should be more complete and involve discerning where and when the observations were made, since those can vary quite a bit depending on location, sea ice extent, etc. This is hinted at on lines 150-151, but should be elaborated on in the introduction to develop the broader picture for the reported observations.

Even though some of the methodologies are described elsewhere, pertinent information should at least be distilled here so the readers do not have to go on a wild goose chase to obtain such information. Much of it is buried in the appendix, but why not include it in the main text? Here are some questions I had after reading through the methods, information that could be briefly added for clarity:

- When I first read it, it was not clear if only 1 filter was collected per flight, for how long, which altitudes, etc. Table A1 has this information, which I think would be important to include in the main paper.
- There is not enough information on filter collection, preservation, and analysis in the main text. Again, this information is in Table A1 and its caption. Should be moved to the main text. For the altitude, was the aircraft spiraling at that one altitude per filter? If so, what was the spatial coverage of these spirals? Or were these profiles and the one altitude value provided is an average? Need more details here.
- I originally assumed the processing was not done on site, however processing was indeed done on site as stated in the caption for Table A1. This may seem like mundane information, but how the filters are preserved and handled can have a large impact on their results, so is important enough to include in the main text.
- What filter pore size was used for the filters? Different filters have different collection transmission efficiency curves.
- Why was Teflon used for INPs? Teflon tends to have high backgrounds compared to polycarbonate. And why polycarbonate for microscopy? How were carbonaceous particles classified if the filter substrate material is carbon-rich?
- How exactly were the sample suspensions prepared? Shaken in water? For how long? What type of water was used? And was a blank water spectrum tested as well to see how much of the background is from the water itself versus the filter handling?
- How many drops and how many separate tests for the drop freezing experiments?

- What type of cold stage is this? Since it is not a commercial instrument, there needs to be some description of the method, even if just a few sentences.
- What is the temperature range of the cold stage?
- Was freezing detection manual (by human eye) or automatic?
- Why was a 68% confidence interval chosen? That seems like an oddly low one...
- I realize the inlet is described elsewhere, but clear referencing is needed, given aircraft inlets can be riddled with artifact issues.
- For HYSPLIT, why were the heights and 5-days chosen? A topic of discussion in the community is how long back is long enough. I don't have a good answer to that, but the authors should at least justify why they chose the parameters that were used. Additionally, typically a good approach is to run trajectory ensembles (i.e., every so many hours during filter collection and at multiple heights around the sampling point). One trajectory per sample is not enough, given the uncertainties with HYSPLIT, especially near the poles.
- There is hardly any information on the microscopy work. Even though it is in a previous paper, a few more sentences on the methodology should be included here. Where was the SEM-EDX analysis done? What about the details of how the samples were processed on the same filters? Were the samples frozen or stored at room temp and why? There should also be a brief synopsis of how the classifications were defined.

These are just a handful of questions I had, but effectively, the authors should take care to include the relevant methodology information in a succinct manner in the main text instead of in the appendix. That would alleviate most of my concerns here. The details on the calculations and blank corrections could remain in the appendix, if the authors desire, but the rest would be more helpful in the main text.

I am concerned about the background INP spectra. These are very high, and I suspect part of it is due to the use of Teflon and the fact that only a few hundred liters of air were collected per filter. Sure, these volumes would be sufficient in the midlatitudes, but we are dealing with the Arctic here! I see in the appendix that the observations are often at or lower than the background filters. This makes interpretation of the results extremely difficult. I am also not clear on why differential spectra were used for the blank corrections, or why there are still data points for the subtracted when the original datapoint was below the background. It is evident the authors were branching into Arctic measurements (which is great!) and were perhaps not aware of the difficult sampling conditions. Obviously, nothing can be done about this at this point, but the authors should spend the necessary time to describe these caveats in detail in the main text and CAREFULLY interpret the results given these caveats. The article is short as it stands, so certainly has the space to spend on appropriate descriptions of sampling issues that may lead to the results observed.

One of the novel aspects of this work is that the INP measurements were conducted ABOVE the ground. Arctic INP measurements are limited in general, but especially in the vertical. The authors should include more details on how these measurements were conducted exactly (e.g., which heights, etc.; see specific comments below) and include discussion and interpretation on these compared to previous ground based measurements, especially given the Arctic can be highly stratified year-round and certainly in the spring. Even if the filters suffer from high backgrounds and sampling statistical issues, this still affords key information on vertical distributions of INPs. The authors should also compare their results to previous airborne INP measurements such as those from MPACE and especially ISDAC (spring campaign in the same region).

Why is there no discussion on flying in clear air versus cloud? And if out-of-cloud, what percentage of time was spent below versus in? I would expect the results to vary quite a bit, depending on these

conditions. The authors need to include details and discussion on cloud conditions, in addition to those conditions over which surface types (ice, snow, open water, land), and the interpretation of those conditions with respect to the observational results.

### **Specific comments:**

Abstract: One of the benefits of this work is INP measurements above ground, however, the abstract does not discern if there was any sort of vertical gradient in INPs. It would be important to note the altitude ranges somewhere in the abstract, and if the INPs were vertically resolved over the whole flight(s).

Lines 54-55: This is only true for the Arctic haze season in the winter/spring.

Lines 59-60: Some of these were samples collected then processed in the lab, so they *could* contribute to the INP population should they become airborne. It is not clear if they actually do in the real environment.

Lines 60-61: What about dust sources that contain biogenic material, like permafrost? Could just reference Creamean et al. (2020) here.

Paragraph starting on line 67: Should reference the new study by Creamean et al. (2022) for Arctic interseasonal annual cycles. Specifically for the western Arctic, what about the other INP measurements in this region, including airborne studies (e.g., ISDAC, MPACE)? This is especially important to bolster since these measurements are above the ground. These references should be included here for the literature background.

Lines 72-73: Seems like this is an incomplete sentence. I assume the authors mean that Rinaldi et al., in contrast, did not observe the seasonal cycle? They actually do report a small increase in INPs from spring to summer. Important to note here too that Svalbard can be partially ice-free in Apr versus western Arctic locations, and the presence of pack ice can modulate the local sources of INPs.

Line 90: Fig 1 shows air mass backward trajectories. Where are the flight paths? Certainly important to include those somewhere.

Line 109-110: Need to describe what “handling experiment” is a bit more. I assume this means collecting a blank by handling it in the same manner as the sampled filters, but this needs to be explicitly stated.

Lines 123-139: This information belongs in the methods section, not the results and discussion.

Lines 145-146: Was this expected, based on the conditions and previous literature? Why or why not?

Lines 150 and on: The literature comparison is great, but the point to make here is that the authors measured ABOVE the ground. Given the Arctic can be highly stratified, it is challenging to compare what was measured at the ground to that aloft. The advantage of this work is that it is above ground, even if not vertically resolved. This concept needs to be highlighted, here and throughout.

Line 173: No mention of OPCs in the methods. This information needs to be included in that section (i.e., which OPCs, the inlet they were sampling on, etc.). Were the optical data corrected or quality controlled at all? If comparing the SEM and OPC data, there should have been a conversion of the different measured diameters, but I do not see this anywhere.

Section 4: The authors reference the unique properties of their samples to “other regions” they have conducted the sample analyses on, but which “other regions”? The Arctic is a very different place when it comes to aerosol sources and concentrations. It seems as if the authors have not conducted much work in the Arctic, which by no means is a problem, but doing the proper legwork is necessary here. What I mean

here is, specifically comparing to other microscopy studies in the Arctic. Kerri Pratt's group has done a number of studies using CCSEM-EDX in Northern Alaska. I strongly suggest looking up her group's papers (<https://prattlab.chem.lsa.umich.edu/pubs.php>) and comparing/contrasting the reported observations to those. Essentially, more interpretation of the results is needed here with respect to previous work done in the same region (albeit, at the ground).

Lines 180-181: I see the distributions for the SEM and OPC in Fig 4, but what are the percentages of particles detected from SEM via the total population in the overlapping size bins?

Lines 190-192: This could LARGELY affect interpretation of the results! But, what do the authors mean by "artefacts"? Do they mean cloud residuals, compared to interstitial aerosol? Then, were cloud particles collected via filters at times and water evaporated / ice sublimated during inlet residence time or once on the filters? See my general comment above about interstitial aerosol versus cloud residuals. Additionally, any information on the cloud phase? Temperature?

Lines 211-213: Need to cite this. But additionally, how likely is this given the air mass analysis in this region?

Lines 219 and on: Sure, this is true for lower latitudes within the dust belt, but is this really relevant for here? Why not look at trajectories farther back in time if this is a possibility? With the evidence shown, this interpretation does not fit. There are no SEM images shown, so how sure are the authors that what they are calling mineral dust is actually dust versus industrial particles from Prudhoe Bay? Did the authors evaluate the chemical spectra in the context of the morphology as well?

Lines 247-248: This cannot be confirmed without INP treatments. The authors should reword this to demonstrate this is a speculation, albeit a legitimate one.

Lines 254-257: Can the authors elaborate on this? What specifically would the biological material be from during this time of year? Should reference papers like Creamean et al. (2022) and Santl-Temkiv et al. (2019) here that do evaluate INPs/biological particles in the spring. Porter was in the late summer, so it is not exactly relevant for the Mar measurements here. Late summer sources can vary quite a bit from spring, given the contrasting transport conditions, surface open water, and marine versus sea ice biological productivity. Need to compare with previous ISDAC results (also a spring flight campaign).

Fig 1: The flight IDs are difficult to discern without the table being in the main text.

Fig 2: Starts at -14C...is this a sample limit or instrumental bias? If the former, is this because of the blanks for sample volume?

Fig 3: "All Arctic" is a misnomer. These are from Porter et al. from one study in Aug/Sep. Why are the authors showing data from other studies in the same timeframe, but from the late summer for Porter? The Porter data are not actually relevant for this figure. If the authors want to show the specific studies indicated in comparison to, truly all Arctic data, then other studies should be included from other times of the year as well.

## References:

Creamean, J. M. et al. Thawing permafrost: an overlooked source of seeds for Arctic cloud formation. *Environ. Res. Lett.* 15, 084022 (2020).

Creamean, J.M., Barry, K., Hill, T.C.J. et al. Annual cycle observations of aerosols capable of ice formation in central Arctic clouds. *Nat Commun* 13, 3537 (2022).

Tina Šantl-Temkiv, Robert Lange, David Beddows, Urška Rauter, Stephanie Pilgaard, Manuel Dall'Osto, Nina Gunde-Cimerman, Andreas Massling, and Heike Wex. *Environmental Science & Technology* 2019 53 (18), 10580-10590.

See link above for papers from the Pratt group for previous microscopy studies in the same region.