Sanchez-Marroquin et al. describe findings from an aircraft campaign that took place in mid-March 2018 primarily on the north coast of Alaska and the adjacent Arctic Ocean. They collected filter samples of the encountered aerosol and analyzed them with a freezing assay to derive the number concentrations of ice-nucleating particles (INPs), and also used SEM-EDS to obtain a size-resolved composition of the aerosol particles. In addition, they examined back trajectories and compared their results with previous studies and parameterizations. The measured INP spectra were always close to the background and all samples very little variation. They conclude that their samples are dominated by mineral dust from low-latitude sources. They also attribute the ice activity of the samples (below -22°C) to mineral dust and speculate biogenic INPs control the INP spectrum above -22°C. The study lacks exciting results, but that should not be held against the authors. The data pool on Arctic INP is limited, especially for measurements on aircraft. Therefore, any addition to this pool is valuable in itself because it helps us as the authors themselves write, "[...] understand the sources and nature of INP in the Arctic during winter and early spring". Overall, I recommend this manuscript for publication in ACP after some of its shortcomings have been addressed (see general and specific comments below)

General comments

- 1. A bit more discussion of aerosol-cloud interaction is needed. I.e. what do the results mean for mixed phased clouds and the overall INP "budget" in this region?
- 2. The authors suspect low-latitude sources for the mineral dust, is it possible to be more specific in that regard? It is known that mineral dust sources have compositional fingerprints (Scheuvens & Kandler 2014, and references therein, <u>https://doi.org/10.1007/978-94-017-8978-3_2</u>). Could the authors use the SEM-EDS results to constrain possible source regions? Could the authors also use satellite products to at least qualitatively verify the transport of dust from low to high latitudes over the relevant time period?
- 3. Several details on the flights and the sampling strategy are missing in my view, but are needed for a reader to assess the conditions in which the samples were collected. Hence I suggest the addition of the following information to the manuscript (at least in the appendix)
 - Individual Flight tracks and/or height profiles with the sections when a samples was collected highlighted.
 - Indicating if a samples was collected within the PBL/MBL or in the free troposhere; Indicating if sample was collected below/above a cloud layer. This could be done as additional columns in Table A1
 - If available, vertical profiles of aerosol number concentration (e.g. derived by integrating the OPC size distribution) and basic meteorological parameters (temperature, humidity)
- 4. For details on the inlet, the authors refer to one of their previous publications, which is fine, but nevertheless the main key should be repeated in this manuscript. That includes cut-off diameter, is the inlet heated or is the aerosol stream dried in some way to avoid the collection of cloud droplets, is the sampling isokinetic, and where on the aircraft is the inlet located?
- 5. Analogous to my previous comment, the main key points of the SEM-EDS should be repeated in this manuscript and not just referenced. This includes whether there was any form of sample preparation, such as gold sputter coating, what accelerating voltage was used, how long was the collection time for an EDS spectrum, whether the analysis was manual or automatic, (if the

latter, whether particle detection was based on contrast in the BSE image or something else, and whether possible artifacts were removed before further analysis).

L64

Since Tobo et al. 2019 has already been cited above, and because it is consistent with one of your conclusions, it should be explicitly mentioned that dust can also be a carrier for biological INPs

L72-73

This statement about Rinaldi et al.in relation to Wex et al. is correct, but also a bit misleading. Wex et al. attribute the seasonal increase to biological INP and see their effect primarily at high temperatures, with a less pronounced seasonality at lower temperatures. Because of the setup used by Rinaldi et al. they can measure at lower temperatures/higher concentrations. So even if there were a seasonality, Rinaldi et al. would not see it as pronounced in their data, as Wex et al. do. The wording here should take this into account.

L116-117

What is meant by "subset"? Were not all PC filters used for SEM-EDS? If so, what happened to the rest and how was this "subset" selected?

L153-155

I would suggest also including a more recent publication on aircraft INP measurements in the Arctic such as Hartmann et al. (2020), which you cited earlier, in the comparison. They differ in location but are very similar in time (March/April 2018). Borys (1989) may be more comparable in location, but his measurements took place more than 30 years ago, before the extraordinary warming of the Arctic was observed (Arctic Amplification), and some might argue that the differences are related to this.

L155

When making the comparison here, it should also be mentioned which method was used in the respective studies to determine [INP] (freezing assay, CFDC, expansion chamber).

L172

There are many ways to obtain particle diameters in SEM images, it should be mentioned what diameter was used here

L173

More details about the optical probes are needed, such as the name of the instrument, the manufacturer, and where it is mounted. Before reading in the caption of Fig. 4 that it was a PCASP-CDP, I wondered if the OPC might have been fed through the same inlet as the filter sampler. PCASP-CDP, on the other hand, indicates that it is a wing-mounted device.

In addition, it should be mentioned whether the OPC size distributions shown in Fig. 4 represent the average over the entire period in which the SEM sample was taken or whether it is one point measurement during that sampling period.

L175 (+ Fig 4)

Is there a reason why only four samples were analyzed with SEM-EDX? An explanation should be given in the text.

L190-192

PCASP-CDP suggests the combination of aerosol and cloud probe. Doesn't this make it possible to confirm rather than speculate whether the artifacts are actually cloud droplets?

L201

Because of the scale of the y-axis in Fig. 1b, it is difficult to tell when the air masses are below 500m as described in the text. A horizontal line indicating the 500m mark might help, and perhaps a logarithmic axis.

L204-207 (+ Fig 1a)

Looking at sea ice concentration (e.g. https://seaice.uni-

<u>bremen.de/databrowser/#day=11&month=2&year=2018&img=%7B"image"%3A"image-</u> <u>1"%2C"product"%3A"AMSR"%2C"type"%3A"nic"%2C"region"%3A"Arctic3125"%7D</u>) instead of sea ice extent or satellite imagery in general (e.g., <u>https://go.nasa.gov/3TevAJn</u>), it is clear that the ocean around the north coast of Alaska is not completely covered by ice, but is highly fragmented with numerous open leads. This makes the marine particle source mentioned in L207 more relevant in my eyes and should be discussed accordingly in the text.

L210-211

Defining mineral as described here is reasonable in many cases. However, considering that there are algae with Si or Ca-based skeletons and the question of the importance of marine or terrestrial INP sources is still debated, I wonder if algal skeletal fragments were misclassified as mineral dust. Can the authors comment on this? Was visual screening done to determine the amount of non-dusty Si and Ca containing particles?

L219-222

Indeed, studies such as Huang et al. (2015) show that dust from lower latitudes can reach the Arctic within 5 days. The back trajectory analysis in this study also states that the air masses containing the dust particles circulated for at least 5 days. Meaning that the found mineral dust particles with a size of 1-5 μ m had to remain suspended in the air for a minimum of 10 days. Can the authors support the claim this typically happens in the atmosphere?

L234 Fig. 5 (instead of Fig. 3)?

L235-236

Is it not possible to use the SEM-EDS analysis to derive a K-feldspar amount that is more representative of the samples of this study? You already have the "Al-Si rich" category, which should contain mainly the feldspars and their weathering products (illite, kaolinite, etc.). If this category is filtered for appropriate amounts of K, an estimate for the K-feldspar content could be derived. I think studies by K. Kandler and co-authors have covered the categorization of mineral dust with SEM-EDS in detail.

L238-242

It should be noted that the parameterization presented by McClusky et al. (2018) is meant to represent

pristine SSA and they intentionally excluded events with elevated INP concentrations. They also state t during a period of elevated marine organic aerosol from offshore biological activity, [INP] at -15°C was 7.7x10⁻³ L⁻¹. A value that is above the mineral dust and below the fertile soil in Fig. 5.

This, of course, does not refute the author's conclusion that ice activity is related to mineral dust, but the possibility of biological INP of marine origin should be discussed.

McClusky et al. (2018) also present a different parameterization based on TOC. Perhaps the authors can derive an estimate for TOC based on the "carbonaceous" category of the SEM-EDS data and add these predicted INP concentrations to Fig. 5 as well.

L252-254

Following the author's train of thought, the sampled aerosol was transported for about 10 days, and as noted here, a decrease in ice activity would have been expected, yet Fig. 5 shows a very good match with fertile soils collected directly from the ground and brought to the laboratory quickly. This would suggest that the aerosol was not transported far, but may have more local sources. Can the authors comment on that?

In this context, could the use of monthly average snowpack (Fig. 1a) or the sensitivity of the satellite product used for the ERA5 reanalysis be obscuring exposed soils?

L255-256

In the INP community, heat and H_2O_2 treatments are commonly used to gain additional insight into the presence of proteinaceous and organic INPs. Could the authors perform such treatments as the results could further support the claim presented here?

Table A1, second column 2018 instead of 2017

Table A1

Are the presented altitude values averages over the sampling time? If yes, that should be mentioned in the text and caption.

Fig. 1 a)

A reader is not able to recognize where a trajectory starts/ends. If the daily marker would shrink with time, the reader might be able to identify the direction of the trajectory and hence start and end. The authors also chose to show only one return trajectory per sample. For short sample times, a single back trajectory could be representative, but for longer sample times, the aircraft could travel long distances and enter air masses with very different histories. Can the authors comment on the representativeness of back trajectories they show. The authors might also consider adding a figure to the appendix showing high frequency back trajectories for each sample.