

Overview:

This study has some really interesting data that are definitely worth sharing with the scientific community in some form. It is great to see these high latitude data from locations where there were hardly any data at all before. And as the authors pointed out, these data offer very valuable information to contrast and compare with CALIPSO, which up until now has been one of the only sources of lidar information at remote Arctic sea ice locations (but which misses the highest latitude areas and has problems with lidar ratios).

However, the methodologies and associated uncertainties in this paper were not well described, and many of the conclusions were not well supported by the presented data. In particular, a lot of new and interesting techniques are used that are not well validated, but the resulting data presented as if they are known to be accurate. For this reason, I honestly do not know whether I should recommend this paper to be published or not, and I would like to re-evaluate it after the authors have been given a chance to better characterize the uncertainties and reframe the discussion in context of these uncertainties. See more specific comments below.

Specific comments:

- The Punta Arenas and Cyprus data the authors cite for CCN and INP validation of this work (Jimenez et al., 2020a; Ansmann et al., 2020, 2019; Mamouri and Ansmann, 2016) were not actually validated with *in situ* data. I think it is really important to be upfront about this fact, which suggests an unknown degree of uncertainty in the CCN and especially INP estimates for these Arctic data. It should also be mentioned that the Arctic environment is colder and cleaner than these other locations, and has different types of aerosol particles, which might affect the estimates and render previous validations efforts less useful here.
- Many high altitude Arctic AOTs will be very small. How can we be sure that determinations about particle properties can be made at such small signals?
- Can the authors take advantage of the available complementary MOSAiC data (e.g., with INP and CCN near the surface or from tethered balloon) to somehow better validate the data?
- I have made various comments below asking the authors to better describe the uncertainties in various parameters. But I think it is very likely in many cases that uncertainties may not be easy to describe because cloud and aerosol parameters estimated from lidar depend not only on things like conversion coefficients and assumptions about mineral dust, but also on variables like optical thickness of the cloud, and the extent to which the lidar signal has been attenuated. For example, note how the signal has been attenuated beyond the top of the cloud in Fig. 3. Therefore, there is a fundamental challenge when trying to use the methods in this paper to compare quantities like estimated CDNC between clouds, or even between the base and top of clouds for a case study. I am not sure how the authors can address these issues. Possibly a sensitivity study might help.

- Places where uncertainties need to be better described:
 - P6L9: “*The retrieval of aerosol microphysical properties such as particle volume, mass, and surface area concentration and estimates of cloud-relevant properties (aerosol-type-dependent cloud condensation nuclei, CCN, and ice-nucleating particles, INPs) is performed by means of the POLIPHON (Polarization Lidar Photometer Networking) approach (Mamouri and Ansmann, 2016, 2017; Ansmann et al., 2019, 2020)*” Please describe the validation for and uncertainties in this measurement in greater detail. For example, in Jimenez et al. (2020b) it is mentioned that uncertainties in lidar-derived CCN are around 50%, but that is not mentioned here. Please quantify the uncertainties, discuss how were derived and where they cannot be quantified, and how this information affects the interpretation of these results.
 - P6L13: “*Alternatively to the POLIPHON method, we used the multiwavelength lidar inversion technique (Müller et al., 1999, 2014; Veselovskii et al., 2002, 2012) to derive microphysical properties of aerosols including the particle size distribution for detected pronounced aerosol layers.*” Please describe the validation for and uncertainties in this measurement in greater detail.
 - Fig. 5: How well validated are these data? Can error bars in these measurements be applied to this figure?
 - Fig. 6. These are extremely low AOTs. It would be helpful to indicate instrument detection limits on both figures, and to show uncertainty bars, as I would expect these to get increasingly large at low AOTs. The discussion of uncertainties in these data, and how they relate to the conclusions of the study should be further expanded upon in the text.
 - Fig 10: what are the detection limits?
 - Fig. 11: Please discuss whether these are averages over the cloud layer, and if so how that cloud layer was determined. Please change to “estimated effective radius” and “estimated droplet number” in the figure and caption. Please describe in the methods text how the uncertainty range was determined, and discuss the extent to which this uncertainty is meaningful.
 - P13L27: “In this way we estimated CCN concentrations of about 30-70 cm⁻³ with an uncertainty of a factor of 2.” Please describe how this uncertainty factor was estimated, and why this uncertainty estimate is meaningful.
 - P14L2: “*Here, the particle number concentration n_{250} of dust particles with diameters > 500 nm is an input parameter and obtained from the respective lidar observation of the extinction coefficient in Fig. 12 and by assuming a dust fraction of 3-10% in the conversion of the extinction profile into the n_{250} profil (Mamouri and Ansmann, 2016).*” Please provide the uncertainties in the input parameter of dust particle concentrations with diameters > 500 nm and discuss the impact of these uncertainties on the INP estimates? Why assume a dust fraction of 3-10%? As it reads now, there seems to be very large uncertainties,

with estimated INP levels based on unsubstantiated assumptions. Hopefully further discussion can clarify this.

- P16L30: *“In this figure, the number concentration of large smoke particles n_{250} (with radii > 250 nm, lower axis) is shown as well. This number indicates the overall reservoir of favorable INPs (Ansmann et al., 2020).”* Please change to *“estimated number concentration of large smoke particles n_{250} .”* Please explain why this estimate should give us the overall reservoir of favorable INPs, and discuss associated uncertainties.
 - Section 3.4: I find the last 2 paragraphs of section 3.4 to be not useful and mainly conjecture because there are so many assumptions. I suggest removing these paragraphs and Fig. 15 entirely.
- Other places further information is required:
 - For the smoke section, could the authors please clarify why it is definitely smoke, and not a mix of pollution and smoke?
 - P7L26: *“This fourth mechanism is responsible for the occurrence of ULTS wildfire smoke over the North Pole region in the MOSAiC winter half year.”* Please provide evidence or the reference for the deduction that this mechanism is the predominant responsible pathway for this transport during this entire time.
 - P7L30: *“Figure 4 presents the optical properties of the smoke layer as measured on 11 December 2019 (Fig. 3g and h). The smoke layer extended from 8 to more than 18 km height.”* I don't see evidence in Fig. 3 of the smoke layer going past ~13 km. In Fig. 4, the instrument detection limits have not been shown. It would be helpful to add those for the reader to better interpret these plots, and to see how high a detectable layer extended.
 - P8L1: *“No other aerosol type (or cloud type) produces an inverse spectral behavior in terms of the particle lidar ratio”* Please describe the lidar ratio of aged pollution plumes, and say why that could not be a main contributor to the haze event determined here to be mostly made up of smoke particles.
 - P9L16: *“the smoke layer was continuously present and probably homogeneously distributed over large areas of the Arctic.”* If CALIPSO could not observe the layer, what reason is there to believe that the layer was homogeneously distributed over large areas of the Arctic?
 - P9L21: *“Note that we corrected our stratospheric smoke observations in Fig. 6 for PSC effects.”* The authors should describe how they corrected for these observations.
 - P13L10: *“The new method was originally designed for pure liquid-water cloud observations but can be applied to mixed-phase clouds as long as backscattering by ice crystals is negligible compared to droplet backscattering in the droplet-dominated cloud top layer. This condition holds here with ice crystal backscatter coefficients of $5\text{-}10 \text{ Mm}^{-1} \text{ sr}^{-1}$ in the virga and thus also in the cloud top layer and droplet backscatter coefficients of the order of $700 \text{ Mm}^{-1} \text{ sr}^{-1}$.”* Please

provide more information on why this backscatter coefficient can be considered negligible.

- P13L18: *“Later on, the updrafts became obviously stronger, and supersaturation levels exceeded 0.2%...”* Please discuss the evidence behind this statement.
 - P13L19: *“With increasing CDNC the effective radius (characterizing the typical droplet size) decreased and vice versa for constant water vapor conditions ”* Please discuss the evidence behind this statement.
 - Fig. 13: This graph and the text describing it on P14 is a bit difficult to understand. The temperature in Fig. 13a is said to be derived from radiosonde, so why is there only one T value shown, and does it only correspond to a height of 2.5 km? I see RH in Fig. 13b, but no temperature at all? The estimated CCNC, CDNC, INPC, and ICNC values are provided in a range. Is this range meant for a single altitude? Or across the whole figure? Or is it a point measurement range? Please specify where the values are relevant for each parameter, and why the values are only shown for that location/set of locations.
 - P15L24: *“During the 7-day travel in the Arctic the Pacific airmass mixed with the smoke above 7 km. These smoke particles then served as ice nuclei when cirrus formed after further lifting.”* Please discuss the evidence behind this statement.
- Other comments:
 - In the text, when discussing CDNC values, please change from “CDNC” to “estimated CDNC” to reflect the appropriate uncertainty and to avoid confusing readers.
 - P14L20: *“The good match between CCNC and CDNC (liquid-water cloud closure) and between INPC and ICNC (ice cloud closure, see numbers in Fig. 13a) during the early phase of the altocumulus development indicates that the aerosol particles controlled the cloud properties and thus had a strong influence of the evolution of the observed altocumulus cloud system as long as the humidity conditions were favorable. It should be emphasized that such a closure study with consistent findings is only possible if primary ice and droplet nucleation dominates and secondary ice formation, ice breakup processes, crystal-crystal collision and aggregation processes, as well as droplet collision and coagulation, and strong mixing and entrainment processes are absent.”*

This statement seems overly confident and simplistic given the very high uncertainties involved (only some of which are discussed here). Please rephrase to reflect a more accurate level of uncertainty. As an example, if I were writing this study I might say,

“During early altocumulus development in the Figure 13 case study, the estimated CCNC values outside of the cloud are in a similar range to the estimated CDNC levels within the cloud, as are the estimated INPC and ICNC (Fig. 13a). Thus, our data suggest that the estimated cloud active particle levels could

be high enough to control the case study cloud given favorable moisture conditions and in the absence of other processes that might influence CDNC and ICNC levels (e.g., secondary ice formation). This hypothesis would be in line with numerous other Arctic studies that have previously observed this phenomenon (e.g., Mautritsen et al. (2011)). However, higher resolution in-cloud microphysical data are required to verify this lidar-based hypothesis."

- P15L19: *"To demonstrate that the observed wildfire smoke particles were able to control cirrus evolution and life time we present the results of a first MOSAiC case study here. The observation is from 6 December 2019 (Fig. 3c and d)."* Is this cloud even a cirrus cloud? The lidar signal extends down to near the surface at times, and the top is below 8 km altitude. What has been done to ensure that this is not actually a mixed phase cloud? The temperatures near the base of the cloud appear to be as high as -30C or so, from Fig. 14b, and liquid water can be present at such temperatures in the Arctic.
- P15L29: *"This part of the smoke layer (above 9.3 km) can be regarded as the main reservoir of INPs."* Why is it assumed that this aerosol layer is in contact with the ice cloud? To me it looks distinctly separate for most of the time.
- I like the introduction, it really gets the reader interested in the study.
- P.3, paragraph starting on L4: Here or elsewhere, you might also consider mentioning relevant Arctic high altitude smoke findings from Schill et al. (2020).
- P4.L22 *"... HSRL is of advantage during the summer half year (when Raman lidar observations are of limited use)"* Please specify why (and if relevant, which) Raman lidar observations are of limited use during the summer. Also, can't HSRL also be used during the winter? If so, for clarity please explain to the reader what additional capabilities the Raman lidar provided that the HSRL could not.
- P4L29: *"Di Biagio et al. (2018) were the first to run lidars (mounted on an ensemble of autonomous drifting buoys) in the Central Arctic, ..."* The authors might consider mentioning that these data were collected on buoys.
- P6 *"'Co' and 'cross' denote the planes of polarization parallel and orthogonal to the plane of linear polarization of the transmitted laser pulses, respectively."* This sentence should probably go in the previous section where the authors first mention the co- and cross terms.
- Fig. 3: It might be easier on the reader to just state: "range-corrected 1064 nm signal" and "linear depolarization ratio" above the columns in the figure instead of in the caption. To avoid confusion, the authors might also want to note in the caption and/or on P7L3 that the y- and z-axis limits were varied between panels in order to highlight different features.
- P8L5: *"The size distributions of the smoke particles were obtained from the Polly observation by applying the lidar inversion method to the layer-mean backscatter and extinction information (Veselovskii et al., 2012)."* This information would be better placed in the methods section.
- Fig. 6a: This figure is not intuitive to me. Please tell readers what the height and base of the bars indicates (the top and bottom of a smoke layer?). Please tell

them whether the colors are the relative fraction, or the dominant feature at that altitude (or something else). I am confused about the colors also because in the caption it says “The color in the bars provides information about the smoke particle concentration in terms of particle extinction coefficient at 532 nm.” Please explain exactly what the particle extinction coefficient tells us about estimated smoke particle concentration. Please state in the figure and not just the caption that colors relate to particle extinction coefficient at 532 nm. Again, how do detection limits play into these bars? Please state whether the bars are only the observations above detection limits of the lidar. If the observations are below the detection limits, please either get rid of them, or clearly state why the data are still useful (I would guess they would not be). To avoid confusion, perhaps get rid of the height levels redundantly shown on the right side of the figure. Are the black dots the tropopause on that day? If so, an arrow from the word “tropopause” pointing to the dots might help clarify things. I know it was mentioned in the text, but can the authors mention in the caption as well in just a few words how the tropopause was determined?

- Fig 6b and 6c captions: Please change “column mass concentration” and “vertical mean particle mass concentration” to “estimated column mass concentration” and “estimated vertical mean particle mass concentration” to indicate appropriate uncertainty
- P9L21: “*Note that we corrected our stratospheric smoke observations in Fig. 6 for PSC effects.*” This note should go in earlier with discussion of Fig. 6.
- P9L22: “*This type is made up of supercooled liquid ternary solutions that consist of H₂SO₄, HNO₃, and H₂O.*” Speculation on the chemistry may be beyond the scope of this paper. I suggest saying “likely consist” instead of “consist.”
- P9: “*The temperature at PSC base height showed values of -78°C and at the backscatter maximum the Polarstern radiosonde measured a temperature of -86°C .*” Wow, that is cold!
- P10L12: “*Height-resolved lidar observations of Arctic haze, prevailing during the late winter and early spring months, are rare (Di Pierro et al., 2013; Di Biagio et al., 2018).*” I suggest rephrasing this. The CALIPSO observations have taken observations in clear conditions over the entire Arctic since 2006, taking observations of plenty Arctic haze events.
- P10L15: “*However, knowledge about the vertical layering structures of Arctic haze is still limited and mostly based on snapshot-like aircraft observations performed during field campaigns, preferably in spring.*” Again, I am not sure that is entirely true, given the extensive CALIPSO observations.
- P10L27: “*Type-Ia PSCs consist of nitric acid trihydrate (NAT) crystals and produce significant depolarization of backscattered laser light.*” Suggest rephrase to “...are thought to consist of ...”
- P12L30: “*After nucleation, the ice crystals grew fast to sizes of 50–100 μm within minutes (Bailey and Hallett, 2012) and immediately started falling out of the altocumulus layer. The ice crystals partly evaporated on the way down, but partly*

reached the ground as precipitation.” Please clarify here whether you are talking about findings from the Bailey and Hallet, 2012 study, or whether you are talking about results observed during MOSAiC.

- Fig. 12: Please replace “CCN” with “Estimated CCN” in the figure and caption.
- P15L19: This paragraph would benefit from a Figure showing the trajectories being discussed.
- P16L34: “*As mentioned, ice nucleation occurs during updraft periods, more precisely when a certain (threshold) supersaturation level is exceeded.*” The authors may want to mention that ice nucleation also requires cold enough temperatures.
- Fig. 15: Again, please put estimated ahead of any parameters that were not directly measured and that include substantial assumptions in the caption.

Technical comments:

- Title: “an introductory” should be changed to “an introduction.” But maybe also consider making the title more succinct to make it more appealing to readers. Note: most readers will likely not know what UTLS is, suggest dropping it from title.
- L5: “... aboard **the** Polarstern.”
- Caption, Fig. 2 (and also corresponding text p. 4, L.9): “Figure 2. Polarstern drifting in the Arctic ice on 10 April 2020 (left panel) and measurement containers for in situ aerosol monitoring (the two first containers on the left side and the first container on the right side), and for remote sensing of aerosols and clouds (right panel). The OCEANET container of TROPOS is the third one on the left side.” Could the authors please clarify whether they meant third one to the back, or the one in the front?
- P6L12: “Hofer et al. (2020) exemplary shows...” Did the authors mean something like, “Hofer et al. (2020) is an example showing...”?
- P7L28 “poleward”
- P14L2: profile not profil.

References

Ansmann, A., Mamouri, R.-E., Bühl, J., Seifert, P., Engelmann, R., Hofer, J., Nisantzi, A., Atkinson, J. D., Kanji, Z. A., Sierau, B., Vrekoussis, M., and Sciare, J.: Ice-nucleating particle versus ice crystal number concentration in altocumulus and cirrus layers embedded in Saharan dust: a closure study, 19, 15087–15115, <https://doi.org/10.5194/acp-19-15087-2019>, 2019.

Ansmann, A., Ohneiser, K., Mamouri, R.-E., Knopf, D. A., Veselovskii, I., Baars, H., Engelmann, R., Foth, A., Jimenez, C., Seifert, P., and Barja, B.: Tropospheric and stratospheric wildfire smoke profiling with lidar: Mass, surface area, CCN and INP retrieval, 1–45, <https://doi.org/10.5194/acp-2020-1093>, 2020.

Jimenez, C., Ansmann, A., Engelmann, R., Donovan, D., Malinka, A., Seifert, P., Wiesen, R., Radenz, M., Yin, Z., Bühl, J., Schmidt, J., Barja, B., and Wandinger, U.: The dual-field-of-view polarization lidar technique: a new concept in monitoring aerosol effects in liquid-water clouds – case studies, 20, 15265–15284, <https://doi.org/10.5194/acp-20-15265-2020>, 2020a.

Jimenez, C., Ansmann, A., Engelmann, R., Donovan, D., Malinka, A., Schmidt, J., Seifert, P., and Wandinger, U.: The dual-field-of-view polarization lidar technique: a new concept in monitoring aerosol effects in liquid-water clouds – theoretical framework, 20, 15247–15263, <https://doi.org/10.5194/acp-20-15247-2020>, 2020b.

Mamouri, R.-E. and Ansmann, A.: Potential of polarization lidar to provide profiles of CCN- and INP-relevant aerosol parameters, 16, 5905–5931, <https://doi.org/10.5194/acp-16-5905-2016>, 2016.

Mauritsen, T., Sedlar, J., Tjernström, M., Leck, C., Martin, M., Shupe, M., Sjogren, S., Sierau, B., Persson, P. O. G., Brooks, I. M., and Swietlicki, E.: An Arctic CCN-limited cloud-aerosol regime, *Atmos. Chem. Phys.*, 11, 165–173, <https://doi.org/10.5194/acp-11-165-2011>, 2011.

Schill, G. P., Froyd, K. D., Bian, H., Kupc, A., Williamson, C., Brock, C. A., Ray, E., Hornbrook, R. S., Hills, A. J., Apel, E. C., Chin, M., Colarco, P. R., and Murphy, D. M.: Widespread biomass burning smoke throughout the remote troposphere, 13, 422–427, <https://doi.org/10.1038/s41561-020-0586-1>, 2020.