

Dear reviewer,

thank you for careful reading of the manuscript and for providing valuable comments and ideas how to improve the paper.

A brief overview of main changes in the beginning:

We have now two separated sections, for MOSAiC instrumentation (Section 2) and for lidar retrievals (Section 3, aerosol microphysics, CCN, INP). Now the instrumental and data analysis methods are better separated. Furthermore, Section 3 is better structured with subsections from 3.1-3.3 and from 3.4.1-3.4.5.

We simplified the INP parameterization significantly. We have two INP retrieval sections now: Sect. 3.4.1. and Sect. 3.4.2. In Sect. 3.4.1., we describe the ABIFM approach (Knopf and Alpert, 2013) to estimate immersion freezing INPs from lidar observations in the lower troposphere. In Sect. 3.4.2, we describe the DIN approach (Wang and Knopf, 2011) to estimate deposition ice nucleation INPs from lidar observations in the upper troposphere. We removed the immersion freezing and deposition nucleation INP parameterizations of Ullrich et al. (2017). This was necessary in order to avoid 'mixing' of time-dependent and time-independent INP parameterizations... and the difficulties, arising from this mixing of methods, when showing the obtained results in ONE MOSAiC time series. Now the full presentation of the annual cycle of INP observations is much more straight forward. We adjusted the discussion of the results after these changes.

We also changed the titles of Section 4 and 5 a bit: Section 4: Observations, part 1: ...optical properties..., and Section 5: Observations, part2: cloud-relevant products, CCN, INP etc. This may better indicate that the RESULT block is separated into two main parts.

We introduced one new figure (Figure 12), as requested by one of the reviewers.

We went through the entire manuscript and improved the text after all the changes and with all the comments of the reviewers in mind.

Now the step-by-step response to all comments with our response in blue. Essential changes in the manuscript are indicated in BOLD.

This paper follows, as many others, the MOSAiC campaign conducted from the oceanographic vessel Polarstern that was conducted over the years 2019 and 2020. It is mainly focused on the exploitation of data from the Raman lidar that was on board the ship. It is a unique dataset from the ice pack because on an annual sampling of the Arctic atmosphere. The paper is very well organized and written. It contains many relevant references to support the statements. It is totally within the scientific domain of ACP.

For all these reasons, I think it can be published without major changes.

In addition, it raises current issues such as the impact of biogenic aerosols on ice cores or the role of biomass burning aerosol that can be mixed with terrigenous particles. These are still open scientific fields of importance for climate projections.

**This comment motivated us to even include recent results of Tobo et al. (2019) and Kawai et al. (2023) who pointed to the importance of high latitude dust (glacier washout products containing biogenic material) regarding heterogeneous ice nucleation at high temperatures, >-15°C (on page 3).**

Below are some questions/comments.

Introduction.

- 1) It is very complete, maybe a little long. Some parts could be more synthesized, such as the discussions on CALIPSO that happen in two different places.

**We agree and shortened the Introduction considerably (submitted version: 3.5 pages, revised version: 2 pages).**

- 2) L16. I think we are talking about the middle troposphere?

**In the Abstract, we now write: ... caused a re-increase of the aerosol concentration towards the tropopause.....**

- 3) L31. The ship was also trapped in the ice in August/September?  
**Polarstern was again at latitudes >85°N from 21 August to 20 September 2020 and thus in the ice. This is now written on page 3 in the Introduction.**

Section 2.

- 4) L212. The optical thicknesses are low, and this will induce very large errors on the Angstrom coefficient. It should also be taken into account in the interpretations.

**Yes, we agree, we re-phrased the text a bit. However, all the AERONET observations in the Arctic shows such a narrow range of Angstrom values. We think, all these Angstrom values are trustworthy.**

- 5) In subsection 2.6, it is assumed that aerosols do not change in nature with altitude in order to apply the same coefficients  $c$ ?

**We mention now at several places in the revised text that we assume similar aerosol conditions up to 3 km height, so that we can apply the conversion factors for the 250 and 2000 m height levels.**

**We also discuss that these conversion factors (based on summer time AERONET observations) may not be fully appropriate for Arctic haze conditions (winter time aerosol conditions).**

- 6) In fact, these coefficients  $c$  are the inverse of cross sections, why not use directly this very explicit quantity in physics?

**You are right! On the other hand, we have these simple formulas, Eqs.(1)-(3), in Sect. 3.1, we leave it as is.... in order to confuse readers not too much.**

- 7) L231. On which dataset is the regression done, it is not very clear?

**We explain all this now in more detail in Sect. 3.1 (in the paragraph after introducing Eqs.(1)-(3)). The full procedure is, however, described in Mamouri and Ansmann (2016). That is mentioned as well.**

- 8) L320. Can't there also be nitrates on the dust?

**Yes! By passing through polluted regions dust particles can become coated with sulfate, nitrate, and organic substances. This is now mentioned on page 11 in Sect. 3.4.1.**

- 9) Subsection 2.8.3. If a thermodynamic model is used to calculate INPs from the lidar measurement, how can this independently validate the climate models? Don't these models use related approaches?

**The INP parameterizations are obtained from laboratory studies, i.e., from observations at well-defined temperature and humidity conditions and for well-defined aerosol properties**

(chemical composition, size distribution). They are not obtained from modelling. In the lidar retrieval we use temperature, humidity, and aerosol information to estimate INP concentrations by using the laboratory findings. Climate modelers may use the same INP parameterizations.

**We do not want to validate climate models?**

Section 3.

10) L464. How can we be sure that it is deep convection, linked to a pyroCb, that injects the wildfire smoke into the lower stratosphere? Is it the altitude at which the aerosols are observed by the lidar?

**No, we cannot be sure! Mike Fromm (reviewer #3) had a similar comment. Now we provide an extended discussion on the potential smoke sources in the last paragraph in Sect. 4.1. We include the findings in the article of Hu et al. (2022). These authors observed the Californian smoke (that partly travelled to the Arctic from 10-19 September 2020).**

**According to Hu et al. (2022), intensive wildfires in California and Oregon injected large amounts of wildfire smoke into the atmosphere on 10 and 11 September 2020. Thick smoke layers at 5-10km height were detected with CALIOP over the Pacific Ocean just west of the west coast of North America Hu et al. (2022) ..... Hu et al. (2022) mentioned that pyrocumulonimbus (pyroCb) development occurred on 9 September and that the smoke was trapped over the eastern Pacific Ocean on 7-11 September due to cyclone activity. It remains open to what extent strong convective motions were responsible for smoke lofting up to the upper troposphere.**

11) L467. We found the same thing on biomass burning aerosols from Canada (<https://doi.org/10.5194/acp-18-13075-2018>)#

**We now consider this (plus reference) in the last paragraph in Sect. 4.1.**

12) L475. In Fig. 6, how many lidar profiles are averaged per month? Are they homogeneously distributed in the month?

**No! They are not homogeneously distributed. We used all the cloudfree periods for aerosol profiling. Now, we provide numbers of observations per months for all MOSAiC months (Sect. 4.2, first paragraph).**

13) L544. Is it normal that the profiles of particle number concentration are not shown? It would have been interesting to see.

**The number concentrations are the most uncertain products (surface area and volume concentrations are much more robust retrieval products). We discuss that a bit more at several places (Sects.3 and 5). This is the main reason why we hesitate to show number concentration profiles.**

Section 4.

14) L591. In Fig. 10, the empty blue circles are not identified.

**We use one set of conversion factors (for Arctic summer aerosol). In the submitted version, we used conversion factors for marine aerosol, i.e., for sea salt, in summer (Mamouri and Ansmann, 2016). That was a mistake! We changed that and use the conversion factors for Arctic summer aerosol throughout the MOSAiC year now. Therefore, we have only closed circles in Fig.10 of the revised version.**

15) On Figures 10, 11 and 12, wouldn't it be clearer to put envelopes of data variation?

**We do not like this idea! However, we now provide seasonal mean CCN values (for winter and summer seasons, for surface, 250, and 2000 m height) in the text, in Sect. 5.1. This helps to see the variations in the data, and these variations are mostly due to natural (atmospheric) variations.**

16) How can we separate natural variability and uncertainty from these figures?

**Table 1 contains uncertainty information for the lidar products. The uncertainty in the in situ observations is lower. We now provide mean values and standard deviations of measured data, and the obtained variability of 50-100% can be interpreted as natural or atmospheric variability. That is at least our position.**

17) L610-611. I don't quite understand the sentence about the dry deposit.

**We removed the paragraph on low level jets to keep the discussion short and to avoid confusion by too many hypotheses.**

18) L631. Why did you take 1% at 250 m? Did the in-situ measurements give 1% dusts in number?

**We adjusted our lidar derived INP values to the in situ measured INP values. This is now described in Sect. 5.2 (page 21).**

19) In-situ measurements are usually on mass, there must be significant errors to pass in numbers. Is this the case?

**In Sect. 2.5, we now provide details on the methods regarding the in situ observations aboard Polarstern. Particle number concentrations and INP number concentrations are derived from these observations. The uncertainty analysis is described in the respective MOSAiC papers of Boyer et al. (2022) and Creamean et al. (2022). Well-characterized and well-established retrieval methods are used. The uncertainties are certainly much lower than 50%.**