

The paper “UTLS wildfire smoke over the North Pole region, Arctic haze, and aerosol-cloud interaction during MOSAiC 2019/20: An introductory” presents and discusses remote sensing observations obtained during the MOSAiC campaign. The unique dataset provides new opportunities to explore aerosol-cloud interactions at the North Pole. Persistent smoke layers originating from large scale fires are discussed and analyzed in terms of their properties as well as the ability to act as Cloud Condensation Nuclei and Ice Nuclei (CCN and IN). Observations of the Arctic haze during the campaign are presented and discussed also, aiming to enrich the current observational dataset with winter time measurements.

The study falls within the scope of ACP. The manuscript is well-written and structured, the presentation is clear, the language is fluent and the quality of the figures are high. In order to help improve the manuscript, I would kindly suggest the authors to take into account the following comments.

General comment:

Please provide the typical uncertainties of the lidar-derived aerosol and cloud microphysical properties discussed, originating from the conversion assumptions and the uncertainties of the optical properties derived from the lidar measurements. Additionally, please discuss the effect of the low aerosol concentrations presented here, on these retrievals. Please provide information on independent validation studies of the lidar-derived microphysical products (e.g. using in-situ measurements).

Specific comments:

Page 2, lines 29-32: “The MOSAiC lidar observations (together with the radar observations of the ARM mobile facility) allow us, for the first time, to investigate the role of smoke in ice formation processes”. The authors should specify the Arctic region in the sentence, since there have been several studies exploring the potential of smoke particles to act as IN (.i.e. Peters et al., 2009; Prenni et al., 2012; Levin et al., 2016; Schill et al., 2020). Or specify if the statement is meant for combined lidar and radar observations in the Arctic.

Page 3, lines 1-3: “A unique opportunity is thus given to explore to what extent the wildfire smoke particles, providing a significantly enhanced number of sites for heterogeneous chemical processes (chlorine and bromine activation), contributed to the strong ozone depletion“: Since the article provides hints on the role of smoke particles on ozone depletion, rather than “findings”, please rephrase accordingly.

Page 3 lines 4-30: “The article is organized as follows... Sect. 4 finally provides some concluding remarks”: This section is hard to follow, since it mixes the scientific objectives of the study with the proposed methodology/techniques and the article layout. Please divide this part in three paragraphs, with the first containing the scientific objectives of the study (e.g. "Organic aerosol particles are ubiquitous in the atmosphere around the world and there is an urgent need to investigate..."), the second one containing the proposed methodology/techniques and the third containing the structure of the article with very brief descriptions for its section (e.g. "... In Sect. 3.2, we present two cases of Arctic haze observations performed in February and March 2020").

Page 3, lines 21-22: “Recently introduced new remote sensing analysis concepts (closure studies) (Ansmann et al., 2019) are applied for the first time to Arctic clouds”. Please also include the work of Marinou et al., (2019).

Page 5, lines 9-10: “which permits accurate aerosol and cloud profiling from about 800 m to 30-40 km height”. Please specify separately the information on aerosol and cloud detection ranges and provide information on the accuracy of the aerosol and cloud retrievals for different altitude ranges. Which are the typical signal-to-noise (SNR) values of this lidar for aerosol layers at 10, 15 and 20km a.s.l.? What are the uncertainties of the lidar-derived properties at these altitudes?

Page 5, lines 16-19: “This technique enables us to determine multiple scattering by droplets in liquid-water dominated cloud layers and to determine from this multiple scattering information cloud microphysical properties (e.g. effective droplet size and cloud extinction coefficient) (Jimenez et al., 2020a). The method is based on depolarization ratio (ratio of cross-to-co-polarized backscatter coefficient) observations at the two FOVs”. This part is more appropriate for section 2.2 where the rest of the lidar products and retrievals are presented. I suggest to move it before **page 6, line 15-16:** “Details of the retrieval of microphysical properties of liquid-water cloud layers can be found in Jimenez et al. (2020a, b)”.

Page 5, lines 26-27: “we use the preliminary radiosonde products that were directly available during the expedition”. Explain the “preliminary” definition in the radiosondes used, and why you used these instead of the consolidated radiosonde products.

Page 6, lines 9-16: “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). Hofer et al. (2020) exemplary shows the full set of POLIPHON aerosol products in the cases of an 18-month Polly campaign in Dushanbe, Tajikistan, for central Asian aerosol. 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. Details of the retrieval of microphysical properties of liquid-water cloud layers can be found in Jimenez et al. (2020a, b)”. Please provide a short description of the assumptions used for the aerosol and cloud microphysical properties retrieved from the lidar products. Please comment on independent validation studies for these products (e.g. with in-situ measurements as in Marinou et al. (2019) study). Please quantify and discuss the uncertainties of the aerosol and clouds microphysical retrievals for the observations presented on this study.

Page 7, line 2: “The measured linear depolarization ratio in the right panels of Fig. 3 allows us to precisely distinguish cirrus from layered mixed-phase clouds as explained above”. Above you mention in **page 3, line 15:** “We start with a case of a shallow mixed-phase cloud consisting of a liquid-water layer on top of the ice virga zone” and in **page 6, line 6:** “...in the case of clouds, liquid-droplet layers show PLDR \approx 0 at layer base where light depolarizing multiple scattering is low, and PLDR of 0.4-0.6 in,

e.g., cirrus layers”, but you haven’t explained how you distinguish cirrus from layered mixed-phase clouds in depolarization measurements above. So I suggest to skip “as explained above ” or explain it.

Page 7, lines 22-25: “The light-absorption-related lifting occurs during the spread of the smoke over the respective hemisphere and continues as long as the smoke layers are optically dense enough (aerosol optical thickness $AOT > 1-2$ at 500 nm) with the consequence that the smoke reaches, e.g., the Central Arctic at heights up to 5-10 km above the tropopause”. Please provide a reference for these AOT required conditions.

Page 7, line 33: “The 532 nm lidar ratio is much larger than the 355 nm lidar ratio. ”: please quantify how larger it was.

Page 8, line 5: “These specific optical properties are linked to the narrow size distribution of absorbing smoke particles which form a well-defined accumulation mode as shown in Fig. 5”. Please provide references to support this claim. Moreover, discuss the role of the shape and refractive index of the smoke particles in defining the unique optical properties measured.

Page 8, line 15: “With increasing age the core structure obviously collapses, gets compact, and the particles become more and more spherical with time (Baars et al., 2019)”. Please revise obviously as probably.

Page 8, line 27: “Downward mixing and transport into the lower troposphere had no impact on the UTLS AOT as well”. Please explain this statement in more detail. How can this be supported, when the AODs observed are decreasing with time from 0.12 to < 0.03 during the time period discussed?

Page 9, line 7: “ But this smoke layer had no clear boundaries, at least no clear upper boundary (see Figure 4a)” and **page 8, line 30:** “The layer-mean 532 nm smoke extinction coefficients in Fig. 6c (obtained from the ratio of AOT divided by the layer geometrical depth in Fig. 6a)”. Please include a comment on the effect of the unclear layer’s boundary to the lidar retrievals presented in this work (e.g. the effect to the AOD and layer top heights).

Page 9, lines 1-10: “It is noteworthy to mention that the CALIPSO data base...aerosol observations and corroborate our hypothesis”. Please revise taking into consideration the Interactive comment of Jayanta Kar on the detection of the smoke layers from CALIPSO.

Page 9, line 21: “Note that we corrected our stratospheric smoke observations in Fig. 6 for PSC effects”. Please explain how.

Page 9, lines 33-34: “According to Vaughan et al. (2020), the volcanic aerosol layer consisting of sulfuric-acid-containing water droplets (75% H_2SO_4 , 25% H_2O) formed above the tropopause with maximum heights reaching 21 km”. In Vaughan et al. 2020 paper they reported that the volcano “...send a plume of ash and sulphur dioxide into the stratosphere...”, “the ash and sulphur dioxide plume initially moved westward before being entrained in a cyclonic circulation over the North Pacific” and “During the latter half of June and in early July, pyroconvection over Canada injected layers of smoke and ice clouds into the lower stratosphere (similar to the case described by Vaughan et al.

(2018)), making it difficult to distinguish the progression of volcanic ash remnants using the CALIOP profiles". Why do you not mention the ash contribution on these layers? As the smoke layers from these fires arrived in the MOSAiC altitudes and are discussed herein, why do you exclude possible mixture with the volcanic ash?

Page 9, line 34 - page 10, line 2: "From the 355 nm Raman lidar observations at Capel Dewi Atmospheric Observatory, United Kingdom (52.4°N, 4.1°W) it can be concluded that the 532 nm AOT was about 0.03 over UK in August 2019, of the order of 0.01 in December, and of around 0.005 during the first months of 2020, respectively". Please provide a reference for these AOTs.

Page 10, lines 10-14: "The original and primary goal of the shipborne MOSAiC lidar measurements was to provide, for the first time, a seasonally and height-resolved characterization of tropospheric aerosols and clouds for the North Pole region. Especially the coverage of the winter half year can be regarded as a valuable new contribution to Arctic aerosol research. 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)". As there are plenty of CALIPSO overpasses in the Arctic region, please rephrase this part including for example the specific latitudes of the campaign.

Page 10, line 22: "The measurements are representative for many days during the winter months". Please quantify the days arctic haze was observed in the period of the experiment.

Page 10, lines 23-24: "The most striking feature in both figures is that aerosol layers were detected everywhere up to the tropopause, and because of the smoke layer even from 8 to almost 20 km height". This is not visible in these figures which are up to 15 and 16 km. Please state if you refer to fig. 6, which shows smoke up to 17km during March and April, and smoke up to 20km in February.

Page 10, lines 27-28: "Type-Ia PSCs consist of nitric acid trihydrate (NAT) crystals and produce significant depolarization of backscattered laser light". Please rephrase to "...are thought to consist of ...".

Page 11, lines 6-7: "The Ångström exponent for the extinction coefficient was around 1.7 in the lofted layer above 3 km height. The lidar ratios were high with values close to 100 sr in the lofted layer on 4 March". Please provide the uncertainties or standard deviation of these values.

Page 11, lines 6-7: "The 532 nm AOT was 0.024 (4 February, for the lowest 307 km height) and 0.022 (4 March, for the lowest 5 km)...". Please provide the uncertainties/errors of these values.

Page 12, lines 29-30: "Favorable conditions with cloud top temperatures around -28.5C at 2.6 km height (at 03:00 UTC) were given for heterogeneous ice formation via immersion freezing, i.e., ice nucleation on INPs immersed in the water droplets". Please provide relevant reference for the "favorable conditions".

Page 12, lines 30-32: "After nucleation, the ice crystals grew fast to sizes of 50–100µm within minutes (Bailey and Hallett, 2012) and immediately started falling out of the altocumulus layer". Please clarify whether you refer to findings from Bailey and Hallett (2012) or to result from this specific case. If the

second appy, please provide a short description on the measurements/methods used for these findings.

Page 13, lines 3-4: “As discussed below in detail, there were always 20-200 droplets per cm³ in the altocumulus top layer, but only 0.1 to 1 ice crystals per liter”. Please rephrase so as to be clear that these concentrations are estimated and not measured.

Page 13, lines 12-14: “This condition holds here with ice crystal backscatter coefficients of 5-10 Mm⁻¹sr⁻¹ in the virga and thus also in the cloud top layer and droplet backscatter coefficients of the order of 700 Mm⁻¹sr⁻¹”. Please include the information that these values are not shown in the study.

Page 13, line 15: “As can be seen in seen Fig. 11, the CDNC values were about 20 cm⁻³ in the beginning and around 100 cm⁻³ later on”. Please rephrase to “estimated values” or “retrieved values”

Page 13, lines 16-18: “Obviously, updraft velocity was weak and correspondingly water super saturation levels were below 0.2% so that fewer particles were activated to become cloud droplets as predicted (CCNC values in Fig. 12). Later on, the updrafts became obviously stronger, and supersaturation levels exceeded 0.2% so that more CCN nucleated cloud droplets as predicted by the retrieved CCNC values”. Please rephrase or justify “obviously”.

Page 13, line 20: “The cloud extinction coefficient showed typical values from 10-20 km⁻¹ most of the time in the droplet-dominated cloud layer”. The plot shows cloud extinction coefficient values <10 km⁻¹ from 7:45 until the end of the cloud (~3hrs) and values between 10-20 km⁻¹ for ~2.5hrs. Please rephrase accordingly.

Page 13, line 30: “Soot and mineral dust particles are good candidates to serve as INPs”. Please provide relevant references (i.e. Sassen and Khvorostyanov, 2008; Boose et al., 2016; 2019).

Page 13, lines 31-33: “Dust is left as potential INP. Our polarization lidar observations indicated the presence of a dust fraction of 3-10% according to the slightly enhanced particle depolarization ratio (not shown) above 2 km height...” Earlier, in page 9, you mentioned the influence of aerosol particles from “a strong eruption of the Raikoke volcano in the Kuril Islands...”. Why do you exclude the possibility of the presence of ash non-spherical particles? Please discuss the effect of possible ash particle presence on the INP retrievals you provide.

Page 14, lines 1-2: “We used the INP parameterization scheme of DeMott et al. (2010) to estimate the dust INP concentration for immersion freezing. Here, the particle number concentration n₂₅₀ 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 n₂₅₀ profil”. Please discuss why you use the INP parameterization scheme of DeMott et al. (2010), which was developed with minimum presence of dust particles in the analysed samples, to convert the lidar derived dust n₂₅₀ concentrations to INP, instead of the dust-tailored INP parameterization scheme of DeMott et al. (2015). Can the authors comment on the effect of the dust-tailored parameterization to the lidar-derived INPC in the study?

Page 14, line 17: “until a dry air mass approached, leading to a strong decrease in relative humidity and dissolution of the stratiform cloud deck”: Please support this discussion with updraft and wind data, if available in MOSAiC. Moreover, please provide a colorbar with more points so that it is easier for the reader to understand which values you are referring to.

Page 14, line 19: “ice crystals could partly reached the ground as precipitation”. for convenience to the readers, you could refer to figure 3f, where this is evident.

Page 14 line 20: “The good match between CCNC and CDNC (liquid-water cloud closure) and between INPC and ICNC”. Please rephrase as “the good match between the estimated CCNC..” or “the good match between the retrieved CCNC..”.

Page 14, lines 32-33: “Organic aerosol (OA, the main aerosol component in wildfire aerosol) is besides dust and marine particles ubiquitous in the atmosphere”. Please comment on the presence of ash particles, as mentioned in previous comments.

Page 15 lines 20-25: “HYSPLIT backward trajectory analysis ... During the 7-day travel in the Arctic the Pacific airmass mixed with the smoke above 7 km”. Consider including a figure showing the trajectories discussed, even in an appendix.

Page 16, lines 13-15: “Ice supersaturation conditions are usually given or produced during updrafts (e.g., during the ascending period of a gravity wave) that could, in principle, be detected and measured with the AMF-1 Doppler radar”. Is this information available for the case discussed here?

Page 17, line 2: “600 s may represent here a typical time period of the lifting phase of a gravity wave”. Please provide a reference on this, or discuss if these updrafts were actually observed during MOSAiC.

Page 17, lines 2-5 and lines 13-16: “As can be seen the n_{ICE} and $n_{INP,I}$ values (blue and red bars) are in the same range of values which suggests that organic particles may be able to control the evolution of the cirrus layer via the immersion freezing mode ... The impact of deposition $n_{INP,D}$ (cyan and orange bars) is comparably weak in this example... However, the successful closure, indicated by a reasonable match between $n_{INP,I}$ and n_{ICE} , indicates that the wildfire smoke was able to trigger cirrus formation (before homogeneous freezing can take place on stratospheric background or even liquid smoke particles) and control of the further evolution of the ice cloud system”. The authors should consider the possibility that the discussed cloud was formed in an aerosol richer environment. As they show in fig. 6, all the dust plumes observed in the period between 1/10 to 30/11, and for altitudes up to 11 km, had extinction coefficient values $>10 \text{ Mm}^{-1}$. Also, indicatively, in Fig. 7 and Fig. 10, the aerosol profiles show that the extinction coefficient values in the middle of the layers were respectively 5 and 8 times higher than the values in the edges of the layers. It would be good if the authors take into consideration that the $n_{INP,D}$ abundance could have been significantly higher in the time of the formation of the cloud, and discuss how this affect their conclusion that the immersion freezing process with uplifting support from a gravity wave is the main driver on the n_{ice} observed in this case.

Figure 5: The size distribution retrievals in Fig. 5 need further support with a) the provision of the corresponding retrieval errors (as these are provided in e.g. Veselovskii et al. (2012)) and b) the comparison with other studies for stratospheric smoke. The effect of the (quite) low AODs on the retrieval errors should be also discussed.

Figure 6: Please provide the products uncertainties in figures 6b and 6c. Also, I would suggest changing legend “col. mass” in fig. 6b to “col. mass conc.”, and legend “Mass conc” to “Vert. mean. mass conc.” for completeness.

Figure 7: “The 532 nm backscatter ratio (total-to-Rayleigh backscatter) peaks at 2.43”. This parameter is not presented in the plot. Please include it or this give more information on this sentence to be in the context of the figure.

Figure 7: “. PSC optical thickness was 0.0125 at 532 nm(computed from backscatter values multiplied by a lidar ratio of 50 sr”. Please explain in the relevant section (in page 9) why you chose this LR value (e.g. Ohneiser et al., (2021) paper).

Figure 8: The VDR quicklooks of these cases would be of interest to the reader. Additionally, the addition of the backscatter profiles from 0-15km, which can support the argument of the authors that “There were no regions with a negligible aerosol content” in **page 10, line 24.**

Figure 13: “The range-corrected 1064 nm signal in (a) is biased by an detector overload in the nearest height range to the lidar”. Include the height you are referring to.

Figure 13:In Fig. 13b (as well as in Fig. 3f) the lidar signal above the cloud seems to be totally attenuated. Please provide additionally the collocated radar measurements to show the extent of the whole cloud and support the cloud top provided from the lidar.

Figure 15: To simplify the plot and make it more clear for the reader, the authors could consider moving the n_{ice} numbers to fig. 15c, where the profiles of these values are plotted.

Figure 15: “derived ranges of INP number concentrations $n_{INP,I}$ (for immersion freezing, blue and red, indicating the respective cirrus layers in (a)”. Please revise this part to state clearly these n_{INP} concentrations from which profiles/days are derived.

Technical corrections:

Page 1, line 2-5: Long sentence. Consider splitting it to two.

Page 5, line 22: “procedures, and to”: skip and

Page 6, line 3-8: “The linear depolarization ratio is defined as the cross-polarized-to-co-polarized backscatter ratio and allows us to sensitively distinguish spherical particles showing particle linear depolarization ratios (PLDR) close to zero from non-spherical aerosol particles showing PLDR values of typically 0.1-0.3. In the case of clouds, liquid-droplet layers show $PLDR \approx 0$ at layer base where light depolarizing multiple scattering is low, and PLDR of 0.4-0.6 in, e.g., cirrus layers. “Co” and “cross”

denote the planes of polarization parallel and orthogonal to the plane of linear polarization of the transmitted laser pulses, respectively”. It will be better read if the “co” and “cross” definitions are closer to the parameter “cross-polarized-to-co-polarized backscatter ratio”.

Page 7, line 11: “In summer, warm, moist and polluted air massed..”. Typo: masses.

Page 7, line 31: “The internal vertical structures were rather smooth and indicate an aged smoke layer”: change “and” to “which”.

Page 8, line 16: “well established and obviously prohibited any mixing”. Please delete “obviously”.

Page 13, line 13: “As can be seen in seen Fig. 11”. seen duplicate.

Figure 13: “... (a) is biased by an detector overload”. Typo: a.

Page 14, lines 32-33: “...(OA, the main aerosol component..). Typo: aerosol .

Page 18, line 1: “As an outlook”. The authors could consider to revise this to “future work”.

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Petters, M.D., Parsons, M.T., Prenni, A.J., Demott, P.J., Kreidenweis, S.M., Carrico, C.M., Sullivan, A.P., McMeeking, G.R., Levin, E., Wold, C.E., Collett Jr., J.L., Moosmüller, H.: Ice nuclei emissions from biomass burning (2009) *Journal of Geophysical Research Atmospheres*, 114 (7), art. no. D07209, DOI: 10.1029/2008JD011532

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