Reply to referee #2: Interactive comment on "The contribution of Saharan dust to the ice nucleating particle concentrations at the High Altitude Station Jungfraujoch (3580 m a.s.l.), Switzerland" by Cyril Brunner et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-643-RC2, 2021.

Reviewer comments are reproduced in **bold** and author responses in normal typeface; extracts from the original manuscript are presented in *red italic*, and from the revised manuscript in *blue italic*.

The authors conduct measurements of INP from February 2020 to December 2020 at JFJ. The INP measurements are constrained to a temperature of 243K and a saturation ratio of 1.04. They classify the INP according to whether or not Saharan dust events were present. The classification is based on four criteria: single scattering albedo; satellite retrievals of dust mass concentration; modelled tropospheric residence times; the backscatter signal from a ceilometer. 14 dust events of high confidence (hcSDEs) were classified where each of the four criteria for Saharan dust were met and 12 events of lower confidence (lcSDEs) were classified where at least one of the four criteria were met. The authors show that INP concentrations increase by generally one to two orders of magnitude during the periods of dust events. They also find some evidence for dust influence in the absence of an identified event. I find the main aspects of the work to be sound and useful. I think the interpretation of the results, initially sound, is carried a bit farther than warranted, reflected in my comments 12-17 below. Overall, I feel the paper could be suitable for publication subject to some revisions.

We would like to thank the reviewer for their valuable comments and address the comments individually below.

Lines 3-4: These two sentences might be better reversed in order, as I find the second seems to contradict the first.

We agree with the reviewer, and changed lines 3-5 (revised manuscript) from the abstract as follows, addressing also the comment about line 4 (line 4 revised manuscript) from reviewer #1:

~~However, the extent of the abundance and distribution of INPs remains largely unknown.~~ Mineral dust has been found to be one of the most abundant INP in the atmosphere at temperatures colder than 258 K. However, the extent of the abundance and distribution of INPs remains largely unknown.

Lines 33-35: Again, slightly contradictory statements: Mixed phase exist between 273K and 235K, yet most clouds warmer than 253-258K are ice free.

We thank the reviewer for the comment. We changed line 33 (revised manuscript) as follows:

Mixed-phase clouds theoretically can exist between 273 K and ~ 235 K. Depending on the measurement location, in-situ measurements revealed that only approximately half of the clouds contain the liquid phase when at 253 to 258 K, while the warmer clouds are mostly ice free (e.g., Korolev et al., 2003; Verheggen et al., 2007; Kanitz et al., 2011).

Lines 41-59: I suggest mentioning the importance of mineral dust as INP before discussing the sources and transport of dust.

We acknowledge the reviewer's comments, however we feel it is better to first address the global distribution and ubiquity of mineral dust in the atmosphere before narrowing down to the INP impacts of mineral dust. As such, we leave the text in lines 41-61 (revised manuscript) the same as in the original manuscript.

Line 73: Remove "Besides".

We agree with the reviewer, and changed line 85 (revised manuscript) as proposed:

This allows to analyze whether all SDEs show an increased INP number concentration, as previous studies imply (Chou et al., 2011; Boose et al., 2016b; Lacher et al., 2018a). ~~Besides, o~~Our data indicate that signals from Light Detection and Ranging (LIDAR) ceilometers can be used to infer INP concentrations, as reported in other studies using depolarization channel LIDARs (Mamouri and Ansmann, 2015; Ansmann et al., 2019).

Line 250: What metric of particulate matter is recorded?

We were referring to mass concentration. We thank the reviewer for the comment, and changed line 292 (revised manuscript) as follows:

The mass concentration of particulate matter with an aerodynamic diameter below 2.5 µm (PM2.5) and below 10 µm (PM10) is continuously recorded with a white light optical aerosol spectrometer (Fidas 200, Palas GmbH, Germany).

Line 256: Please clarify what you mean by "and involved microphysics of dust".

We thank the reviewer for the comment, and changed lines 298-300 (revised manuscript) as follows:

To assess the atmospheric transport of dust and the presence and retrieved phase of nearby clouds in the case study (see subsection 3.3), post-processed lidar data from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instruments of the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellites was used.

Lines 264-270: Perhaps clarify that the median of all single events is the median of the individual event medians, and that the collective refers to the median of all INP concentrations during the SDEs.

We thank the reviewer for the comment, and changed lines 307-313 (revised manuscript) as proposed:

The median INP concentrations of the individual event medians was 15.3 ± 1.2 INP std L^{-1}. Analogously, the medians of all single event 25 %, ~~50 %,~~ and 75 % quartile INP concentrations were 9.1 ± 1.1, ~~15.3 ± 1.2,~~ and 32.4 ± 1.4 INP std L^{-1} (event-based; see Table 1, and Table A1 for more detail), generally one order of magnitude higher than the median INP concentration of 1.1 ± 1.0 INP std L−1 during periods without SDEs (non-SDE). All SDE quartiles except the 25 % quartile exceed the 95th percentile during non-SDE conditions. ~~Considering the collective INP concentration during all SDE periods, t~~The median of all INP concentrations during all SDEs combined increases to 22.5 ± 1.4 INP std L−1, rendering the longer dust events generally to contain more ice-active particles. This median concentration is consistent with previously reported values at the JFJ of 26.1 INP std L−1 (Lacher et al., 2018a).

Lines 277-279: They clearly differ based on the stated uncertainties, yet you say the difference is not significant. Please explain.

We thank the reviewer for the comment, and changed lines 322-325 (revised manuscript) as follows:

The observed INP concentrations during SDEs were lower in FT conditions compared to periods with BLI with median concentrations of 17.3 ± 1.1 and 23.7 ± 1.5 INP std L−1, respectively, however, they did not significantly differ, as the median INP concentrations of one class does not exceed the interquartile range INP concentrations of the other class and vice-versa.

Lines 297-298: With only 12 lcSDE cases, the statistics for these events cannot be strong, unless one criterion was dominant.

The statement on line 297-298 (original manuscript, now lines 343-344) is based on all 26 SDEs detected. I.e., we believe that increased INP concentrations can be detected as long as one of the tracers exhibits and SDE. While we are limited with how many SDE occurred during the year, and more statistics are of course always desirable, this is a substantial leap forward in the statistics of the number of events detected compared to previous studies where single field campaigns on the order of 2-6 weeks were used to inform the impact of SDEs on INP concentrations.

Lines 310-311: Maybe, you can't say this is true, or even implied, without some sort of chemical ID.

We agree, and changed lines 356-357 (revised manuscript) as proposed:

This could mean that the kind of INPs detected during SDE also contributes to the overall INP population during non-SDE periods, but chemical analysis would be necessary to categorically conclude this.

Figure 6b – You mention having normalized the area under each curve. I suggest adding that this does not allow the "SDE" plus "no SDE" curves to equate to "all".

We agree with the reviewer, and changed Figure 6 caption as proposed:

Figure 6. Frequency distributions of INP concentrations (a); and dustCAMS (b) between February 7 and December 31, 2020 (solid black), for all classified SDEs (lcSDE and hcSDE, green) and for periods without SDEs (pink). A log-normal curve with stated curve parameter in (a) has been fitted to the frequency distribution of all INP concentrations (dashed blue). The area under each frequency distribution is normalized to unity, which does not allow the sum of the areas below the SDE and non-SDE frequency distributions to be equal to the area below the frequency distribution of all classified SDEs.

Lines 334-339: This a simple back-of-the-envelope calculation. The inclusion of uncertainties of 0.1 % and 0.2 % suggests otherwise and I think is inappropriate. I suggest reducing this discussion to something like "With our assumptions, we estimate that about 23 % of the INPs measured during non-SDE periods were dust-related.".

We agree with the reviewer, and changed lines 396-401 (revised manuscript) as proposed:

If 76.5 % of the dust is responsible for 74.2±0.2 % of the INPs (74.7±0.2 % - 0.5 %), and 23.5 % of all dust$_{CAMS}$ was advected to the JFJ during non-SDE periods, we estimate that about 23 % of the INPs measured during non-SDE periods were dust-related with our assumption that ~~this dust would proportionally contribute 74.2 / 76.5 × 23.5 % = 22.8±0.1 % of INPs during non SDE measured at the JFJ, assuming~~ a constant mass fraction of dust acts as INPs. Therefore, the total contribution of dust to the INP population measured at the JFJ at T = 243 K and saturation ratio of Sw = 1.04 is estimated to be 74~~.2±0.2~~ % + 23 ~~22.8±0.1~~ % ≈ 97~~.0±0.3~~ %. Note, during non-SDE periods, dust contributed 23 / 25.3 % ≈ 91 % ~~22.8 / 25.3 % = 90.1 %~~ to the overall INP population.

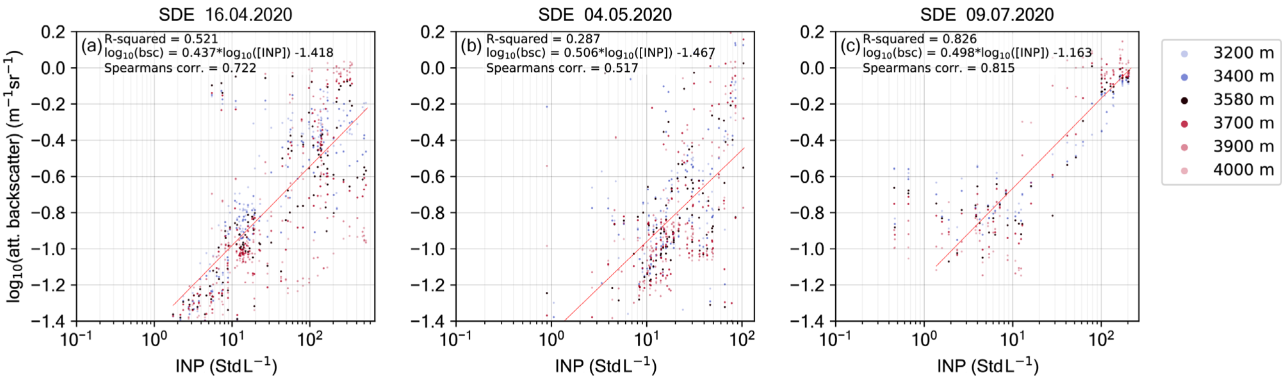
Lines 351-363 - The potential for correlation of INP with dustcams is based on Figure 6 showing consistency between the INP and dustcams distribution. However, the dustcams distribution for the SDE cases does not exhibit a log normal, which suggests that the sizes of dust particles vary, perhaps substantially. One consequence of that is the number concentrations of dust particles will not necessarily scale with dust mass. The authors note that INP scales with r^2, but that is largely related to the process of ice nucleation. Actual INP concentrations likely also scale with simply r or just the number of viable dust particles. Significant improvement of this discussion is needed.

We now clarify the discussion to better reflect the reviewer concerns as follows (lines 421-427 revised manuscript), addressing also the comment about lines 355-356 (lines 421-424 revised manuscript) from reviewer #1:

This does not come as a surprise, as the INP concentration is a particle number concentration per volume of air that tends to scale with particle surface area ($\propto r^2$) for an identical INP type or air mass dominated by a certain INP species, or with the number concentration of viable dust particles, while $dust_{CAMS}$ provides a mass concentration ($\propto r^3$). Given the distribution of the dust particles during the SDEs is not log-normal (see Figure 6b) suggests that the size of dust particles varies, and thus, the number concentrations will not scale with dust mass. Therefore, dustCAMS is also compared to PM10, which both are in units of mass per volume of air.

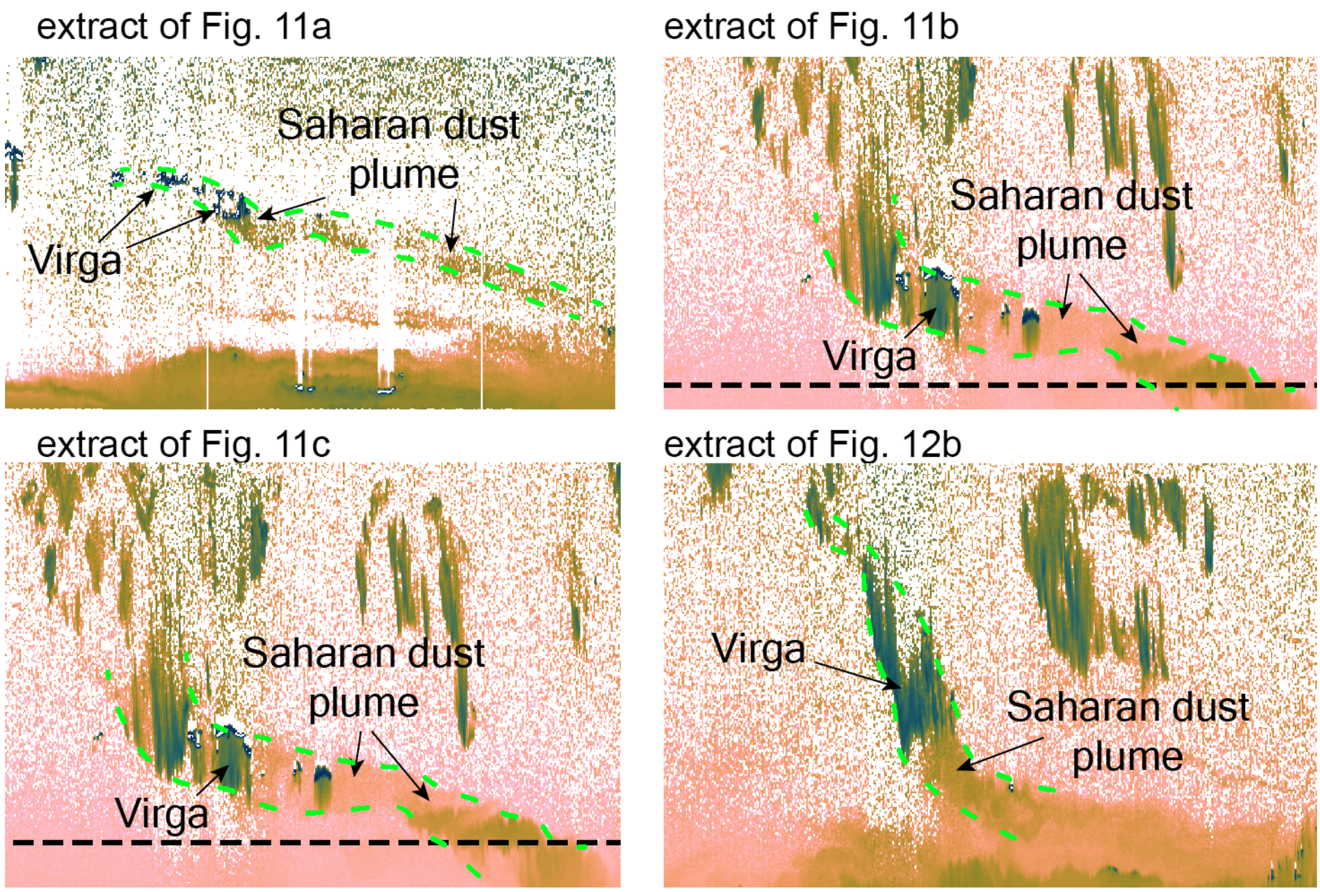
Line 378 and Figure 9 – The "R^2" in Figure 9c looks to be 0.826. Is that correct?

We thank the reviewer for the question. Indeed, R^2 in Figure 9c is 0.826. We increased the font size of the in-figure text to improve readability. However, we note that what is referred to in line 378 (now line 448) is not the R^2 but the Spearman's rank correlation coefficients.

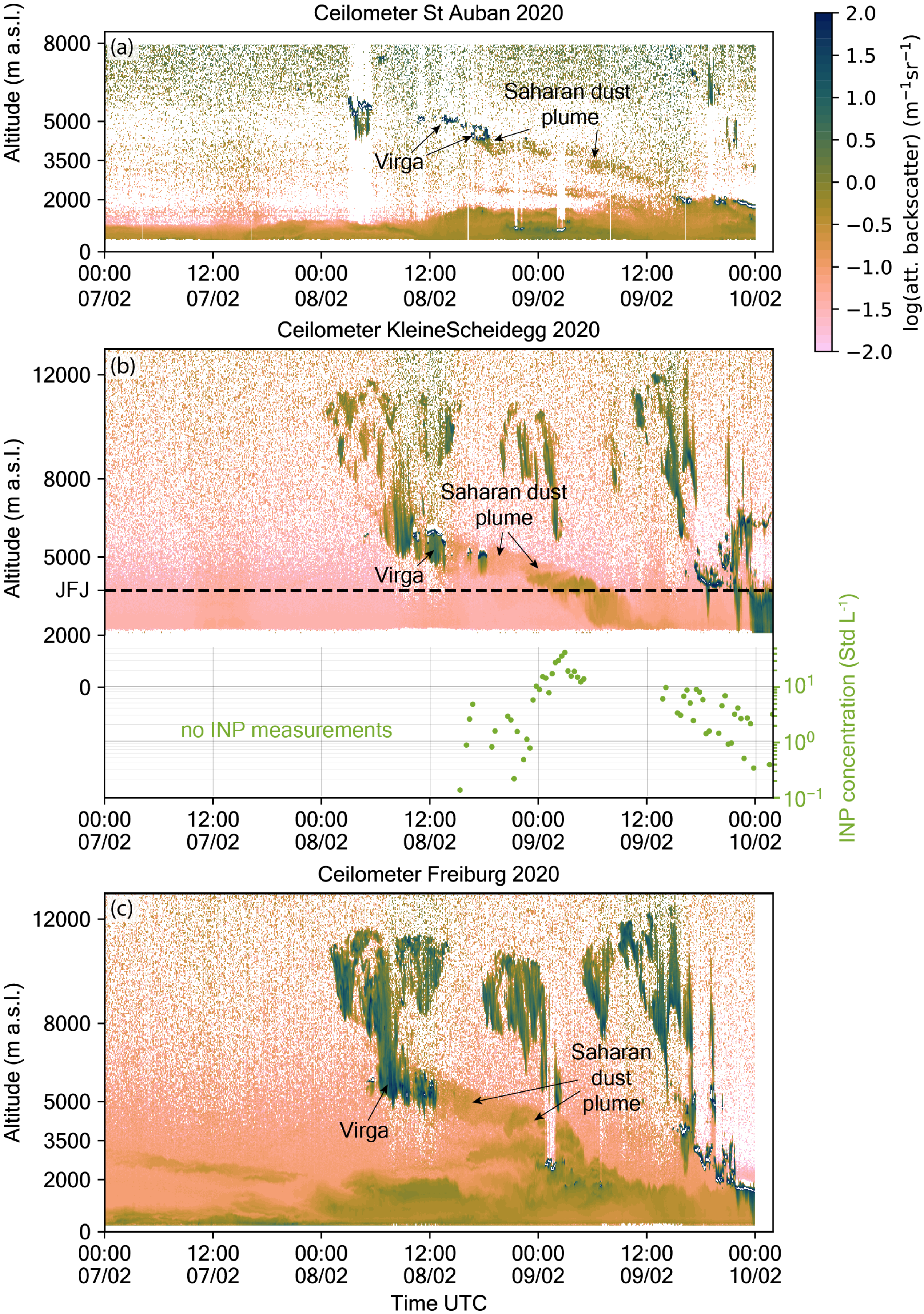


Lines 393-394: Please explain the evidence for a connection between the dust and the virga, which to me looks tenuous at best.

As stated in Line 414, we only formulate the hypothesis, that the dust are residual particles after the sublimation of the virga, and state in lines 431-434 that the hypothesis remains to be proven in future work. Thus, we do not present any evidence, but rather want to share our observations, that in two case studies Saharan dust plumes, which showed increased INP concentrations compared to background concentrations, are collocated with the low altitude part of virga with higher attenuated backscatter signals than the Saharan dust plumes, as illustrated in Figures 11 and 12. Here extracts from the Figures, where the green dashed lines indicate the collocation of the boundaries of the virga and the Saharan dust plumes:



Given that we found a visually similar pattern for various ceilometer locations, we do not assess this to be an artefact or chance, but an interesting pattern worth sharing. To further aid the reader, we marked the virga and Saharan dust plumes in Figures 11 and 12:



Ceilometer St Auban 2020
(a)
Saharan dust plume
Virga
Altitude (m a.s.l.)
8000
5000
3500
2000
0
00:00 07/02
12:00 07/02
00:00 08/02
12:00 08/02
00:00 09/02
12:00 09/02
00:00 10/02
log(att. backscatter) ($m^{-1}sr^{-1}$)
2.0
1.5
1.0
0.5
0.0
−0.5
−1.0
−1.5
−2.0
Ceilometer KleineScheidegg 2020
(b)
Saharan dust plume
Virga
12000
8000
5000
JFJ
2000
0
no INP measurements
INP concentration (Std L^{-1})
10^1
10^0
10^{-1}
Ceilometer Freiburg 2020
(c)
Saharan dust plume
Virga
12000
8000
5000
3500
2000
0
Time UTC

Figure 11

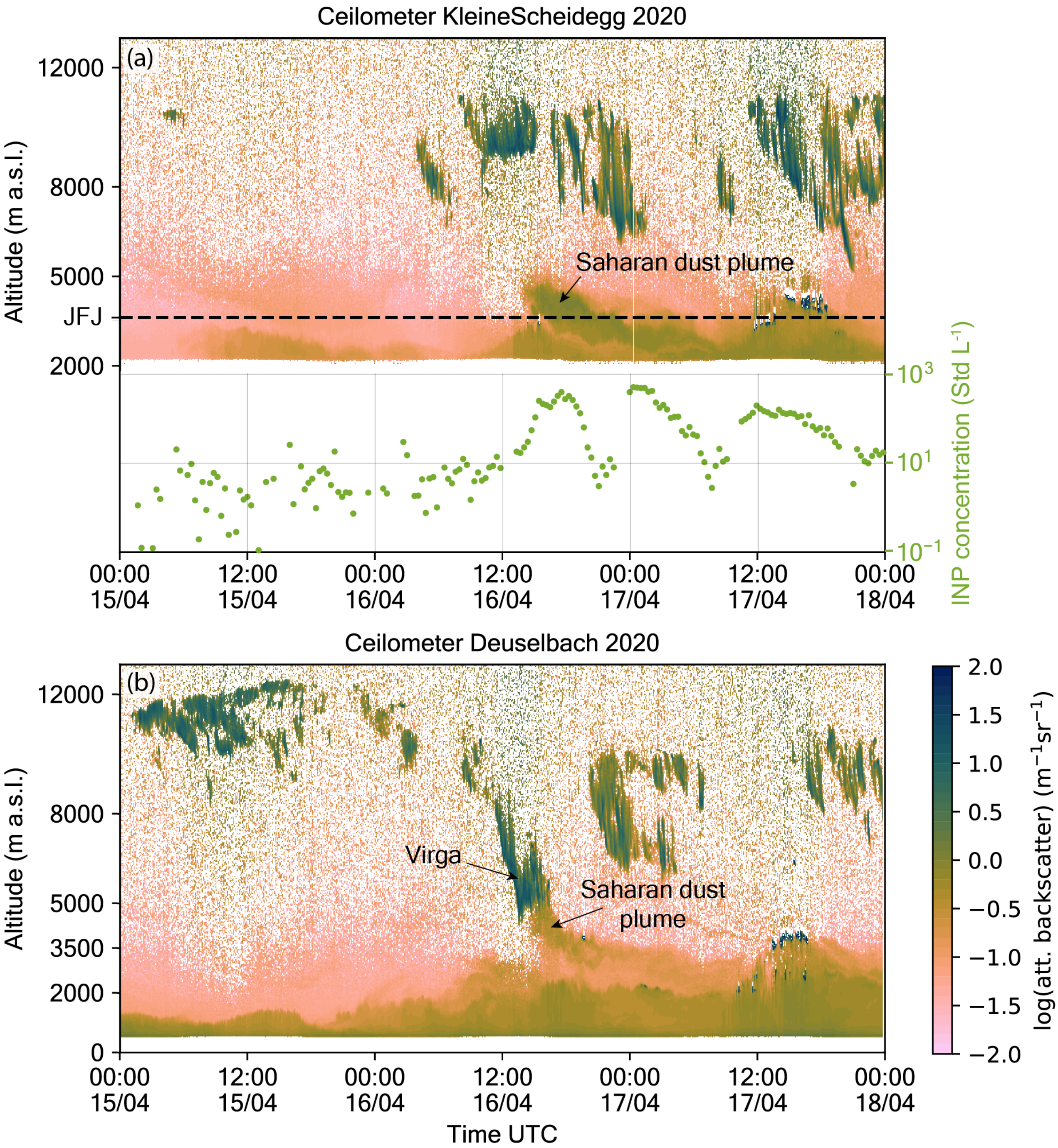


Figure 12

Are you suggesting that the dust is responsible for the virga or that the dust may be modifying it?

We are hypothesizing that the dust could make up a substantial fraction of the responsible INPs, which formed the virga. I.e., that the dust is being transported within the virga.

Line 415: Where are the dust particles between 10200 m and 11200 m? It looks like noise above 8 km.

We agree with the reviewer that the attenuated backscatter only shows noise at the stated altitudes except for the stronger signal of the cirrus clouds. Our altitude statement in line 415 was referring to the relative humidity profile. To reduce the vagueness, we changed lines 485-490 (revised manuscript) as follows:

This raises the hypothesis, whether the ice-active particles ~~within the plume~~ nucleated the ice clouds at altitudes above 7790~~10200~~ m a.s.l., ~~reducing the ambient ice saturation ratio to Si ≈ 1.0 due to diffusional growth~~ with subsequent sedimentation of the ice crystals to below 7790 m a.s.l., where they sublimated leaving behind dry INPs. These INPs further sedimented, however, due to the lower mass, at a much smaller rate. Thus, signals of the plume appear in the ceilometer more elongated after sublimation (e.g., in FRE at 13:00 UTC on February 8) compared to prior in the virga. What opposes this hypothesis is the fact, that no Saharan dust plume can be observed in the ceilometer measurements at altitudes above 7790 m a.s.l.

To remain consistent, we changed lines 543-548 (revised manuscript) in the conclusions as follows:

We found examples of SDEs with upstream virga from altitudes above 8000 m a.s.l., which led to the hypothesis of INPs being transported ~~at these high altitudes~~ to the midlatitudes, where they nucleate ice at altitudes above 5500 m a.s.l. and sediment to lower altitudes where they sublimate in drier air ~~and sediment as ice crystals to dryer altitudes, sublimate~~, and act as INPs at these lower altitudes. This could have important implications, as these INPs can be pre-activated and/or were subjected to atmospheric processing during the freeze-thawing cycles. This hypothesis will be subject of a future study, as pre-activated INPs loose their pore ice in the heated and dried sampling lines used in this study.

Lines 420-442 and Figure 12: I don't see any evidence for lifting of the dust above 7 km at best.

We thank the reviewer for the comment. Indeed, there is no evidence of dust being lifted above 7 km within the ceilometer data. We changed lines 491-498 (revised manuscript) as follows (see also the second comment below for the other changes):

Figure 12 shows another example observed pattern ~~a plume connected to virga~~, with the ceilometers KSE and DEU. The simultaneous ceilometer and INP measurements at the KSE and at the JFJ, respectively, which are shown in Figure 12a, indicate that the Saharan dust plume on April 16-17 contains INP number concentrations above 100 INP std L^{-1}, however, no connection between the Saharan dust plume and a virga is apparent. The Saharan dust plume can be tracked north to other ceilometer locations, such as DEU, following the outline of the dust plume in Figure 10b. At DEU, shown in Figure 12b, the lower end of the virga and the onset of the dust plume are collocated as before in the case study from February 8. However, also in this case no signal of the Saharan dust plume is apparent above 5500 m a.s.l.

Why can't the "connection" be simply the virga settling until it hits the drier air containing the dust?

We thank the reviewer for the comment. We assess the statement plausible, however, why does SDE arrive at the same altitude as the virga? From the ceilometer time series, it is clearly an evolution of the virga rather than the arrival of a dust mass to the same altitude as the virga. Why do the boundaries match so well across the locations? We assess a coincidence to be rather unlikely. After all, we only try to find a hypothesis for what we observe in different events and at different locations.

The virga does not correlate with the dust, which suggests that, if the dust is involved, it may only modify the virga.

We agree that the virga in Fig. 12a shows no direct connection with the Saharan dust plume. Here, we wanted to indicate in Fig. 12a, that the Saharan dust plume shows high INP concentrations, and as indicated by CAMS, the same Saharan dust plume further north at DEU shows the same collocated lower end of a virga with the onset of the Saharan dust plume as in the first case study. We would like to emphasize that we do not mention any statistical analysis, such as correlation, but refer to the feature, that the lower end of the virga and the onset/top of the Saharan dust plume are collocated. We are only presenting the observed pattern and formulate a hypothesis, which needs to be proven or rejected in future studies, as mentioned in the manuscript in lines 431-434 (now lines 508-511).

We clarify, changed lines 491-498 (revised manuscript) as follows:

Figure 12 shows another example of the observed pattern ~~a plume connected to virga~~, with the ceilometers KSE and DEU. The simultaneous ceilometer and INP measurements at the KSE and at the JFJ, respectively, indicate in Figure 12a that the Saharan dust plume of the SDE of April 16-17 shows INP number concentrations above 100 INP std L^{-1}, however, no connection between the Saharan dust plume and a virga is apparent. The Saharan dust plume can be tracked north to other ceilometer locations, such as DEU, following the outline of the dust plume in Figure 10b. At DEU, shown in Figure 12b, the lower end of the virga and the onset of the dust plume are collocated as before in the case study from February 8. However, also in this case no signal of the Saharan dust plume is apparent above 5500 m a.s.l.

One might expect more and smaller ice crystals associated with more INP, which might result in a shorter fall streak. However, the shortest fall streaks appear to be farther from the dust and completely unconnected.

We support the statement, that more and smaller ice crystals would be expected for a higher INP concentration. However, we argue, that the vertical extent of fall streaks depends on the ambient relative humidity profile with respect to ice and not on only on the number of ice crystals. Our analysis presented supports that the sublimation of ice crystals within the virga coincides with the atmosphere getting dryer, while the ice crystals could maintain their size above given the relative humidity was at ice saturation or above.