Reply to referee #3: 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-RC3, 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*.

This study provides interesting insights into the linkage between mineral dusts from the Sahara and ice nucleation activity at low temperature at the Jungfraujoch station. By using a wide array of instruments, techniques, and model data the authors were able to assign Sahara dust events and link it to INP concentrations. Even though many assumptions were necessary for the data curation, the authors clearly stated the limitations of each technique. The authors further hypothesize possible dust transport pathways within the atmosphere. Generally, the article is well written, concise and the results are of general interest for the scientific community. The provided data will improve the knowledge of the field and are important for further model studies on clouds and climate. However, I suggest minor revisions before publication:

We would like to thank the reviewer for their comments and compliments on the manuscript, and address the comments individually below.

Can you explain in the introduction in more detail why you have chosen 243 K and Sw of 1.04 as the parameters for your measurements? I assume it is due to general mixed phase conditions. However, why is this particular temperature area so important for ice nucleation including mineral dust and not e.g. 250K? Please elucidate in more detail so a general reader can better follow the content.

We thank the reviewer for the comment, and changed lines 70-80 (revised manuscript) as follows:

In this work, we investigate and quantify the INP concentrations at 243 K and S_w = 1.04 (immersion freezing). These conditions were chosen to align with previous INP measurements at the JFJ between 2014 and 2017 (Boose et al. 2016a, Lacher et al., 2018a). Ice formation in stratiform mixed-phase clouds is frequently observed close to the cloud top where temperatures of 243 K are a common lower bound cloud top temperature of mixed-phase clouds in central Europe (e.g., Bühl et al., 2016). In addition, 243 K is the warmest temperature where the instrument's signal-to-noise ratio allows for statistically acceptable data analysis when the sampling site is located in a remote region such as mountain top stations or the Arctic without using an aerosol concentrator. There is an uncertainty of relative humidity and variation in the vertical position of the particles within the aerosol layer in the chamber (DeMott et al., 2015; Brunner and Kanji, 2021) amounting to S_w + 0.007 and - 0.009 and ± 1.11 K at 243 K and set S_w of 1.04. To ensure that the entire sample layer experiences S_w > 1.0, a nominal S_w = 1.04 was chosen. All INP concentrations were measured at the JFJ during all SDEs between February 7 and December 31, 2020.

Line 116: I think it would improve the manuscript and makes it clearer to the reader if you could explain the Single scattering albedo Angström exponent in more detail.

We thank the reviewer for the proposition, and added lines 131-135 (revised manuscript) as follows:

The single scattering albedo Ångström exponent (αSSA) is an indicator of aerosol optical properties, which change during the presence of SDEs. Collaud Coen et al. (2004) observed that the exponent of the single scattering albedo during SDEs decreases with wavelengths, which counteracts the usual increasing trend. This is a combined effect of the peculiar spectral dependence of Saharan dust complex refractive index and large particle size. αSSA is retrieved from a nephelometer (Airphoton, IN101) and an aethalometer (MAGEE scientific, AE33) according to Collaud Coen et al. (2004).

Line 203: The classification of air masses comes a little bit abrupt. Could you mention that at the of the introduction where you summarize the investigations? (just a suggestion)

We thank the reviewer for the proposition and added lines 81-85 (revised manuscript):

In this work, we investigate and quantify the INP concentrations at 243 K and Sw = 1.04 (immersion freezing) at the JFJ during all SDEs between February 7 and December 31, 2020. During this time, continuous high-resolution (20 minutes) online INP measurements were performed for the first time at the JFJ. Because the data are not tied to single field campaigns in active SDE seasons, it also includes ~~SDEs~~ measurements in seasons where SDEs are infrequent. 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). Furthermore, the classification of SDEs is based on four distinct tracers (see Section 2.2) and analyzed with regard to the type of air mass present at the site, i.e., free tropospheric air or boundary layer intrusions (see Section 2.4). 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). In contrast to Mamouri and Ansmann (2015) and Ansmann et al. (2019), the topographic setup of the present study allowed for the ceilometer to scan the same altitude that the INP concentrations were measured at. Estimating the INP concentrations from the ceilometer backscatter signals from all ceilometer stations across Europe allows us to (back-)track the aerosol masses with enhanced INP concentrations and look into their atmospheric pathway, which we demonstrate in a case study. Finally, the contribution of (Saharan) dust to the INP concentration is estimated.

Figure 4: The figure gives a good overview of the collected data to assign a dust event and compare it to INP concentrations. The caption reveals much information. However, I miss an explanation of the figure in the continuous text.

We thank the reviewer for the comment. We added the following explanations to lines 274-290 (revised manuscript):

Figure 4 shows an example of a SDE with all introduced tracers, the air mass type and the INP concentration. example of a SDE with all at KSE showed an increase from background levels to ~1 $m^{-1}sr^{-1}$ at altitudes similar to the JFJ after midnight on July 10, indicating the presence of an aerosol plume. At 11:00 UTC, the signal of the aerosol plume was attenuated by the low-level clouds during the remaining period of the plume event. α_{SSA} decreased to below zero with decreasing wavelength after 2:00 UTC on July 10 (Figure 4c). The signal becomes less separated with α_{SSA} above zero at 15:00 UTC and noisy after midnight of July 11, indicating the end of the SDE according to this tracer. $Dust_{CAMS}$ mass concentrations exceeded the threshold concentrations on July 9 at 23:00 UTC, as shown in Figure 4d, peaking at 3:00 UTC of July 10 with 19.6 µg m^{-3}, followed by a decay, until falling below the threshold at 20:00 UTC. FLEXPART particle surface residence times in Figure 4e indicate between 3:00 UTC on July 10 and 5:00 UTC on July 11 that the air mass is expected to have had ground contact over the Saharan domain. Following all four tracers showing a signal, the SDE was classified as hcSDE. The INP concentrations show an increase from background INP concentrations on July 9 at 23:00 UTC (Figure 4a), to concentrations above 200 INP std L^{-1}, followed by lower concentrations coinciding with α_{SSA} exceeding zero for one hour. After a brief, second increase in INP concentrations, a decline to background levels at midnight of July 7 followed. No tracer shows an identical onset and decline as observed in the INP concentrations, however, α_{SSA} was the closest. The BLI air masses were present during the SDE, with ^{222}Rn initially at FT levels at 4:00 UTC on July 10, followed by a rapid change to BLI levels. At the end of the SDE, N_{90} indicated FT conditions, while ^{222}Rn still pointed to BLI, resulting in the air mass being classified as BLI. ~~During this SDE, all tracers showed clear signals, however, with different start and end times. Note the increase in INP concentrations during the SDE.~~

The results point out that further characterizations are necessary in order to estimate the role of mineral dust and other INPs on microphysical processes in the atmosphere. You mentioned in the manuscript in one sentence (line 321) that you cannot draw any conclusions on the influence of biogenic coating.

However, I wonder whether this would be an important information? I think the manuscript would benefit if this would be discussed and commented in more detail.

We thank the reviewer for the valuable comment. We agree with the comment and changed lines 369-382 (revised manuscript) as follows, addressing also the comment about section 2 from reviewer #1 regarding the influence of arable dust:

Thus, 74.7±0.2 % of all INPs were detected during SDEs. Whether this is because of the ice nucleation activity of the mineral dust or because of biological surface features, as proposed by other studies (e.g., see O'Sullivan et al., 2016; Augustin-Bauditz et al., 2016), is outside of the scope of this study. Biological surface features on the dust particles as the predominant cause of the ice nucleation activity of the particles would have two main implications. Firstly, the underlying mechanism leading to ice nucleation might differ, as proteins and other macromolecules could induce the ice nucleation as compared to topological mineral surface features, such as cracks and pores. Secondly, it would raise the question of the source of the dust particles containing ice-active biological surface features to be potentially from dried lake beds in desert regions or agricultural regions that are not differentiated in this work. In addition, a contribution of arable dust cannot be ruled out, however, during the 26 SDEs the modelled FLEXPART particles surface residence times indicated in 24 cases that the air mass had robust surface contact in the Saharan Desert, and in the other two cases, they had weak surface contact. The median INP concentration during SDE periods with FT air masses is 17.3 INP std L^{-1}, while with BLI it is 23.7 INP std L^{-1}. If we assume arable dust to show a substantially larger signal within the well-mixed PBL than in the FT, we could attribute the difference between median INP concentrations in FT SDE periods and BLI SDE periods to be because of arable dust, which is 6.4 INP std L^{-1}. There were three SDEs detected with lower concentrations than 6.4 INP std L^{-1}, one with a signal in α_{SSA}, one without and one where the nephelometer was offline. If during SDE periods the median contribution of arable dust was 6.4 INP std L^{-1}, then this should be also similar during non-SDE periods. However, the median INP concentration during non-SDE periods was only 1.1 INP std L^{-1}. We do not see any indications why during SDEs the contribution of arable dust should be substantially larger than during non-SDE periods. If at all, we expect arable dust would contribute to BLI INP concentrations during non-SDE periods.

In addition, how could you improve future studies to target this issue?

We thank the reviewer for the comment. We added lines 401-406 (revised manuscript), which also address the comment of reviewer #1 concerning lines 338-339:

Note, during non-SDE periods, dust contributed 23 / 25.3 % ≈ 91 % ~~22.8 / 25.3 % = 90.1 %~~ to the overall INP population. To validate the stated contribution in future studies or investigate the presence of biological surface features causing the ice-activity, we propose to separate INPs from the bulk aerosol population to analyze the chemical composition of the INPs as well as study the surface using scanning electron microscopy. However, to our knowledge, such equipment to separate INPs has not been used in a continuous annual study. Detailed suggestions of how this can be achieved in a long-term automated study by modifying HINC-Auto is presented elsewhere (Brunner 2021), and is beyond the scope of the current manuscript.

Line 82: Jungfraujoch (JFJ) – abbreviation was already introduced above.

We thank the reviewer for catching that, and changed line 95 (revised manuscript) as follows:

The Sphinx observatory at the JFJ ~~Jungfraujoch (JFJ)~~ is located on a saddle between Mt. Mönch and Mt. Jungfrau in the Swiss alps (46.330° N, 7.590° E) at an altitude of 3580 m a.s.l. (see Fig. 1).

Line 88: space is missing between the number and the unit (check throughout the whole paper).

Thank you for catching that. We have now included a space between the number and percent symbol throughout the manuscript.

Line 89: m/s or m s^{-1} – only use one style

We thank the reviewer for catching this, and changed line 101 (revised manuscript) as follows:

The principal local wind directions between February and December 2020 were 320° (NW) for 62 %, and 150° (SE) for 27 % of the time, while calm wind situations below 1 m s^{-1} had a frequency of 11 %.

Line 119: […] more 'than' 6 consecutive […]

We thank the reviewer for catching this, and changed line 136 (revised manuscript) as follows:

A SDE is detected if the α_{SSA} is negative for more than 6 consecutive hours.

Line 119: This is longer than the 'previously' […] (?)

Thanks for catching that. We changed line 136 (revised manuscript) as proposed:

This is longer than the previously used 4 hours in Collaud Coen et al. (2004) in order to decrease the number of false or suspicious signals due to construction work at the JFJ (see below).

Line 127: […] MERRA-2 'with' […]

Thanks for catching that. We changed line 144 (revised manuscript) as proposed:

Gueymard and Yang (2020) performed a worldwide validation of the aerosol optical depth and Ånsgtröm exponent from CAMS and MERRA-2 with ground-based AERONET stations over the period 2003–2017.

Line 135/136: meters were abbreviated before

We thank the reviewer for the comment, and changed lines 152-155(revised manuscript) as follows:

For the classification of SDEs we used the dust reanalysis data from CAMS ($dust_{CAMS}$) at 1000 m above the surface in hourly resolution and units of µg m^{-3}, accessed via the Copernicus Atmosphere Data Store. 1000 m above surface was chosen to be closest to the real altitude of the JFJ and accounting for the smoothed surface elevations in the model domain due to the coarse grid spacing.

Line 227: introduce the abbreviations and delete them below (line 231/232); Carbon dioxide (CO) and reactive nitrogen (NOy)

We thank the reviewer for the comment, and changed lines 254-260 (revised manuscript) as proposed:

A further tracer of BLI is the ratio of total reactive nitrogen (NO_y) to carbon monoxide (CO) according to Zanis et al. (2007). Both NO_y and CO are emitted from anthropogenic sources, however, NO_y reacts and decays on the order of days, while CO can be considered inert within the same time period. Consequently, the ratio of NO_y to CO decreases with increasing ageing time, leading to smaller ratios found in the FT compared to BLI (Zanis et al., 2007). ~~Carbon monoxide (CO)~~ CO is continuously measured with a Cavity Ringdown Spectrometer at the JFJ (Zellweger et al., 2019). ~~Total reactive nitrogen (NOy)~~ NO_y was measured *until March 2020 with a chemiluminescence detector after conversion to NO on a heated gold catalyst (573 K) in the presence of CO as a reducing agent (Pandey Deolal et al., 2012).*

Figure 4 caption: superscript '222'-Radon

Thanks for catching that. We changed Figure 4 caption as proposed:

(f) The probability for free tropospheric (FT) or boundary layer intruded (BLI) air mass to be sampled at the JFJ for the left hand side factor of equation (1), dependent on the ^{222}Radon concentrations (^{222}Rn, blue line) and the right hand side factor, dependent on the particle concentrations with a mobility diameter larger than 90 nm (N_{90}, red line) as well as their product, the probability of the sampled air to be of free tropospheric origin (PFT, black dashed line).

Line 253: '85°' is missing a degree sign

Thanks for catching that. We changed line 295 (revised manuscript) as proposed:

The spectrometer measures the intensity of single particles at an angle between 85° and 95° and infers the particle diameter solving the inverse Mie problem.

Table 1: All INP 'concentrations' missing the letter s in the end

Thanks for catching that. We changed Table 1 caption as proposed:

All INP concentrations are in units of INP std L^{-1}.

Line 302: Figure or Fig.? chose one style

We thank the reviewer for the comment, and changed line 350 (revised manuscript) as follows:

[…] (blue dashed line in Figure 6a; […]

Line 349: I guess at the end of the sentence it should be 'can'?

Indeed. We thank the reviewer for catching that and changed lines 416 (revised manuscript) as proposed:

Also, at measurement locations further away from the dust source than the JFJ is, or locations closer to local sources in the PBL, the contribution of dust on the total INP population can be expected to be much smaller.

References

Boose, Y., Kanji, Z. A., Kohn, M., Sierau, B., Zipori, A., Crawford, I., Lloyd, G., Bukowiecki, N., Herrmann, E., Kupiszewski, P., Steinbacher, M., and Lohmann, U.: Ice Nucleating Particle Measurements at 241 K during Winter Months at 3580 m MSL in the Swiss Alps, Journal of the Atmospheric Sciences, 73, 2203–2228, https://doi.org/10.1175/JAS-D-15-0236.1, 2016a.

Bühl, J., Seifert, P., Myagkov, A., & Ansmann, A.: Measuring ice- and liquid-water properties in mixed-phase cloud layers at the Leipzig Cloudnet station. Atmospheric Chemistry and Physics, 16(16), 10,609–10,620. https://doi.org/10.5194/acp-16-10609-2016, 2016.

Brunner, C.: Monitoring of Ice Nucleating Particles (INP) at the Jungfraujoch: Automation of the Horizontal Ice Nucleation Chamber for Continuous INP Monitoring (Doctoral dissertation), chapter 6.2.3, available from ETH Zurich research collection, doi: 10.3929/ethz-b-000493877, 2021.

Brunner, C. and Kanji, Z. A.: Continuous online monitoring of ice-nucleating particles: development of the automated Horizontal Ice Nucleation Chamber (HINC-Auto), Atmospheric Measurement Techniques, 14, 269–293, https://doi.org/10.5194/amt-14-269-2021, 2021.

DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., et al.: Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles. Atmospheric Chemistry and Physics, 15(1), 393–409. https://doi.org/10.5194/acp-15-393-2015, 2015.

Lacher, L., DeMott, P. J., Levin, E. J. T., Suski, K. J., Boose, Y., Zipori, A., Herrmann, E., Bukowiecki, N., Steinbacher, M., Gute, E., Abbatt, J. P., Lohmann, U., and Kanji, Z. A.: Background free-tropospheric ice nucleating particle concentrations at mixed-phase cloud conditions, Journal of Geophysical Research: Atmospheres, 123, 10,506–10,525, https://doi.org/10.1029/2018JD028338, 2018a.