

Reply to referee #1, Dr. Paul DeMott: Interactive comment on “The contribution of Saharan dust to the ice nucleating particle concentrations at the High Altitude Station Jungfrauoch (3580 m a.s.l.), Switzerland” by Cyril Brunner et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-643-RC1>, 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 is a very interesting study using the automated HINC instrument, which is packed with useful results and honest (refreshing) caveats, limited only perhaps by the assessment being made at a relatively low temperature (243K, for conditions that emphasize water droplet co-activation and expected immersion freezing). That is a modest critical comment, really, as it does not terribly limit the insights reflected in the deep analyses conducted. The study reveals important links between ice nucleation at lower temperatures and Saharan dust transports in this region, an apparently ubiquitous and strong influence, made further relevant by the observation that higher altitude dust appears to be drawn down via virga following likely heterogeneous ice formation at cirrus levels. The authors emphasize that this raises the potential issue of pre-activation and the known inability of most INP instrumentation for assessing this without special preconditioning of sampled air (needs more discussion). I wonder if this behavior does not also point to an important potential link between dust and precipitation in storms where deep clouds form over the region under dust transports. Precedence for this occurrence already exists in the literature over other regions that are more distant from major mineral dust sources. If so, transported dust INPs at high altitudes could more importantly impact precipitation (via ice sedimentation effects) than one might guess based on their characteristics of activating only at lower temperature, versus some other INP types that are imagined to have a greater impact on mixed-phase clouds due to their higher activation temperatures. I feel that a potential missed opportunity in this paper is relating the INP data more directly to aerosol size distribution measurements. An important relation to aerosol surface area is only inferred in the paper, and indirectly supported via the relation shown with attenuated backscatter. Closure with actual measured aerosol surface area to show relation to INPs could be useful (not required, just a suggestion). Presently it is only stated that some of the surface area (35 % estimate) is missed by the measurement limitations on capturing all sizes. That seems a minor amount to miss, something that I think should be emphasized as a positive aspect of the real-time, automated methods. I have only an assortment of other specific comments added to this, which I do below in order of appearance.

We would like to thank Dr. Paul DeMott for reviewing the manuscript, for the valuable comments and compliments and address the comments individually below.

Line 4: I suggest that this statement is not true in general, and so requires more generalization without attribution of a specific temperature. There are many reports of other influences on ice formation dominant at this temperature that are not only mineral dusts. For example, also soil dusts, which made me think about whether (or why not) this site does or does not see influence for more local/regional soil sources.

We agree with the reviewer, and changed lines 3-4 (revised manuscript) from the abstract:

Mineral dust has been found to be one of the most abundant INP in the atmosphere at temperatures colder than 258 K.

Line 8: I suggest to mention the method for derivation of Ångström exponent here (nephelometer), just as satellite retrieval is mentioned for mass concentrations.

We agree with the reviewer, and changed lines 8-10 (revised manuscript) as proposed:

Using the single scattering albedo Ångström exponent retrieved from a nephelometer and an aethalometer, satellite retrieved dust mass concentrations, simulated tropospheric residence times, and the attenuated

backscatter signal from a ceilometer as proxies, we detected 26 SDEs, which in total contributed to 17 % of the time span analyzed.

Line 15: Can you distinguish mineral from soil dust particles (due to attribution scheme for SDEs)?

No, we cannot distinguish the two types of dust with the sensing methods used in the current work. Thus, we changed lines 15-16 (revised manuscript) from “mineral dust” to “dust”, which is also consistent with the statement in the conclusions. Thank you for catching that.

We estimate 97 % of all INPs active in the immersion mode at 243 K $S_w = 1.04$ at the JFJ to be ~~mineral~~ dust particles.

Line 52: This is obviously not critical, but just to note that DeMott et al. (2003) was revised in 2009 to indicate a correction to maximum INP concentrations, reducing them to a few hundred per liter.

We thank the reviewer for the note, and changed lines 51-52 (revised manuscript) as follows:

In these studies, mineral dust showed the ability to be ice-active in the deposition or immersion mode with concentrations exceeding 500 to 1000 INP std L⁻¹ at 236.6 to 248 K (DeMott et al., 2003; DeMott et al., 2009; Bi et al., 2019).

Lines 52-55: I feel it is necessary to say that Zhao et al. was a modeling study, depending both on proper modeling of aerosols and parameterization of INPs.

We agree with the reviewer, and changed lines 53-57 (revised manuscript) as proposed:

Zhao et al. (2021) studied the global contributions to INP concentrations at 248 K using the Community Earth System Model version 2 (CESM2) and found dust over the terrestrial midlatitudes to be the dominating INP species by one to two orders of magnitude higher INP number concentrations at 248 K compared to marine organic aerosols. In general, the contribution of different INP species is expected to vary depending on the implementation of the INP parameterization in the model and aerosol representations in the model.

Lines 92-93: “There is no appreciable natural source of mineral dust in proximity of the site.” Is it true that there are no regional sources through the year of other dusts, such as agriculture?

We thank the reviewer for the comment. Given the remote location of the site in the Swiss Alps, the closest agricultural fields are 15 km to the north; ~3000 m lower in altitude and not used to grow crops (no plowing). Given the structure of the valley, the total area of the agricultural fields is very small. For all remaining directions, agricultural fields are further away. We added lines 105-106 (revised manuscript) as follows:

There is no appreciable natural source of mineral dust in proximity of the site. Potential local sources of arable dust are isolated agricultural fields 15 km north of the JFJ.

Lines 109-110:

“If the plumes appeared not to originate from within the Planetary boundary layer (PBL), the period for which the plume is observed between 3200 - 4000 m a.s.l. is marked as SDE.” Is it always so that origination in the boundary layer is so easily determined, for the entire history of a parcel?

We thank the reviewer for the excellent comment. No, it is not, also not in this case: the ceilometer does not reflect the history of an air parcel, but only the change of a passing aerosol plume above the measurement site over time. To reduce potential misinterpretation, we changed lines 123-124 (revised manuscript) as follows:

If the plumes appeared not to originate from within an aerosol plume was detected above the planetary boundary layer (PBL), the period for which the plume is observed between 3200 - 4000 m a.s.l. is marked as a SDE.

Section 2.2.2: Could you add to say a little more about the Ångström exponent calculation, and why a negative value clearly marks an SDE? I think it would help the general reader.

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

The single scattering albedo Ångström exponent (α_{SSA}) 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. α_{SSA} is retrieved from a nephelometer (Airphoton, IN101) and an aethalometer (MAGEE scientific, AE33) according to Collaud Coen et al. (2004).

Lines 174-179: This is an important discussion, perhaps for reasons that are not highlighted. Although there is size limitation on transmission of particles to the HINC, the losses of INPs are not so great. That is, if indeed mineral dust INPs scale with surface area, then a 35 % loss is not so significant in consideration that different INP measurement methods easily disagree at times by numeric factors up to 10. Hence, it would be great to see documentation of this statement with a size distribution typical of SDE periods. Not a requirement, but a suggestion. And could you also state if an upstream impactor is used for the HINC or not as part of INP detection?

We thank the reviewer for the important comment. We agree that the stated missed fraction of 35 % is low compared to other caveats. Thus, we changed lines 197-198 (revised manuscript):

This is a noteworthy limitation, as studies have reported a majority of INPs at $T > 248$ K to be supermicron particles (e.g., Mason et al., 2016; Creamean et al., 2018; Gong et al., 2020), and for a constant ice active fraction INP concentrations scale with total particle surface area for a given temperature (e.g., Connolly et al., 2009; Niemand et al., 2012). However, instrument comparisons typically report substantially larger discrepancies between individual instruments than 35 % (e.g., see Hiranuma et al., 2015; DeMott et al., 2018).

Furthermore, we add as proposed the following Figure to the appendix of the manuscript to line 551 (revised manuscript):

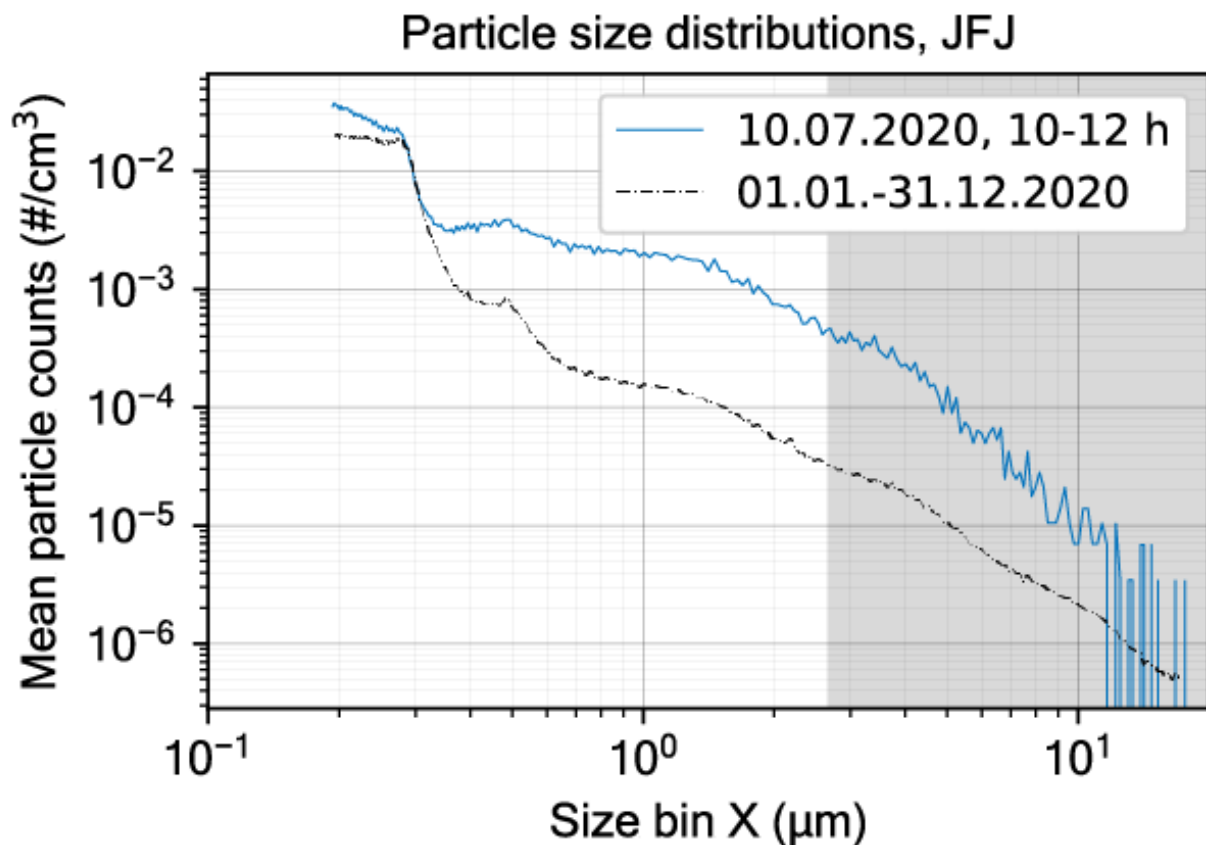


Figure A2: Mean particle size distributions at the JFJ during a SDE at 10:00-12:00 UTC on July 10, 2020 (blue line) and between January 1, and December 31, 2020 (black dashed line), both measured with an OAS. Only particles with diameters $< 2.5 \mu\text{m}$ are sampled by HINC-Auto due to the sampling line geometry and flow rates used. All particles $> 2.5 \mu\text{m}$ (gray shading) between February 7 and December 31, 2020 contribute during all SDE periods to 35 % to the overall particle surface area.

Furthermore, we changed lines 189-194 (revised manuscript) as follows to refer to the new Figure and comment on the lack of an impactor:

Due to the low volume flow of HINC-Auto and the sampling line geometry, particle survival measurements using an optical particle counter (MetOne GT-526S) and an OAS showed limitations when sampling particles $d > 2.5 \mu\text{m}$ (see Appendix A for detailed information), despite the fact that no impactor was used upstream of HINC-Auto. According to particle size distribution measurements from the OAS, particles with $d > 2.5 \mu\text{m}$ contributed during the investigated SDE periods on average to 35 % to the overall particle surface area (see Figure A2 for an example of particle size distributions during a SDE).

Section 2.3: The nature of the correction for frost background is unclear. Simple subtraction? Counts exceeding the 1 sigma value of the filtered air value? And in general, it would be useful to repeat how INPs are distinguished from other large particles in the HINC, especially during SDEs. I realize that an entire instrument paper was recently published, but some of these things bear repeating in short form.

We thank the reviewer for the comment on the nature of the correction for frost background. We changed lines 216-230 (revised manuscript) as follows:

False-positive counts can arise in HINC-Auto. Frost grows on the chamber walls, breaks off and is detected at the outlet as ice. This happens irrespective of whether ambient or particle-free air is sampled with HINC-Auto. Thus, to correct the measured INP concentrations, the number of frost particles is measured

separately and subtracted from the uncorrected INP measurements. This is done by sampling particle-free air for a period of 5 min before and after an ambient air measurement. During these periods, the number of false-positive frost particles are counted and subtracted time-proportional from the ambient air measurement in between. The recorded false-positive counts per unit time follow a Poisson distribution. Therefore, in a fraction of cases more or fewer false-positive counts per unit time are recorded during the particle-free measurements than during the ambient air measurement. This results in fluctuations of the measured INP concentrations even if the true atmospheric INP number concentration was to remain constant. The standard deviation of the resulting probability density function corresponds to the stated counting uncertainty of $\pm 1 \sigma$, which is provided with INP concentrations stated in the present work. This counting uncertainty is also considered to be the 1σ limit of detection (1σ -LOD) for a single data point. However, in the present work all background-corrected INP concentrations are retained, including positive values below the 1σ -LOD and negative values. This approach ensures that the random noise in background-corrected INP values caused by subtracting the mean frost particle counts does not introduce a systematic bias in mean or median values, which would occur if data below the 1σ -LOD were discarded.

To address how we distinguish INP from other large particles in HINC-Auto, we added lines 199-202 (revised manuscript):

INPs are detected if particles with an optical diameter of $\geq 4.0 \mu\text{m}$ are counted at the chamber exit. Only particles with a diameter $< 2.5 \mu\text{m}$ are sampled by the chamber, and the maximum expected size of a droplet that activated on a $2.5 \mu\text{m}$ particle is well below $4 \mu\text{m}$. Therefore, the method is robust as only ice crystals grow past the set size threshold of $4 \mu\text{m}$ in the set conditions. The frequency of the INP measurements is every 20 min (15 min sampling plus 5 min background), corresponding to $N = 19561$ measurements between February 7 and December 31, 2020. See Brunner and Kanji (2021) for more information on the sampling and derivation of the INP concentrations.

Figure 4: Would one infer from this figure that the SDE encompassed primarily boundary layer air transport? Again, I was confused because there was such an emphasis placed on indicating FT origin, and I was expecting that FT would encompass most SDE. Also, in this Figure caption and in the main manuscript text, I suggest writing out the meaning of hcSDE and lcSDE.

We thank the reviewer for the comment. SDEs are often transported in the FT, but then sediment into the PBL. We quantify the fraction of SDE periods at the site for which FT or BLI was present thus proposing that both scenarios are possible for SDEs. To clarify this, we added lines 318-322 (revised manuscript):

In former studies, SDEs were reported to be occurring in the FT only (Lacher et al., 2018a); however, our results indicate that FT conditions ($P_{FT} \geq 50\%$) made up 14.5% of the total SDE-time, compared to non-SDE periods, where FT conditions prevailed for 40.5% of the time. This A smaller FT fraction during SDE periods versus non-SDE periods is expected because of the seasonality of SDEs, with few events in winter, when also FT conditions prevail.

Furthermore, we added the meaning of hcSDE and lcSDE to the Figure caption as proposed:

Figure 4. An example period classified as high confidence Saharan dust event (hcSDE) between July 7 and July 11, 2020: ...

And accordingly to the caption of Figure 7:

Figure 7. Dust_{CAMS} mass concentrations with simultaneously measured INP number concentrations (a), and dust_{CAMS} mass concentrations with simultaneously measured PM₁₀ concentrations in (b), each for periods of high confidence Saharan dust events (hcSDEs), low confidence Saharan dust events (lcSDEs) and non-SDEs, which the corresponding correlation parameters and linear the fit (blue line).

Please note, I could not find anywhere else in the paper where these were specifically defined. That is, what makes for attributing hcSDE versus lcSDE?

The definition of high confidence Saharan dust event (hcSDE) and low confidence Saharan dust event (lcSDE) is provided in lines 98-100 of the original manuscript (now lines 113-114).

Line 250: Particulate matter means mass concentration? Or total counts in a certain size range?

We mean mass concentration. We thank the reviewer for the comment, and changed lines 292-293 (revised manuscript) as proposed:

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

Section 2 general comment: It seems like the CAMS, FLEXPART modeling and CALIOP data are the key to assuredly attributing dust to a Saharan source. That is, an aerosol layer in ceilometer data and Angstrom exponent will only indicate larger particles present from somewhere. I am curious if there were ever indications of arable dust sources reaching JFJ. It seems common in other locales, including mountaintop sites that are perhaps not so elevated. Unfortunately, I cannot offer a reference for that because the separate works I am aware of are still in preparation. It is simply an honest question about whether any such influences were inferred during the non-SDE periods or if any lcSDE events could have captured such.

We thank the reviewer for the very good question. We are not aware of any work at the JFJ quantifying the contribution of soil dust to the overall aerosol or dust population. We added we added lines 369-385 (revised manuscript) as follows, addressing also the comment within the results section from reviewer #3 regarding the influence of a biogenic coating:

Thus, $74.7 \pm 0.2\%$ 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 $17.3 \text{ INP std L}^{-1}$, while with BLI it is $23.7 \text{ INP std L}^{-1}$. 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 $6.4 \text{ INP std L}^{-1}$. There were three SDEs detected with lower concentrations than $6.4 \text{ INP std L}^{-1}$, 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 $6.4 \text{ INP std L}^{-1}$, then this should be also similar during non-SDE periods. However, the median INP concentration during non-SDE periods was only $1.1 \text{ INP std L}^{-1}$. 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.

Lines 338-339: I appreciate the strong qualifications added to the conclusion on the major role of mineral dust outside of SDEs for the conditions examined, but I wonder if somewhere here or in the conclusions you could speculate on what other types of measurements (or extensions) could be done to support the major role of mineral dust that you hypothesize here for this site (and by proxy perhaps, these altitudes over the region)? I say that because, in boundary layer measurements, my impression is that mineral dust cannot be assumed as the only influence at 243K, though I will not list the references.

We thank the reviewer for the constructive comment. We assess that separating the INPs from the aerosol population with subsequent chemical composition analysis can be a way to verify the stated hypothesis. We also have an idea of how to use one of the drawbacks of horizontal CFDCs to achieve the separation of INPs and CCN/remaining aerosols without the need of a virtual counter flow impactor. The idea is published in the Ph.D. thesis of Cyril Brunner, chapter 6.2.3. To elaborate, we added lines 401-406 (revised manuscript) as proposed, addressing also the comment within the results section from reviewer #3 regarding the detection of ice-active biological surface features:

Note, during non-SDE periods, dust contributed 23 / 25.3 % \approx 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 an 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.

Lines 355-356: Do natural INP concentrations scale with aerosol surface area?

No, they generally do not unless an air mass is dominated by a specific INP species. Only for if the INP type remains the same. We thank the reviewer for the comment, and changed lines 421-424 (revised manuscript), addressing also the comment about lines 351-363 (lines 421-427 revised manuscript) from reviewer #2:

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 dustCAMS provides a mass concentration ($\propto r^3$).

Line 434: Regarding these results apparently linking cirrus virga to descent of mineral dust plumes to levels where they can impact ice formation at lower levels, the mystery to me is why the virga connect to regions of apparent higher dust concentrations than ever seem to appear aloft.

We also were wondering the same but have no conclusive answer. Multiple studies state the top of the Saharan dust layer was at 6000 m a.s.l. and transport to higher altitudes seems improbable. Nonetheless, we detect that the plumes in the ceilometer show substantially elevated INP concentrations, and these plumes often connect to virga. The virga could also be spotted by eye as well at the time of the occasion.

Lines 440-442: You say as much in the abstract, but I expected a few more words here. It is possible I suppose to explore this topic in a laboratory setting, or by exposing instruments to the natural environment. I suspect that unreported efforts have already been attempted. It will be immensely more difficult to do this in an aircraft setting, where the actual nature of the inlet outside the aircraft would have to be modified in order to not heat particles due to pressure. In any case, I think you should expand the discussion a little bit, for the sake of those who may not have deeply considered how this can be done (or how hard it may be).

We agree with the reviewer, and changed line 519-526 (revised manuscript) as follows:

If the pre-activated particles are again exposed to ice supersaturated conditions, spontaneous ice crystal growth can occur at much lower S_i than with other INPs (David et al., 2019). To capture and study this effect, design adaptations need to be implemented in our sampling equipment in upcoming studies, as the pore ice sublimates or melts in the heated and dried sampling lines upstream of the INP counter used here. We attempted such a measurement of the number concentration of pre-activated particles in this way, using a supercooled diffusion dryer at the JFJ before directly sampling the ambient particles with HINC, in addition to sampling with a reference chamber with the standard heated inlet. Nevertheless, we were unable to reach any reliable conclusions. In part, due to the clogging of the inlet from frost build-up, the conditions along the trajectory of the potentially pre-activated particles had to be precisely adjusted, such that neither particle sedimentation due to activation, nor pore ice sublimation occurs. Furthermore, the set conditions do not affect all pore sizes similarly, and pore-ice can melt if the pore ice shrinks below the critical size of an ice embryo within the pore with a constrained width. In summary, there were too many parameters that needed to be simultaneously and meticulously controlled.

Lines 25: It is unusual to start a sentence with a number. Perhaps write it out.

Thanks for catching that. We changed line 25 (revised manuscript) as proposed:

Sixty-three \pm 7 % of global precipitation is initiated via the ice phase (Heymsfield et al., 2020), predominately over land and in the midlatitudes (Mülmenstädt et al., 2015).

Line 31: cover clouds → do clouds cover

Thanks for catching that. We changed line 31 (revised manuscript) as proposed:

Not only do clouds cover 68 % of Earth's surface...

Lines 69: Suggest to spell out 20 “minutes” for the INP measurement time resolution.

Thank you for the proposition. We changed line 80 (revised manuscript) accordingly:

During this time, continuous high-resolution (20 minutes) online INP measurements were performed for the first time at the JFJ.

Lines 90: hosts → has hosted

Thanks for catching that. We changed line 103 (revised manuscript) as proposed:

The JFJ has hosted long-term aerosol measurements for more than 30 years ...

Lines 119: previously is misspelled.

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).

Lines 127: with is misspelled.

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 Ångström exponent from CAMS and MERRA-2 with ground-based AERONET stations over the period 2003–2017.

Lines 272: where → were

Thanks for catching that. We changed line 316 (revised manuscript) as proposed:

..., and the highest concentrations measured were half as high as those measured in the Saharan Air Layer in Tenerife...

Lines 425: Probably do not need the word “for”.

Thanks for catching that. We changed line 502 (revised manuscript) as proposed:

The analysis showed ~~for~~ trajectories to originate from the Saharan desert within Algeria,...

Lines 461: Suggest rewrite, e.g., “and sediment to lower altitudes where they sublimate in drier air...”

Thanks for catching that. We changed line 545 (revised manuscript) as proposed, addressing also the comment of reviewer #2 about line 415 (line 485 in the revised manuscript):

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.~~

Lines 463: loose → lose

Thanks for catching that. We changed line 547 (revised manuscript) as proposed:

This hypothesis will be subject of a future study, as pre-activated INPs lose their pore ice in the heated and dried sampling lines used in this study.

References

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