

EGUSPHERE- 2025-3842:

“Aerosol Size Distribution and New Particle Formation in High Mountain Environments: A Comparative Study at Monte Cimone and Jungfraujoch GAW Stations”

Answers of the authors to Reviewer#1

While the reviewer’s comments are given in **bold red**, our answers are given below in black letters. Additionally, we added the changes made in the revised manuscript in *italic black text*.

The paper by Mazzini et al., investigates aerosol number size distributions (PNSD) and new particle formation (NPF) in the free troposphere (FT) using long-term measurements from two high-altitude observatories, Monte Cimone (2165 m, Italy) and Jungfraujoch (3580 m, Switzerland). The aim is to understand how aerosol formation, growth, and variability differ between the two sites. The results show that CMN is affected by more frequent and intense NPF and stronger PBL influence, while JFJ shows fewer NPF events, weaker growth despite more ultrafine particles. This is a good quality study that makes an important contribution to understanding aerosol formation in the free troposphere (FT). It is especially valuable because it contrasts two representative European high-altitude sites, applies three independent NPF classification methods and is based on a solid dataset consisting of two years of harmonized measurements. The paper also highlights how the condensation sink, cloud processes and boundary-layer strongly regulate NPF, providing a strong link between observations and their climate relevance.

The manuscript is well written and carefully developed. With the addition of some clarifications and details, it should be suitable for publication.

We would like to sincerely thank the reviewers for the time, effort, and thoughtful consideration given to our manuscript. We truly appreciate the positive assessment of our work and the constructive suggestions that have helped us improve the clarity, consistency, and overall quality of the paper. The feedback was extremely valuable and has been carefully addressed point-by-point below.

Specific comments:

Line 6: Sometimes “free troposphere (FT)” written in full, sometimes only “FT.” Suggest spell out first time and then “FT” for consistency.

Revised for consistency as suggested.

Line 29: “The frequency of NPF events vary widely” → should be “varies widely.”

Corrected to “*varies widely.*”

Line 59: “at 2165 m a.s.l.” double period. Should be “a.s.l.”

Corrected to “*a.s.l.*”

Line 63: the sentence “with this influence being most pronounced during summer daytime hours and diminishing at night, when the site predominantly reflects free tropospheric characteristics” is too long. I suggest splitting into two sentences.

The sentence was split into two shorter sentences for clarity. The revised text reads: *The observatory is considered representative of the Mediterranean and Southern European FT. However, it can be significantly affected by air masses from the PBL, particularly those originating in northern Italy and the highly polluted Po Valley. This influence is most pronounced during summer daytime hours and diminishes at night, when the site predominantly reflects free-tropospheric characteristics (Cristofanelli et al., 2018; Rinaldi et al., 2015).*

Line 63: “2.5–560nm” should have space: “2.5–560 nm.” Please check the entire document.

All instances have been checked and corrected.

Line 97: Datasets coverage: “31.2% at CMN... In contrast, summer months show notable data gaps”. Could you provide the percentage of the different seasons for clarity?

We now report the seasonal coverage directly in the paragraph (Section 2.2). The revised text reads: *Overall, both datasets provide a representative seasonal distribution, though with some variability in coverage. At CMN, data availability was highest in winter (31.2%), followed by autumn (28.6%), spring (21.6%), and summer (18.7%). At JFJ, the corresponding seasonal coverage was 35.9%, 28.5%, 14.4%, and 21.2%, respectively. The summer months show the largest data gaps at both sites (Fig. S1).*

Line 104: “Scaling factors were derived individually for each measured size distribution, and applied to all NAIS channels accordingly.” Please remove comma before “and.”

The comma before “and” has been removed. We also identified and corrected a few other instances in the manuscript, where a comma was unnecessarily placed after “and.”

Line 107: “a common range of 2.5–560nm” should be “2.5–560 nm” (space before nm). Please check the entire document.

Corrected throughout the manuscript.

Line 122: “originating from the nucleation of gas-phase precursors forming clusters approximately (~1–2nm)”; parentheses awkward, just write “forming ~1–2 nm clusters.”

Revised to: *Atmospheric NPF events are characterized by rapid bursts of particles in intermediate and nucleation mode, originating from the nucleation of gas-phase precursors forming ~1–2 nm clusters that subsequently grow into the Aitken mode.*

Line 149 and 150: Equation 2 formatting inconsistent: bracket notation $N_{[2.5-7]}$ inconsistent with earlier $N_{2.5-7}$. Please harmonize notation.

Harmonized using $N_{2.5-7}$.

Line 161-163: about RH threshold, you define out-of-cloud as $RH < 94\%$, in-cloud as $> 97\%$. But what about 94–97%? Please clarify explicitly.

Revised text:

Periods with RH between 94% and 97% were considered uncertain and, together with in-cloud periods ($RH > 97\%$), were excluded from the NPF analysis. However, all measurement periods, including in-cloud, out-of-cloud, and intermediate cases, were retained in the overall statistical analysis of the dataset to ensure full representativeness of atmospheric conditions.

Line 163: “Webcam images were used to further validate cloud presence”, could specify frequency or reliability (continuous, daily snapshots?).

Specified as follows:

At CMN, the cloud classification was further verified using webcam images with a 1-min time resolution, confirming station immersion within cloud layers during high-RH periods.

Line 171: “(770–3)” should be “(770 cm^{-3})”.

Corrected.

Line 171: “across four defined mode” should be “modes.”

Corrected.

Line 206: “These categories reflect distinct stages of aerosol.....These modes reflect distinct stages of particle growth and transformation”, the two repeated sentences are redundant. Please rephrase them

Rephrased as:

The particle number size distribution at CMN and JFJ was initially divided into four a priori modes (intermediate, nucleation, Aitken, accumulation), based on commonly used size ranges in aerosol studies, as described in the Data Analysis Sect 3.1. These modes represent distinct steps of aerosol evolution from freshly nucleated particles to aged particles that

efficiently scatter or absorb radiation and act as cloud-condensation nuclei (CCN), thereby influencing radiative forcing and cloud properties.

Line 210: In Table 2, confidence intervals are in parentheses but not explained in caption. Please clarify.

Specified in the caption:

Statistics of modal structure for hourly particle number size distributions. The table reports the median geometric mean diameter (d_{pg}) and the median geometric standard deviation (σ) and particle number concentration (N), with 25th–75th percentile ranges shown in parentheses, for each lognormal mode at CMN and JFJ.

Line 227: “These elevated ratios has been observed” should be “have been observed.”

Corrected following the suggestion.

Line 233: “One more possible explanation for the relative abundance...”, awkward phrasing, better “Another possible explanation...”

Corrected following the suggestion.

Line 243: Figure references: sometimes “Fig. X,” sometimes “Figure X.” Pick one style.

Harmonized to “Fig. X” throughout.

Line 265: “Ions also shows different behaviour between out-of- and in-cloud” should be “Ions also show different behaviour between out-of-cloud and in-cloud conditions”

Corrected following the suggestion.

Line 274: Table 3 should be harmonized (e.g., JFJ summer values missing \pm).

We thank both reviewers for noting the missing uncertainty values in Table 3 and the limited statistics for JFJ summer data. Indeed, the small number of valid summer observations at JFJ (only 2 days) did not allow for a meaningful calculation of standard deviations.

EGUSPHERE- 2025-3842:

“Aerosol Size Distribution and New Particle Formation in High Mountain Environments: A Comparative Study at Monte Cimone and Jungfraujoch GAW Stations”

Answers of the authors to Reviewer#2

While the reviewer’s comments are given in **bold red**, our answers are given below in black letters. Additionally, we added the changes made in the revised manuscript in *italic black text*.

This study by Mazzini et al. compares two years of observations of aerosol size distribution data at two high-altitude GAW stations in central Europe, Jungfraujoch (JFJ) and Monte Cimone (CMN). The focus is on the frequency and strength of New Particle Formation (NPF) and the abundance of ultrafine particles. As free tropospheric aerosol production is important in the climate system, such high-altitude location studies are interesting tool to investigate the processes related to NPF in the free troposphere and its coupling to the planetary boundary layer. The manuscript is well written, uses appropriate methods to analyze NPF and gives some insights into the role of boundary layer influence on free tropospheric processes. It merits publication in Atmos. Chem. Phys.. Apart from some minor points, I have some concerns about the merging of NAIS and SMPS for the combined particle number size distribution (PNSD) which should be addressed carefully, before publication.

Major comments:

- The authors describe a procedure to combine the NAIS particle mode data with the SMPS data to obtain a merged PNSD. In the overlapping size range, they derive scaling factors which are then applied across the entire NAIS dataset. This is problematic, as the NAIS offset to SMPS data might not be constant across all sizes, as its origin is partly due to unknown multiple charged particles in the larger sizes and agreement is often better at smaller sizes (Kangasluoma et al., 2020). This might induce some considerable error on the absolute number concentrations derived for the modes measured mainly with NAIS and the percentage shares in Figure 2 might therefore be highly uncertain. In addition to that the SMPS sample is dried, and the NAIS is not, according to the description. This introduces some additional uncertainty in the merging of the two instruments, as PNSD are merged at the same diameter, while the one might be dry and the other might be a wet diameter. The authors should investigate the overlap of the size distribution in dependence on RH to see if there is a significant**

difference. Last, I couldn't find an explanation why different size ranges are used for JFJ and CNM to merge the size distributions. This should be clarified.

The best approach to address all these concerns would be a comparison of the integrated merged PNSD with a CPC measuring the total number concentration of aerosol particles, which to my knowledge should at least be available for JFJ.

We thank the reviewer for this detailed and constructive comment. In response, we performed several additional analyses to (i) investigate the size dependence of the NAIS/SMPS offset, (ii) test the influence of relative humidity on the overlap region.

1) Size dependence of the NAIS–SMPS offset

For every size bin in the overlap region, we computed the median NAIS/SMPS ratio together with two indicators of its stability:

- (i) the interquartile range (IQR), reflecting the spread of the ratio, and
- (ii) the coefficient of variation ($CV = \sigma/\mu$), quantifying its relative variability.

Size-dependent disagreements between the instruments manifest as large or progressively changing values of these metrics, whereas stable agreement appears as low and approximately diameter-independent IQR and CV.

The two stations show distinctly different behaviour (Figure 1).

CMN: Both IQR and CV decrease quickly with increasing diameter and form a low, flat baseline from 18 nm onward. In this range the median ratio varies smoothly, the IQR is narrow, and no systematic diameter trend is apparent. We therefore identify 20–40 nm as a stable overlap interval and use it to derive the scaling factors; the merged PNSDs are joined at 25 nm.

JFJ: Here the ratio remains more variable at small sizes and only levels off above 30 nm, where the IQR becomes small and the CV fluctuates around a constant baseline. The 30–40 nm interval is therefore used to determine the scaling factors, and 35 nm is chosen as the merging diameter. SMPS data below 20 nm were excluded because their uncertainty is known to be high at this station.

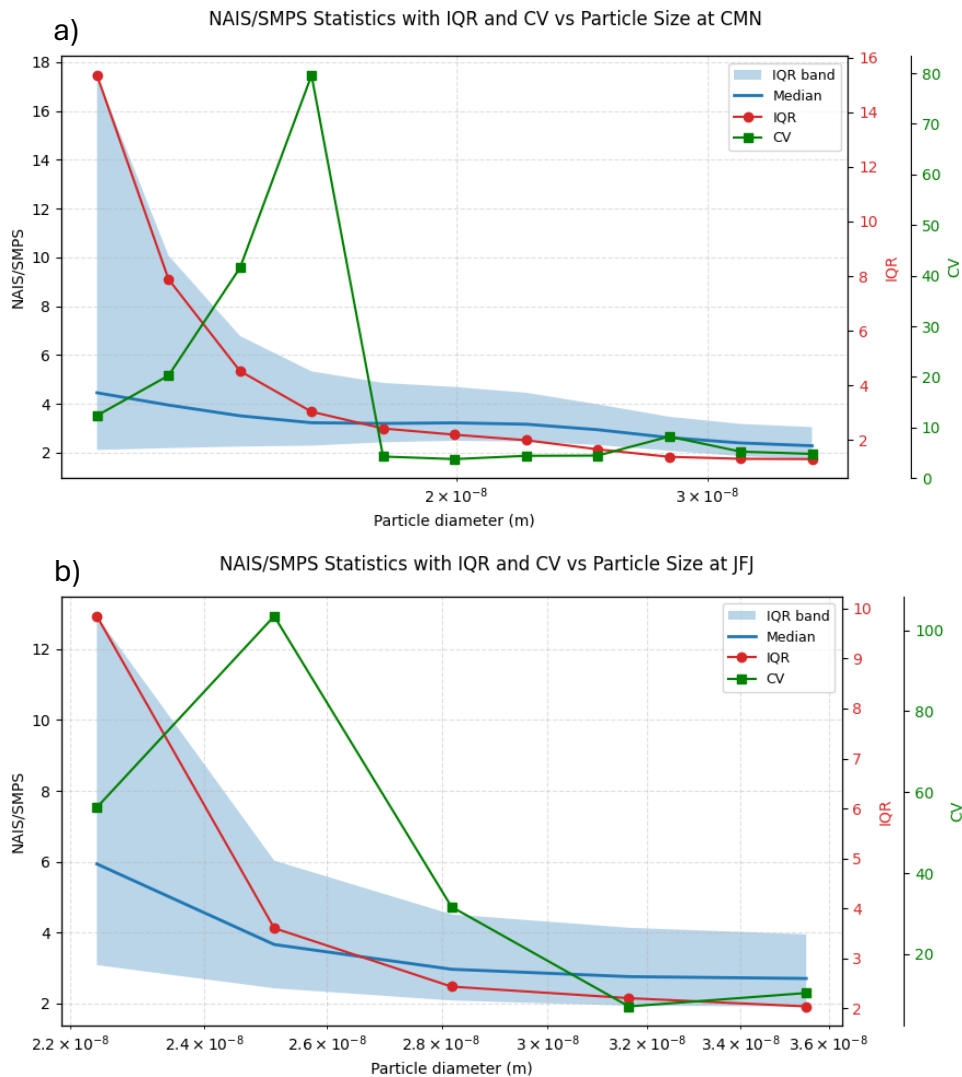


Figure 1. Size dependence of the NAIS/SMPS ratio at (a) CMN and (b) JFJ. For each station, the blue curve shows the median NAIS/SMPS concentration ratio and the shaded area its interquartile range (IQR). The red and green curves display the IQR and coefficient of variation (CV), respectively, providing independent measures of the stability of the NAIS–SMPS offset across particle diameter. At CMN, both metrics stabilise above 18–20 nm, while at JFJ stability is reached only above 30 nm.

2) Effect of SMPS drying vs. NAIS ambient humidity

The reviewer correctly notes that differences in drying between the NAIS and SMPS can introduce uncertainty when merging size distributions. In the diameter range relevant for the overlap (10–40 nm at CMN and 20–40 nm at JFJ), however, hygroscopic growth is expected to be small. Theoretically, particles below 40 nm take up little water because the curvature term (Kelvin effect) in the Köhler equation suppresses condensation at typical atmospheric RH. This expectation is consistent with observational evidence from H-TDMA (Hygroscopic Tandem Differential Mobility Analyzer) measurements, which show that hygroscopic growth decreases strongly with decreasing particle size and is generally limited for diameters below

40 nm (Swietlicki et al., 2008). In addition, the NAIS inlet is trace-heated, further reducing the effective RH inside the instrument.

Our RH-binned analysis supports this expectation: within the dominant RH range (0–80%), the NAIS/SMPS ratio remains nearly constant (Fig. 2). A slight increase appears only in the highest RH bins (>80–90 %), primarily in the smallest NAIS channels. This behaviour is unlikely to reflect hygroscopic growth and more probably arises from artefacts or processes occurring under very high RH or cloud-influenced conditions. Crucially, the scaling factors are derived from the 20–40 nm interval at CMN and the 30–40 nm interval at JFJ, where no systematic RH dependence is observed. Therefore, the drying difference between NAIS and SMPS should not compromise the robustness of the harmonization procedure.

The text has been revised as follow:

When operating in total particle mode, the NAIS may differ substantially from reference measurements, with reported discrepancies of up to an order of magnitude compared to SMPS data (Kangasluoma et al. 2020). Therefore, a detailed comparison was performed with the reference SMPS at both stations within overlapping size ranges: 20–30 nm for CMN and 30–40 nm for JFJ. These size intervals were selected because they correspond to the diameters where the NAIS/SMPS concentration ratio showed the lowest relative variability (coefficient of variation) and a narrow interquartile range, indicating that both instruments provided the most reliable and size-independent agreement in this region. At JFJ, diameters below 20 nm were excluded because SMPS performance is known to degrade in this range. Scaling factors were derived individually for each measured size distribution and applied to all NAIS channels accordingly. The resulting mean scaling factors were 3.38 ± 2.05 for CMN and 3.18 ± 2.07 for JFJ. After scaling, NAIS and SMPS distributions were merged at 25 nm for CMN and at 35 nm for JFJ (see Fig. S2). To ensure that drying differences between instruments did not bias the merging, we also examined the NAIS/SMPS ratio in the overlap region as a function of ambient RH. The ratio showed no systematic RH trend within the selected intervals, consistent with the limited hygroscopic growth expected for particles below 40 nm and with the trace-heated NAIS inlet. A weak RH sensitivity is visible mainly at the smallest NAIS channels, but these diameters are not used for harmonization and do not affect the derived scaling factors.

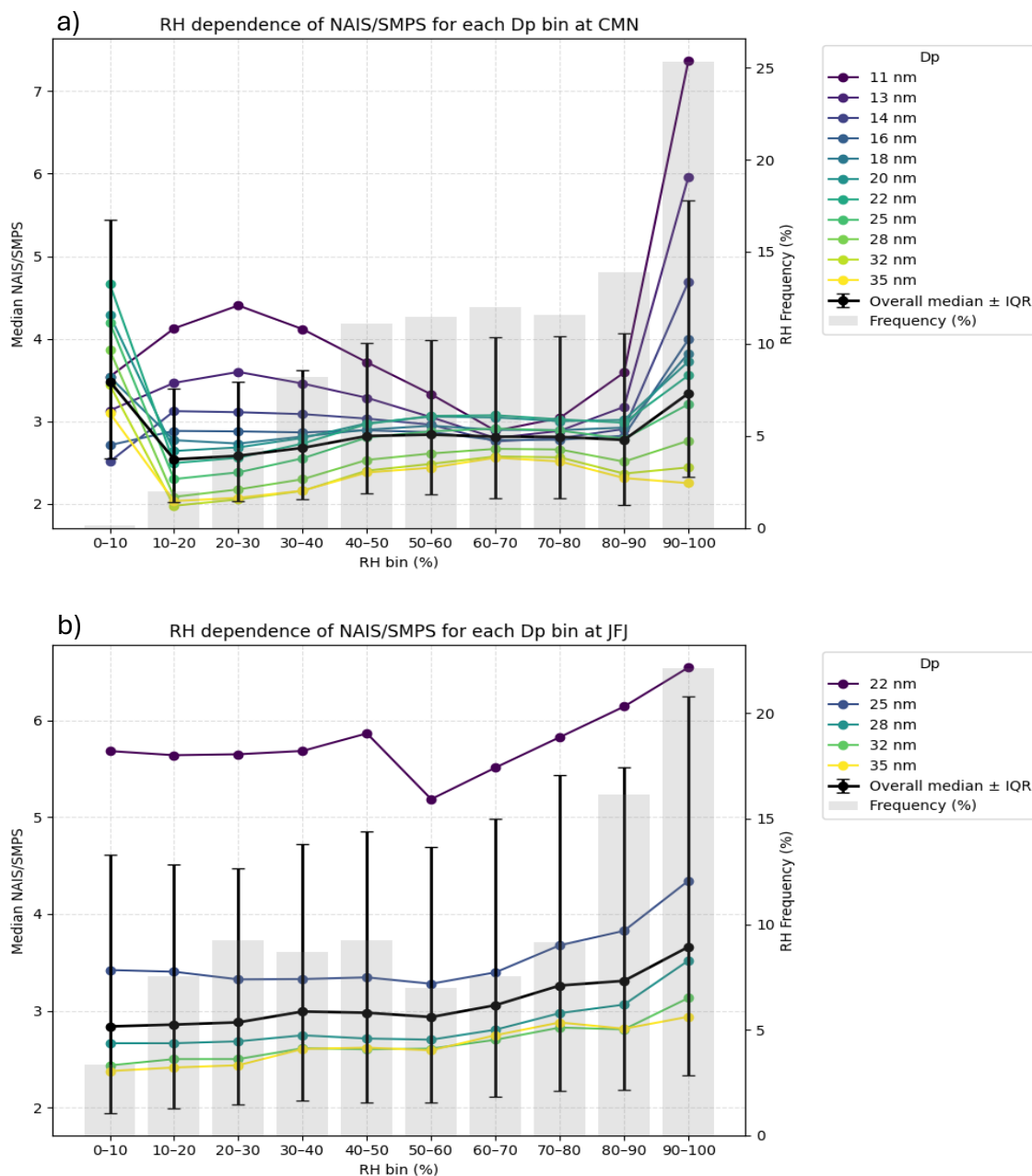


Figure 2. Relative-humidity dependence of the NAIS/SMPS ratio at (a) CMN and (b) JFJ. For each station, the curves show the median NAIS/SMPS ratio for individual NAIS diameter bins as a function of ambient RH (10% bins). Black markers denote the overall median \pm IQR across all diameters, and grey bars indicate the frequency of observations Dp in each RH bin.

3) CPC comparison

We agree that CPC data would provide an independent reference for assessing the merged size distribution. However, at both sites the available CPCs had a lower cutoff of ≈ 10 nm, which means they do not capture the nucleation and early intermediate modes measured by the NAIS. Because the SMPS already measures the >10 nm size range with high reliability comparing the merged NAIS+SMPS PNSD to the CPC would only validate the SMPS-dominated part of the distribution, not the NAIS-dependent size range (<10 nm).

Indeed, we performed this analysis at CMN and found a median CPC/SMPS ratio of 1.01 ($R^2 = 0.91$), confirming that the SMPS provides a reliable reference for particles >10 nm under typical conditions.

Minor comments:

- **Line 110: Technically the survival probability (growth versus coagulation loss) is even lower for the intermediate mode and therefore fast growth even matters more there.**

We fully agree with the reviewer's observation. The text has been revised to reflect the greater importance of rapid condensational growth in the intermediate mode, where coagulation losses are strongest. The updated paragraph now reads:

The intermediate mode (2.5–7 nm) comprises the smallest detectable particles, marking the early stage of NPF where rapid condensational growth is essential to counter strong coagulation losses. The nucleation mode (7–25 nm) represents particles that have survived initial coagulation and continue to grow through condensation of low-volatility vapors. Their growth efficiency determines whether they can reach the Aitken mode (25–100 nm), from which CCN activation becomes possible. The accumulation mode (100–560 nm) consists of aged particles shaped by long-range transport and chemical processing.

- **Line 139: Clarify in the text what the variables $\text{Max}(\Delta N_{3-6})_{\text{active}}$ and $\text{Median}(\Delta N_{3-6})_{\text{non-active}}$ represent.**

The text has been revised to clarify the definition of these variables. The updated sentence now reads:

This ratio compares peak daytime (09:00–16:00) particle concentration, $\text{Max}(\Delta N_{3-6})_{\text{active}}$, to nighttime background (16:00–09:00), $\text{Median}(\Delta N_{3-6})_{\text{non-active}}$, serving as a continuous index of NPF intensity.

- **Line 142: The presence of small particles at high RH is attributed to cloud processing and not instrument artefacts here. Is there any reference the authors can provide?**

While we could not identify a published reference addressing this feature directly, similar observations have been reported at other locations. We changed to: *likely associated with cloud processing or potential instrumental artefacts at elevated humidity*

- **Line 161: Related to the above: What does it mean the PNSD remained stable? Anything quantitative?**

We thank the reviewer for this valuable comment. We agree that the term “stable” was imprecise and have revised the text to provide a clearer and more quantitative description. The revised sentence now specifies that below 94% RH, the PNSD showed no humidity-related variations in the sub-10 nm range (except during nucleation events). The updated text reads:

These exploratory analyses, later supported by the findings presented in Sect. 4.2, indicated that for $RH < 94\%$, the PNSD showed no humidity-related variations in the sub-10 nm range, apart from those associated with nucleation events, and thus represented out-of-cloud conditions. Conversely, when RH exceeded 97%, distinct modifications in PNSD consistent with cloud processing became apparent, supporting classification as in-cloud, in line with Herrmann et al. (2015).

- **Line 168: unit missing**

Corrected

- **Line 176: The higher fractional share of Aitken mode particles at CMN does not necessarily mean that survival of NPF particles is enhanced (the share could just be higher as the “background”, i.e. accumulation mode, is overall lower at JFJ. In fact, when comparing nucleation mode to Aitken mode concentrations they are more similar at JFJ in terms of absolute numbers, indicating a lower loss.**

We thank the reviewer for this insightful comment. We fully agree that a higher fractional share of Aitken-mode particles at CMN does not necessarily imply enhanced survival of newly formed particles, as mode fractions are influenced by the background aerosol load. The text has been revised accordingly to reflect this clarification. The updated passage now explicitly states that the larger Aitken-mode share at CMN may result from higher overall particle concentrations and that absolute nucleation- and Aitken-mode numbers are more comparable at JFJ, suggesting lower relative losses there. The updated paragraph now reads:

JFJ exhibits a higher relative contribution of smaller particles, with the intermediate mode contributing 26.5% of total particles compared to 14.1% at CMN, and the nucleation mode making up 32.2% at JFJ versus 28.8% at CMN. While this suggests a higher fraction of small clusters at JFJ, it does not necessarily indicate more efficient particle formation, as these clusters must survive and grow into larger sizes to contribute to total aerosol load. The Aitken mode dominates the number concentration at CMN, accounting for 38.9% of total particles versus 27.1% at JFJ. However, this higher fractional share does not directly reflect enhanced survival of newly formed particles, since the relative contribution of each mode also depends on the overall aerosol background. In absolute terms, nucleation- and Aitken-mode concentrations are more comparable at JFJ, suggesting lower relative losses there.

- **Line 179: In the same sense the absolute concentrations of the accumulation mode at CMN are twice as large as at JFJ (while the relative share doesn't increase that much).**

We agree with the reviewer's observation and have revised the text to explicitly mention that accumulation-mode particle concentrations at CMN are roughly twice those at JFJ, despite only modest differences in fractional contribution. This change clarifies that the higher total aerosol load at CMN reflects both enhanced regional influence and a stronger accumulation-mode background. The updated paragraph now reads:

Furthermore, accumulation-mode particle concentrations at CMN are about twice those at JFJ, indicating a generally higher aerosol load at CMN despite only modest differences in fractional contribution (see Fig. 2).

- **Line 181: Fig 3 is referenced while the text probably references to Fig. 2 and moreover the total number concentrations across the seasons are never shown.**

Corrected:

Seasonal total particle number concentrations peak in summer (CMN: 1743 cm^{-3} , JFJ: 1360 cm^{-3}) and reach their lowest in winter (CMN: 718 cm^{-3} , JFJ: 568 cm^{-3}). As visible in Fig. 2, the seasonal pattern is mainly driven by the Aitken and accumulation modes, which increase substantially in warm months due to enhanced boundary layer activity (Rose et al., 2021; Herrmann et al., 2015)

- **Figure 3 deserves a more detailed caption explaining the shaded areas (interquartile ranges) and colors.**

Better caption:

Diurnal variation of hourly median particle number concentrations across four modes (intermediate 2.5–7 nm, nucleation 7–25 nm, Aitken 25–100 nm, and accumulation 100–560 nm) for each season is shown for the CMN (solid black line) and JFJ (dashed blue line) stations. Shaded areas represent the interquartile range (25th–75th percentiles) of the hourly medians. All times are given in UTC.

- **Line 194: There seems to be sometimes a kind of bimodal pattern in the intermediate particles both at JFJ and CMN. Any explanations for this? Showing some median diurnal PNSD (as surface plots) in the Supplement would help to better understand what is shown in Fig. 3.**

We thank the reviewer for this observation. A weak bimodal structure occasionally appears in the smallest size ranges. The first peak tends to coincide with the initial rise in solar radiation, while the later and more dominant peak is likely linked to the arrival of boundary-layer air once mixing has fully developed. To provide additional context, we have added the seasonal median diurnal PNSD surface plots together with the

median seasonal solar radiation to the Supplementary Material (Fig. S5). We also incorporated a short explanation into the main text, which now reads:

Diurnal patterns at CMN and JFJ (Fig. 3) show that intermediate particles remain relatively stable throughout the day, with a slight midday increase aligning with peak solar radiation. However, larger variations in the interquartile range (IQR) indicate that nucleation events are episodic, with pronounced midday peaks (10:00–12:00 UTC) in all seasons at both sites. These peaks are particularly evident in spring and autumn, suggesting a seasonally varying nucleation activity. To provide additional context, the corresponding seasonal median diurnal PNSD surface plots, together with the median seasonal solar radiation, are shown in the Supplementary Material (Fig. S5). The nucleation mode shows a clear progression of particle growth beyond the smallest clusters, peaking shortly after the intermediate mode, showing that a fraction of freshly formed particles successfully grow past the critical survival size. This growth is most evident in spring and autumn, while in summer, less favorable conditions for particle formation and growth reduce the intensity of this cycle. In spring and summer, a weak bimodality can occasionally be seen in the smallest size bins, most clearly in the 75th percentiles, with an earlier enhancement coinciding with the first increase in solar radiation and a later, more pronounced peak as boundary-layer air arrives. Consistently, Aitken and accumulation mode particles peak later in the afternoon, reflecting both the continued growth of nucleated particles and contributions from boundary-layer transport.

- **Line 198: I am struggling with the word “nucleation efficiency”. What does it mean? High GR, low sink, high J or just high GR and low sink?**

Thank you for pointing this out. The term “*nucleation efficiency*” was indeed ambiguous, as it could refer to several competing processes (e.g., formation rates, growth rates, or condensational sinks). To avoid confusion, we revised the sentence to describe the physical processes more explicitly without using this unclear term. The new wording reads: “*This growth is most evident in spring and autumn, while in summer, less favorable conditions for particle formation and growth reduce the intensity of this cycle.*”

- **Line 228: I think that “personal communication” is not a valid citation in Copernicus journals.**

Thank you for pointing this out. We agree that “personal communication” is not an appropriate citation format for Copernicus journals. We have replaced the personal communications with published (or in-preparation) sources and updated the sentence accordingly. The revised text now reads:

These elevated ratios have also been reported at other high-altitude stations, including

Chacaltaya (Aliaga et al., 2025) and Zeppelin (Heslin-Rees et al., 2025), and preliminary data from Izaña indicate similar behaviour.

- **Line 229: The lower negative cluster ion concentrations are also very much related to reduced sensitivity of the instruments towards these highly mobile ions. Negative cluster ions have on average higher mobilities than positive cluster ions and more easily get lost in the inlet system. Without perfect corrections for this, their measured concentration will be lower due to higher losses. This should be mentioned here clearly.**

We agree that the reduced sensitivity of ion instruments to highly mobile negative cluster ions is an important factor that should be addressed explicitly. We have now added a clear explanation of how the higher mobility of negative ions leads to enhanced diffusion and inlet losses, and how lower atmospheric pressures at mountain sites further increase these effects, potentially shifting negative ions beyond the detectable size range. The revised text is as follows:

While laboratory calibrations do not reveal systematic biases between polarities, environmental factors and ion chemistry likely contribute to the observed asymmetry. Variations in core ion composition, precursor availability, and ion sink mechanisms may influence ion mobility and size distributions, particularly in the cluster and intermediate ranges. In addition, topographic effects and the associated atmospheric electric fields at mountain sites may modulate ion charge distributions. A further and important explanation for the relative abundance of positive over negative cluster ions is related to instrumental and sampling line limitations. Negative cluster ions have higher mobilities than positive ones and therefore experience stronger diffusion and inlet losses; without appropriate corrections, this leads to systematically lower measured concentrations. At the lower atmospheric pressures typical of mountain sites, ion mobilities increase even further, and highly mobile negative ions may shift beyond the instrument's effective detection range, resulting in additional underestimation of their concentrations (Hirsikko et al., 2011). Further investigation is warranted to better understand the instrumental, chemical, and physical drivers of these charge distributions. Moreover, within ACTRIS RI new guidelines for uncertainties and correction schemes for sampling lines should be published, as done for SMPS (Wiedensohler et al., 2012).

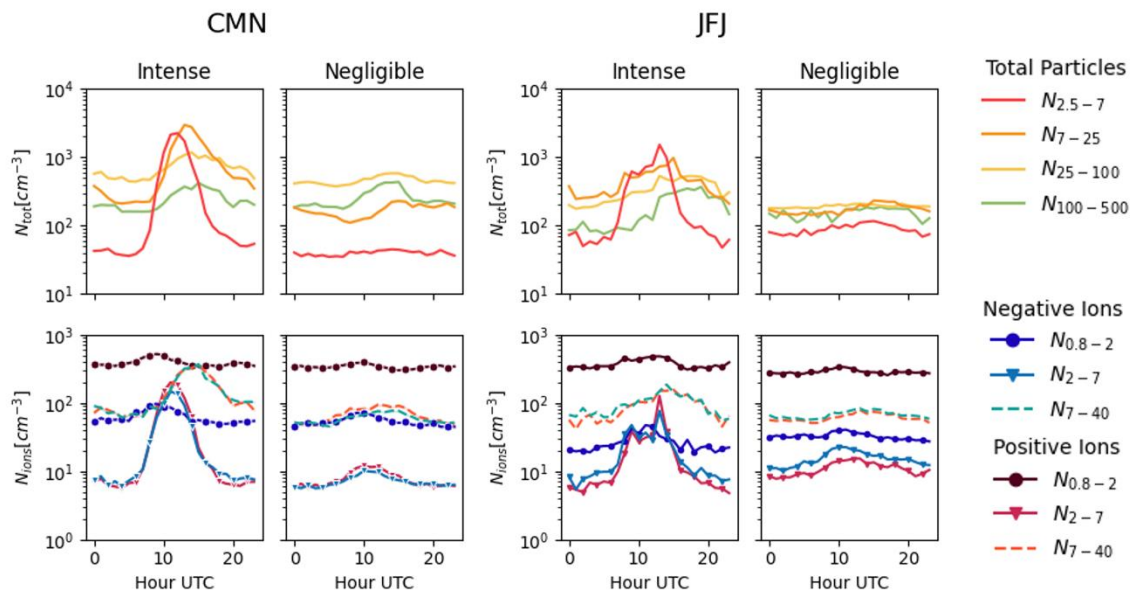
- **Line 310: Or the lower absolute accumulation mode background at JFJ allows an easier detection of smaller changes, i.e. few grown particles reaching accumulation mode sizes, which would not be detectable at CNM.**

Thank you for this helpful remark. We agree that the lower absolute accumulation-mode background at JFJ may allow smaller changes to be detected more easily. We have incorporated this explanation into the revised text, which now reads:

Concerning the accumulation mode, CMN does not show significant differences between intense and negligible NPF events, whereas at JFJ an increase in accumulation-mode particles is observed in the afternoon during intense events. On the one hand, this may reflect the influence of vertical transport, peaking in the afternoon, on NPF occurrence, for example when particle formation is initiated at the interface between clean free-tropospheric conditions and PBL injections. On the other hand, the lower absolute accumulation-mode background at JFJ might make small changes more detectable, meaning that a few particles grown into the accumulation-mode size range could be observed there but remain undetectable at CMN.

- **Figure 9: The coloring for the ion plot could have better contrast.**

The plot has been changed, the lines style and the colors have been modified so that it is clearer.



- **Line 318: It reads like sulfuric acid stabilizing the early clusters, while in fact, it might be the main component of them. The ions are stabilizing the cluster. Please rephrase.**

Rephrased into:

The critical role of negative cluster ions in nucleation under clean and low condensation-sink conditions, such as those prevailing at high-altitude sites, is highlighted by Rose et al. (2018), who showed that newly formed clusters often consist of sulfuric acid and highly oxidized organic molecules carrying a negative charge.

- **Line 331-333: The authors discuss some results here without showing the data or referencing it.**

Thank you for pointing this out. We have added a new supplementary figure (Fig. S6) showing the mean diurnal cycles of temperature, wind speed, shortwave radiation, and relative humidity for intense and negligible NPF days at both stations. The corresponding sentence in the main text now explicitly refers to Fig. S6. This ensures that all meteorological interpretations are directly supported by visible data.

The revised text reads:

To investigate the atmospheric conditions that control NPF, we analyzed the meteorological variables and condensation sink (CS) under out-of-cloud conditions, grouping days by NPF intensity (intense vs. negligible) as determined by the nanoparticle ranking method. The diurnal behaviour of solar radiation, RH, temperature, and wind speed on intense and negligible NPF days is shown in Fig. S6.

- **Line 334: Isn't the warmer temperature also indicative for a higher PBL air mass intrusion?**

We agree with the reviewer. The revised text now clarifies that the higher temperatures observed on intense NPF days at CMN are consistent with enhanced boundary-layer influence.

The updated text reads:

Temperature and wind speed exhibited clear site-specific behaviour. At CMN, intense NPF days were generally warmer than negligible ones, consistent with periods of stronger boundary-layer influence and associated precursor transport. At JFJ, temperature differences between intense and negligible events were much smaller, indicating that temperature plays a weaker role in modulating NPF at this site. Wind patterns showed a similarly contrasted behaviour: at CMN, winds were slightly stronger during the early hours preceding intense nucleation events, potentially facilitating precursor transport, whereas at JFJ wind speeds remained relatively constant throughout the day, with higher values generally coinciding with suppressed NPF, likely due to enhanced dilution and reduced precursor residence time.

- **Line 345: The statement that clean atmospheric conditions are needed for enabling nucleation and early particle growth is partly correct, but here and, more importantly, also in the conclusions I am missing the following thought process: While a lower CS is better, at JFJ we see that if the conditions get too “clean” in a sense of less boundary layer interaction (the real FT) we are at the same time missing the precursor vapors. It seems to be a very delicate balance between having clean conditions in terms of low background and at the same time a high abundance of precursor vapors such that NPF proceeds efficiently. This is seen at many places around the world (in megacities, too high CS typically prevents NPF, still NPF is possible to proceed at higher CS than in rural regions as also more precursors are available). I think the comparison between JFJ and CMN shows exactly this: Cleaner conditions in term of background do help, but we also need**

the precursors, which at CMN are more abundant. This argument should be brought forward very clearly in a revised discussion.

We thank the reviewer for this insightful comment. We fully agree that clean conditions alone do not guarantee efficient NPF and that an optimal balance between low condensation sink and sufficient precursor availability is required. To incorporate this important point, we revised the discussion to emphasise that excessively clean free-tropospheric conditions (i.e., very weak boundary-layer influence) can limit nucleation because precursor vapours become scarce. Conversely, periods with moderate boundary-layer coupling can provide the vapours needed for nucleation while still maintaining sufficiently low background aerosol levels.

The updated text at line 342 reads:

This seasonal variability aligns with the role of precursor and condensable vapours availability in high-altitude environments. A lower CS reduces competition for condensable vapours and favours particle survival (Sellegrì et al., 2019), but if conditions become too clean, such as during periods with limited PBL influence, the availability of precursor vapours can become insufficient to sustain NPF. Efficient nucleation therefore requires a delicate balance: a background clean enough to minimize vapour losses, yet sufficiently influenced by the boundary layer to supply the necessary precursors. This behaviour is clearly illustrated by the contrast between CMN, experiencing stronger PBL influence and higher precursor concentrations, and JFJ, representing cleaner but precursor-limited conditions. Similar dynamics have been reported at various locations worldwide, where NPF efficiency depends on the combined effects of condensation sink and precursor abundance (Bianchi et al., 2016; Sellegrì et al., 2019; Zheng et al., 2021).

Moreover, we revised the conclusion section starting at line 392 to reflect this balance between low CS and sufficient precursor availability:

Condensation sink (CS) emerged as a key limiting factor for nucleation, with intense NPF events consistently associated with lower early-morning CS values. Growth rates at CMN were typically higher than at JFJ, especially during summer, highlighting more favorable conditions for particle survival and growth. While background concentrations of small ions and particles were higher at JFJ, they did not translate into higher nucleation or growth efficiency, indicating that ion-induced nucleation alone is insufficient to sustain NPF in the absence of favorable thermodynamic and chemical conditions.

However, our results also demonstrate that clean conditions alone are not sufficient for efficient NPF. When the atmosphere becomes too weakly coupled to the boundary layer, as occasionally occurs at JFJ, the scarcity of precursor vapours can limit

nucleation despite low CS values. Efficient NPF therefore requires a balance between low background aerosol concentrations and sufficient supply of condensable vapours, conditions that are more frequently met at CMN due to its stronger and more regular boundary-layer influence.

Cloud conditions, inferred from relative humidity thresholds ($RH > 97\%$), significantly altered the observed size distributions, particularly suppressing particles in the CCN size range and introducing anomalous sub-10 nm modes. Ion concentrations, particularly positive cluster ions, were strongly reduced under in-cloud conditions, consistent with efficient ion scavenging.

In conclusion, mountain observatories, even within the same continental region, can exhibit markedly different aerosol dynamics due to differences in atmospheric coupling, precursor availability, and altitude. CMN is characterized by higher variability of aerosol populations and stronger coupling with regional sources, while JFJ represents a predominantly free-tropospheric environment that still experiences episodic local and anthropogenic influences. The contrast between these two sites underscores that NPF depends not only on clean background conditions but also on adequate precursor availability. These results highlight the need for long-term, harmonized observations of ions and ultrafine particles to improve parameterizations of NPF in chemical transport and climate models, particularly in regions where the FT is intermittently influenced by boundary layer intrusions.

- **Line 358: I guess summer data doesn't provide enough statistics to really conclude this (no SD given in the Table, so I assume the number relies only on two values).**

We thank both reviewers for noting the missing uncertainty values in Table 3 and the limited statistics for JFJ summer data. Indeed, the small number of valid summer observations at JFJ (only 2 days) did not allow for a meaningful calculation of standard deviations.