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*.

However, I still have concerns about the way the authors use the scaling factors of the SMPS/NAIS ratio at large diameters to correct the entire NAIS size-distribution. The authors now show that the ratio between the two instruments is most stable at larger sizes and they also show why they chose a different size for deriving the scaling factors at the two stations. However, their response does not address my concern why the scaling factor derived at >=25 nm should be reflecting the scaling the NAIS needs at small diameters (<10 nm). In fact, their size-dependent analysis shows that the ratio has a higher variability the smaller they get and while they use it as an argument to use the scaling factor derived at >25 nm, it is also an argument against the apüplication of that scaling factor at smaller diameters. The scaling factor is probably just not constant in size (as it depends on multiple-charging effects as I mentioned in my first comment). If they find a scaling factor of 3 at >25 nm, there could be an actual offset of 10 at small diameters, or no offset at all. The point is, that merging the two instruments and scaling one of them based on a comparison at larger sizes will not necessarily improve the results at smaller sizes. For the final version of this paper, I would need to see that clarified very explicitly that the uncertainties on small particle number concentrations are significant and that this introduces uncertainty in all calculation so relative shares of the modes, etc. Only after this is clarified, I can recommend publication.

Public justification (visible to the public if the article is accepted and published):
The authors have addressed the majority of the reviewers' comments satisfactorily. However, one issue raised by Reviewer 2 remains mostly unresolved. The authors are, therefore, requested to carefully consider this comment and provide an appropriate response, and/or revise the relevant part of the manuscript accordingly. They are also asked to consider the Notification from review file validation. (While Reviewer 2 suggested a minor revision, the editorial decision has been designated as a major revision to allow Reviewer 2 to participate in the next round of assessment.)

We thank the reviewer for raising this important point concerning the application of a scaling factor derived at larger particle sizes to the smaller size range measured by NAIS. We agree that the SMPS/NAIS ratio is not constant with particle size and that differences between the two instruments increase toward smaller diameters, implying substantial uncertainty in absolute particle number concentrations below the merging diameters.

In the Supplementary Material (Section S2), we now present a dedicated sensitivity analysis in which results obtained with the applied scaling are compared to both unscaled data and alternative scaling factors derived from smaller overlap ranges. This analysis quantitatively demonstrates that absolute particle number concentrations below the merging diameters (25 nm at CMN and 35 nm at JFJ) are sensitive to the choice of scaling factor. The full text of the newly added Supplementary section S2 is reported below.

S2 Sensitivity analysis of scaling applied to NAIS–SMPS harmonization

A quantitative sensitivity analysis was performed to assess how particle number concentrations depend on both the magnitude of the applied scaling factor and the size range used to derive it. The scaling configuration adopted in the paper, based on the most stable NAIS–SMPS overlap range (30–40 nm at JFJ and 20–30 nm at CMN), was used as the baseline scenario. The impact of the scaling and the resulting NAIS–SMPS merging on the median particle number size distributions is illustrated in Fig. S2, which shows the individual NAIS and SMPS PNSDs together with the final merged distributions. In the merged datasets, NAIS-derived concentrations are used below the merging diameters (35 nm at JFJ and 25 nm at CMN), while SMPS data dominate at larger sizes. The resulting mean scaling factors were 3.38 ± 2.05 for CMN and 3.18 ± 2.07 for JFJ.

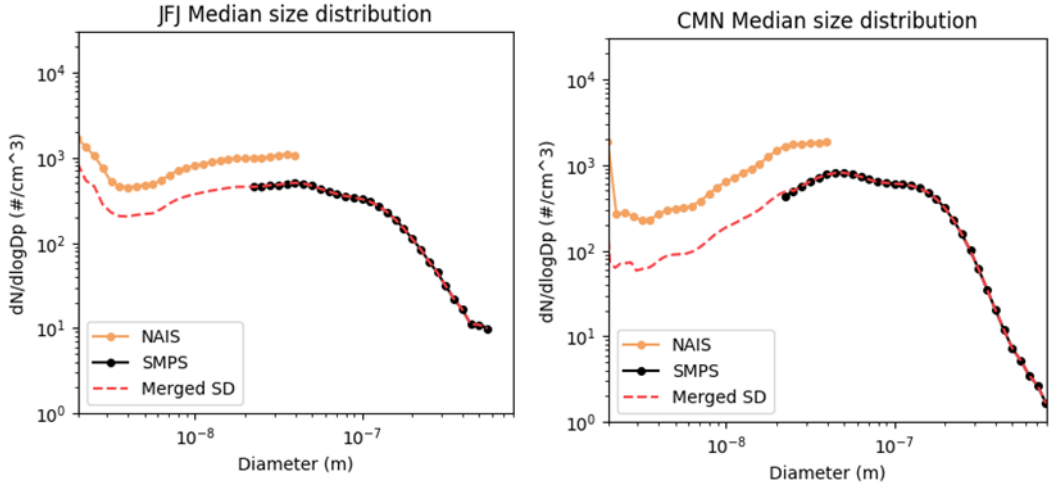

Figure S2. Median PNSD derived by NAIS and SMPS instruments at JFJ and CMN stations. The red dotted line represents the final merged PNSD, after NAIS data correction.

The baseline results were compared with two alternative configurations: (i) a case without any scaling, in which uncorrected NAIS data were used below 25 nm at CMN and 35 nm at JFJ, and (ii) cases using stronger scaling factors derived from smaller overlap ranges (10–20 nm at CMN and 20–30 nm at JFJ). The resulting mean and median particle number concentrations for all size modes and scaling scenarios are summarized in Table S1.

At JFJ, particle number concentrations in the NAIS-only size range show a strong dependence on the applied scaling. In the intermediate mode $N_{[2.5-7)}$, the mean concentration increases from 379.5 cm^{-3} in the baseline case to 789.6 cm^{-3} without scaling (+108%), while the median increases from 91.5 cm^{-3} to 248.4 cm^{-3} (+171%). Applying a stronger scaling factor derived from the 20–30 nm overlap range (4.63 ± 3.56) reduces the mean concentration to 234.9 cm^{-3} (−38%) and the median to 67.7 cm^{-3} (−26%). A comparable sensitivity is observed in the nucleation mode $N_{[7-25)}$, where the mean varies by +130% (without scaling) and −34% (stronger scaling) relative to the baseline. In the Aitken $N_{[25-100)}$ range, which is partially constrained by SMPS data, the mean changes by +33% and −3%, respectively.

At CMN, removing the scaling increases the mean concentration of particles in the intermediate mode $N_{[2.5-7)}$ from 273.5 cm^{-3} to 864.1 cm^{-3} (+216%) and the median from 65.0 cm^{-3} to 188.2 cm^{-3} (+189%). Applying a stronger scaling factor derived from the 10–20 nm overlap range (6.97 ± 15.54) reduces the mean concentration to 209.4 cm^{-3} (−23%) and the median to 54.8 cm^{-3} (−16%) relative to the baseline.

	JFJ [cm^{-3}]					**CMN [cm^{-3}]**				
	N_{2-7}	N_{7-25}	N_{25-100}	$N_{100-500}$	N_{2-500}	N_{2-7}	N_{7-25}	N_{25-100}	$N_{100-500}$	N_{2-500}
	Baseline Scaling (30–40 nm)					**Baseline Scaling (20–30 nm)**				
Mean	379.5	442.1	336.1	165.9	1323.6	273.5	546	665	302.2	1786.6
Median	91.5	197.3	222.4	68.6	770.2	65	220.2	443.2	176.2	1218.9
SD	1654.2	993.3	384.7	234.6	2387.3	909.4	1179.8	777.4	322.7	2252.3
P25	41.2	96.4	126.6	32.8	482.7	27.6	100.5	253.1	60.3	733.5
P75	231.9	424.7	404.1	177.7	1444.6	182.6	525.8	776.5	449.2	2029
	No Scaling					**No Scaling**				
Mean	789.6	1016.1	448.5	165.9	2418.7	864.1	1592.3	665	302.2	3646
Median	248.4	515.8	295.4	68.6	1404.3	188.2	633.8	443.2	176.2	2101.2
SD	2348.8	2051.9	497	234.6	3891.6	3067.2	4838.3	777.4	322.7	7475.9
P25	128.5	297.5	174.2	32.8	898.7	87.9	327.3	253.1	60.3	1248.3
P75	549	974.7	538.3	177.7	2505.8	512.2	1483.7	776.5	449.2	3830.6

	Scaling (20-30 nm)					Scaling (10-20 nm)				
Mean	234.9	292.8	327.2	165.9	1021.2	209.4	420.4	665	302.2	1595.9
Median	67.7	135.1	214.7	68.6	634.1	54.8	159.5	443.2	176.2	1039.7
SD	908.2	618.5	377.1	234.6	1510.4	1044.9	971	777.4	322.7	2138.5
P25	29.9	63.8	122	32.8	389.1	22.8	76.7	253.1	60.3	624.1
P75	166.2	295	391.6	177.7	1199.4	138.7	356.9	776.5	449.2	1806

Table S1. Mean, median, standard deviation (SD) and percentiles (P25, P75) of particle number concentrations (cm^{-3}) for different size ranges at JFJ and CMN under alternative NAIS–SMPS scaling configurations.

Overall, the sensitivity analysis demonstrates that particle number concentrations below the merging diameters are sensitive to both the magnitude of the applied scaling factor and the size range used to derive it. When expressed as fractional contributions, the relative partitioning between the intermediate and nucleation modes varies depending on whether scaling is applied. However, the combined contribution of small particles (2.5–25 nm) remains consistently higher at JFJ than at CMN for both the scaled and unscaled datasets, while CMN remains dominated by the Aitken and accumulation modes. Seasonal and diurnal variability patterns, as well as the relative ordering of particle size modes and the site-to-site contrasts, are preserved across all tested scaling scenarios. Consequently, uncertainties associated with the harmonization procedure propagate predominantly into the smallest size classes and affect absolute concentrations and derived quantitative metrics, while the qualitative conclusions presented in the main text remain robust to reasonable variations in the scaling assumptions.

We have also revised the section “Instrument Harmonization and Data Processing” to explicitly motivate this approach and to place it in the context of existing literature that adopted a similar approach for data harmonization. The manuscript now states:

When operating in total particle mode, NAIS-derived number concentrations may differ substantially from reference measurements, with discrepancies of up to an order of magnitude reported relative to SMPS observations. Previous intercomparisons have shown that NAIS often reports higher particle number concentrations than SMPS-based or CPC-based techniques in the sub-10 nm size range; however, these differences are primarily attributed to fundamental differences in detection principles and multiple charging efficiencies rather than to a demonstrated systematic bias of NAIS (Kangasluoma et al., 2020). As a consequence, no generally transferable conversion scheme currently exists between mobility spectrometers and neutral cluster instruments such as NAIS, and the two techniques are often analysed separately, leaving a gap between their operational size ranges.

In this study, we adopt a harmonized approach to construct a continuous particle number size distribution from NAIS and SMPS measurements. Rather than treating the instruments

independently or imposing an absolute correction, the harmonization ensures consistency between the two datasets within a well-constrained overlap region where both instruments operate most reliably. A detailed comparison between NAIS and the reference SMPS was therefore performed at both stations. Scaling factors were derived individually for each measured size distribution based on this overlap analysis and applied uniformly to the NAIS size distributions. The resulting mean scaling factors were 3.38 ± 2.05 for CMN and 3.18 ± 2.07 for JFJ, obtained from size ranges where SMPS measurements exhibit minimum uncertainty and where the impact of scaling on smaller particle sizes is minimized (20–30 nm at CMN and 30–40 nm at JFJ). After scaling, the NAIS and SMPS size distributions were merged at 25 nm for CMN and 35 nm for JFJ (see Fig. S2).

The implications of this harmonization are now explicitly clarified in the manuscript:

This harmonization implies that absolute particle number concentrations are reported across the combined size range of the NAIS-SMPS system. The applied scaling affects the absolute magnitude of the NAIS-derived concentrations, while preserving shape and temporal evolution of the size distributions. Sensitivity tests show that particle number concentrations below the merging diameters can decrease by approximately 30–40% when stronger scaling is applied and can increase by more than a factor of two in the absence of scaling. The sensitivity of particle number concentrations to the applied scaling is quantified in Supplementary Section S2.

A similar harmonization approach, in which scaling factors were derived within a stable overlap region, has been adopted in previous studies combining NAIS with mobility spectrometers (Dada et al., 2023).

Importantly, we now explicitly show that the qualitative conclusions of the study remain unchanged. In particular, the combined contribution of small particles in the intermediate and nucleation mode (2.5–25 nm) remains consistently higher at JFJ than at CMN across all tested scaling scenarios, while CMN remains dominated by the Aitken and accumulation modes. Furthermore, the seasonal and diurnal patterns and the relative contribution of particle size modes and site-to-site contrasts are preserved. These points are now stated explicitly in the concluding paragraph of Section S2.

In the Results section, we have explicitly clarified that absolute concentrations in the smallest size modes should be interpreted with higher uncertainty and primarily in terms of relative patterns. The revised text now reads:

It should be noted that absolute concentrations and relative contributions of the intermediate and nucleation modes are subject to higher uncertainty due to the harmonization of the sub-20 nm size range. A dedicated sensitivity analysis (Supplementary S2) shows that while absolute values vary with the applied scaling, the relative patterns and site-to-site contrasts discussed below remain robust.

We also clarify that the particle formation rate $J_{2.5}$, which is directly derived from particle number concentrations at the smallest sizes, exhibits a comparable sensitivity to the applied scaling. Specifically, the text now says:

Because the particle formation rate $J_{2.5}$ is directly derived from particle number concentrations at the smallest sizes, its absolute values exhibit a comparable sensitivity to the applied scaling (see Supplementary Material Section S2). Removing the scaling increases $J_{2.5}$ by factors of approximately 2–3 at both stations, while the seasonal patterns and the relative differences between CMN and JFJ remain unchanged.

The decision to construct a harmonized dataset also reflects the methodological challenge that different instruments, despite sampling the same aerosol population, often show substantial and poorly constrained discrepancies at the smallest particle sizes.

We believe that these revisions directly address the reviewer's concern by transparently acknowledging the limitations of the scaling approach and by demonstrating, through quantitative sensitivity analysis, that the main conclusions of the study are robust to reasonable variations in the scaling assumptions.

Notification to the authors:
Regarding figure 1: OpenStreetMap images must include visible attribution on images/snippers/captions. Please add the full credit in the figure caption (e.g. "... © OpenStreetMap contributors, https://www.openstreetmap.org/copyright").

We apologize for any confusion. We clarify that OpenStreetMap is not involved in Figure 1. The map imagery is based on Esri data. The figure has been updated to include visible attribution directly on the image, and the full Esri credit is also reported in the figure caption ("© Esri, Maxar, Earthstar Geographics, and the GIS User Community, https://www.esri.com").