

Authors' response to the reviewers

Reviewer's comments in black, Authors' response in blue.

Line numbering in the reviewers' comments refers to the numbering used by the reviewers, i.e., the line numbering from the first submission. Line numbering in the answers referring to the updated text corresponds to the line numbering of the revised manuscript.

Reviewer #1:

General comments:

Schmale et al. present valuable new datasets from balloon-borne meteorological, aerosol, and trace gas measurements across multiple campaigns in the Arctic, Antarctic, and Alpine regions. These observations from remote areas are challenging to obtain and represent an important contribution to the field. The scientific community stands to benefit from such data to improve process understanding, evaluate models, and achieve closure with remote sensing. However, there are some essential elements missing from the data descriptor document and the data itself that should be addressed prior to publication.

One key area for improvement is the inclusion of calibration procedures and data validation. Could the authors clarify the calibration procedure for each sensor and the frequency with which it was performed? Additionally, were common standards used for calibration and comparison? Information on how the mobile measurements compared to stationary, full-size instrumentation would also be valuable. For variables acquired by different instruments, a discussion on their agreement in flight (e.g., total N by CPC vs mSEMS) would strengthen the manuscript.

Response: Thank you for this valuable comment. A general paragraph describing the intercomparison and calibration procedures has been added to the manuscript at the end of the Instrumental section:

“Standard calibration and intercomparison procedures. Depending on the specific setup, each instrument is calibrated before the campaign either in the laboratory or on site. This includes standard calibrations recommended by the manufacturers for each instrument, intercomparisons between instruments measuring the same quantities, such as particle and droplet total number concentrations and size distributions, aerosol light absorption, or trace gas mixing ratios. In addition, when ground-based instrumentation is available at the field site, the MoMuCAMS instruments are compared with the on-site instrumentation. Intercomparisons generally involve operating the entire instrumental payload or individual instruments for several hours alongside the ground-based instrumentation ideally on the same inlet system. The different intercomparison procedures are described in the Supplement (Table S1). An overview of the laboratory intercomparison and calibration results, as well as the intercomparisons with ground-based instrumentation, is also provided in the Supplement (Table S2).” (L210-218)

Further details on the procedures and results are provided in the Supplementary Material, included at the end of this document. The Supplement references the intercomparisons and calibrations that were previously published alongside the results of the respective campaigns and also presents some of the unpublished results.

In addition to the results presented in the Supplementary Material, we would like to note that an intercomparison between the miniCDA and LOAC was performed at HAC2 on Mt. Helmos during the

CHOPIN campaign. However, the LOAC instrument used for this intercomparison was an older version (version 1.2) than the version planned for use in future campaigns. We therefore consider this intercomparison to be of only approximate relevance for the datasets presented here and have not included it in the Supplementary Material. In addition, the newer LOAC version is expected to result in an improved intercomparison. We provide the results here in the response to the reviewer comments.

CHOPIN: miniCDA and LOAC intercomparison at HAC2 on Mt. Helmos. During the CHOPIN campaign, the locally installed LOAC and the miniCDA were run side-by-side at the HAC2 site on 3 November 2024. The two instruments were compared in terms of both total particle number concentration and particle size distribution. For the 12-hour comparison, there were two periods that the station (HAC2) was measuring within a cloud with both instruments detecting droplets (Fig. 1).

Both units were only compared throughout their shared size range, which spans from 0.80 to 40 μm . Although the miniCDA measures particle sizes from approximately 0.66 to 43 μm , the LOAC has distinct size classes that cannot be separated at the same fine size resolution. During the first cloud period (19:00-20:15 November 3), the miniCDA measured higher concentrations than the LOAC, reaching more than 400 cm^{-3} while the LOAC remained generally below 200 cm^{-3} . This difference is particularly pronounced in the first cloud period and may indicate saturation of the LOAC under the high droplet concentrations. During the second cloud period (00:45-01:45 November 4), the difference is much less pronounced and reversed, with the LOAC generally measuring higher concentrations than the miniCDA. The reason for this difference is not yet clear and requires further investigation of the instrument response and sampling conditions.

It is important to note that the LOAC was version 1.2 for this intercomparison which has since been updated to version 2.0 which incorporates improved optics and reduced noise. Results presented here should be considered an intercomparison of the specific LOAC v1.2 configuration used by the University of Stockholm team during the CHOPIN and ArtOfMelt campaigns.

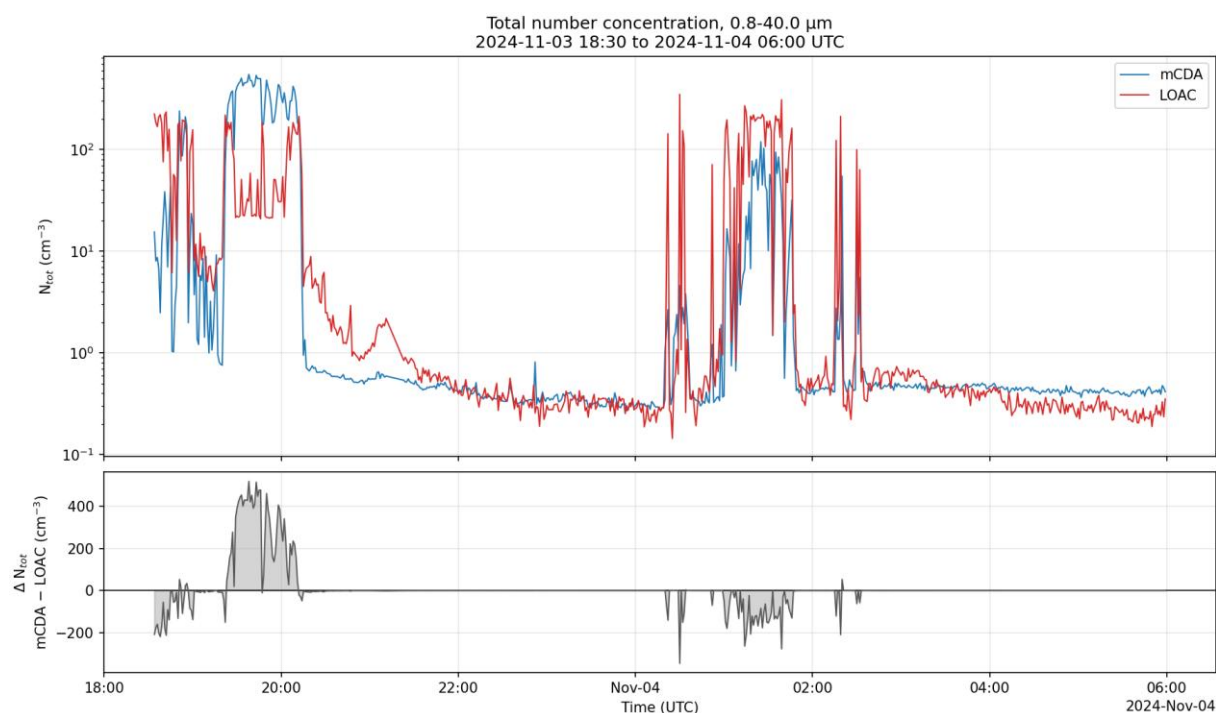


Figure 1. Intercomparison of total number concentrations measured by the miniCDA and LOAC at the HAC2 site between 3 November 2024 and 4 November 2024. The upper panel shows the total particle number concentration measured by the two instruments, while the lower panel shows the number concentration difference between the measurements.

It would be helpful to provide error estimates and discuss sources of error for each variable. This information is important for understanding the accuracy of the sensors in the field and their limitations under certain conditions. For example, the Partector is known to exhibit increased uncertainty at low particle concentrations and sizes, which is particularly relevant in the clean Arctic environment.

Response: We agree with the reviewer that this information is needed and have added Table S4 into the Supplementary Material providing information on the uncertainties for each variable in the datasets.

The missing data sets from the CHOPIN and CLAVIER campaigns must be made available. To ensure reusability, all datasets should be stored in an official data archive that ensures quality assurance, findability, and long-term availability (rather than only on Zenodo) is recommended.

Response: The data sets for CHOPIN and CLAVIER had been made available for the review as ‘Assets’ to the article, because their publication on the ACTRIS database was still in progress. Meanwhile, they have been published under the following links (CHOPIN: <https://zenodo.org/records/20050006>, CLAVIER: <https://zenodo.org/records/20038987>), which are also listed in the manuscript now.

5 out of 9 campaigns have a campaign-specific data archive where the corresponding datasets can be found, ensuring their findability and long-term availability. For the 4 campaigns that do not have a dedicated data archive, the datasets are currently available on Zenodo.

Additionally, please clarify how the filter samples can be accessed.

Response: We agree that information on the accessibility of the filter samples is important. We have therefore added the missing information to the Data Availability section at the end of the manuscript:

“Filter samples from the ALPACA campaign are stored in the EERL freezer and are available upon request. For the ORACLES campaigns, the results from the NEMS-FTIR filter samples collected during both summer seasons will be made available on PANGAEA together with the corresponding flight data in due course. Filter samples from other campaigns are no longer available.” (L562-565)

Please note that the focus of this data descriptor is on the real-time vertical profile data. Physical aerosol samples on various substrates are sparse and little systematic at this point. In the future, we aim for denser chemical information that will then be published and substrates will be kept.

It was also noted that the LOAC data is missing from the ArtOfMelt dataset, and a random check of the PaCE campaign dataset showed unexpectedly high mSEMS_total_N values compared to the Partector and POPS.

Response: The LOAC was flown within MoMuCAMS during the ARTofMELT campaign, but at that time it was not yet integrated into the MoMuCAMS measurement and data-processing framework. The LOAC data were therefore processed by the collaborators responsible for the instrument and are maintained as a separate dataset. Consequently, the LOAC data are not included in the ARTofMELT dataset presented in this manuscript.

Regarding the PaCE campaign with "unexpectedly high mSEMS_total_N values compared to the Partector and POPS" we performed a new quality check on the dataset. As the POPS measures in a different size range, we assumed this is a typo and the reviewer refers to CPC3007. Three flights

exhibited larger total concentrations of mSEMS compared to Partector 2 (Flights 13 and 19) and CPC (Flight 15). Our check revealed that during parts of Flights 13 and 19, some mSEMS bins showed erroneously high values, which had originally been attributed to a later stage of a particle formation event. These were instead caused by overheating of the mSEMS. The affected mSEMS data were therefore removed. For Flight 15, the mSEMS PNSD appears to correctly capture a new particle formation event, which explains the higher mSEMS concentration relative to the CPC. As these very small particles are at the lower edge of the CPC measurement range, the CPC may not have detected a large fraction of them. Thus, for Flight 15, the higher concentration measured by the mSEMS compared to the CPC is considered valid.

What makes me suspicious is the assumption of a constant flow rate for the POPS and miniCDA throughout the flight in the particle concentration calculations. Under changing ambient pressure and temperature, the sample flow is unlikely to remain constant, especially given the pump-control loop dynamics of these instruments. A clarification of how this was addressed or a discussion of the potential implications would help align the methodology with general standards.

Response: The flow rates of both the POPS and miniCDA show some variability during flights, but the variations are small. For example, during a randomly chosen ORACLES flight, 95% of the POPS flow measurements were between 2.97 and 3.01 L/min, while 95% of the miniCDA flow measurements were between 2.788 and 2.811 L/min. Relative to the set flow rates of 3.00 and 2.800 L/min, respectively, these ranges correspond to deviations of less than 1% for the POPS and less than 0.5% for the miniCDA. Since the number of particles sampled per unit time is directly proportional to the volumetric flow rate, the resulting effect on the sampled particle number is of the same relative magnitude, assuming a constant particle number concentration. To avoid introducing additional uncertainty into the concentration data, we therefore use the mean flow rate measured during each flight for the particle concentration calculations.

In addition, the final particle number concentrations are reported at STP. The measured ambient pressure and temperature are therefore accounted for when converting the concentrations to STP conditions. Consequently, although the flow rates exhibit some variability during the flights, the relatively small magnitude of this variability, together with the STP correction, is expected to have only a limited impact on the reported particle number concentrations.

The example analysis section currently focuses primarily on POPS and mSEMS data, with several approaches to plotting number concentration profiles. It may strengthen the manuscript to include examples or references to other instruments and variables.

Response: We thank the reviewer for this suggestion. Given the already substantial length of the manuscript, we have chosen to keep the example analysis section focused on the POPS and mSEMS data. Examples of analyses using other instruments and variables are available in the manuscripts already published from the respective campaigns, to which we refer the reader.

Additionally, the analysis presented may go beyond the intended scope of a data descriptor and could be simplified or supplemented with more background information for clarity.

Response: To provide better context for the examples included in the manuscript, additional background information was added to each example, as suggested by the reviewer.

In section 4.2.1, the following general information was added: *“Differences in aerosol sources, transport and atmospheric mixing between the surface and free troposphere can be investigated using the vertical profiles of aerosol particle number concentrations. Measurements over different environments and seasons can therefore be used to characterize the variability in aerosol vertical structure and identify situations with vertically homogeneous or distinct aerosol populations.”* (L416-419)

In section 4.2.2, the context for the example is introduced with: *“Temperature inversions are an important meteorological feature to consider when trying to understand the vertical distribution and transport of aerosols. Surface-based inversions can suppress vertical mixing, which can lead to differences in aerosol properties near the surface and above the inversion.”* (L443-445).

In section 4.2.3, aerosol-cloud interactions are introduced with *“Aerosol size distributions and concentrations can be substantially modified by cloud processing. When activated into cloud droplets, aerosol particles can undergo aqueous-phase processing, which can change their size and chemical composition. This can lead to characteristic changes in the aerosol size distribution after droplet evaporation, such as the formation of a Hoppel minimum. Comparing aerosol properties below and above clouds can therefore provide insights into the aerosol size ranges that are activated into cloud droplets and the potential sources of aerosols contributing to droplet formation, i.e., whether they are from below or above the cloud layer.”* (L467-472)

Given the current length of the paper and the need to include the essential elements mentioned above, it may be advisable to consider removing the entire data example analysis section 4.2.

Response: We have carefully considered the reviewer’s comment. However, we prefer to retain Section 4.2, as it provides a general overview of the types of analyses that can be performed with the datasets and it shall inspire future users of the datasets, specifically modelers. We consider it important to provide guidance on how to work with the datasets.

Finally, please provide some more details on the Helikite operations and how they were embedded into other measurements. Where were the launch sites located in relation to local stations or adjacent measurements? It would also be very useful if you could link to other datasets or publications that are related to your data.

Response: Following the reviewer’s suggestion, we have added Table S3 to the supplementary material, providing information for each field campaign on the additional data available, the distance from the helikite launch site to other measurement sites, and key references for accessing the data.

Detailed comments:

L31 – 33: The sentence has weird grammar, which gives it the wrong meaning.

Response: Thank you for the comment. To improve clarity, the sentence has been modified in the manuscript to:

“In the Arctic, black carbon has the strongest radiative effect when located in the lowermost part of the atmosphere, while its impact is substantially weaker at higher altitudes (Flanner, 2013). With reference to ACI, the number and size of aerosol particles at the altitude, where clouds form and where particles act as cloud condensation nuclei (CCN) or ice nucleating particles (INP), influence the cloud droplet number concentrations and size distributions as well as ice crystal formation (Carslaw et al., 2013; Kanji et al., 2017).” (L29-34)

Table 1:

- Add “above ground” to the maximum flight altitude, as the elevation of the launch sites differs
“Above ground (a.g.l.)” has been added into the maximum flight altitude header in Table 1.
- The coordinates of CLAVIER are wrong: the longitude has to be 16.641 W

Thank you for noticing, the coordinates of CLAVIER have been corrected in Table 1 of the manuscript; the longitude is now given as 16.641° W.

L115: introduce abbreviation EERL

The full name of the Extreme Environments Research Laboratory (EERL) is introduced at L114.

L116 – 120: Are all instruments installed inside MoMuCAMS? Are the different inlets connected to the main inlet, or are they separate? If separate, please provide details on the cloud droplet sampler inlets.

Response: The instrumental setup inside MoMuCAMS has been clarified in the manuscript by adding the following text at L119-121: *“All instruments are installed inside MoMuCAMS and connected to the main payload inlet, with the exception of the cloud droplet samplers, which have individual inlets. Further details on these inlets are provided below.”*

Further details on the inlets are provided in the Droplet number size distribution section (L155). To improve clarity regarding the miniCDA, L156-158 have been modified to: *“It has a specific inlet oriented vertically extending through the top of MoMuCAMS, providing a flow rate of 2.800 l min⁻¹. To avoid the direct impaction of large, gravitationally settling droplets, a manufacturer-provided perforated inlet cap is installed at the end of the inlet.”*

For the LOAC, details were added at L165-166: *“Its individual inlet is oriented vertically and extends through the top of MoMuCAMS. An inlet cap is used to avoid direct impaction of droplets.”*

L127: Which type of particle charge neutralizer was used with the mSEMS?

Response: A Brechtel Soft X-ray Charger (Model 9002) is used as the particle charge neutralizer for the mSEMS. It uses soft X-rays to neutralize the particle charge rather than a radioactive source. This clarification has been added to the manuscript at L130-131, in the sentence:

“The mSEMS measures the electrical mobility diameter of particles ranging from 8 to a maximum of 300 nm, following charge neutralization with a Brechtel Soft X-ray Charger (Model 9002).”

L154 – 167: Are the cloud droplet probes installed inside the heated MoMuCAMS compartment? If so, how do you treat the effect of droplet evaporation?

Response: All aerosol and droplet counting instruments are installed inside MoMuCAMS. The temperature difference between ambient air and conditions inside MoMuCAMS varies mainly based on outside conditions and can be as large as 30°C. Also relative humidity becomes significantly smaller inside MoMuCAMS. That means that droplet evaporation could indeed be an issue, despite maximally short inlet lines. As shown in Pohorsky et al. (2026), we typically compare the liquid water content derived from the in situ counters with other available observations, e.g., lidar/radar combinations. From that there is no indication that there is a substantial loss of droplets.

L169 – 174: Which type of filter was used with the STAP?

Response: This has been clarified in the manuscript by adding “...using quartz-fiber filters as provided by the manufacturer.” (L173-174).

L204: Please remove the TriSonica if it is not contained in any of the data sets. This gives data users the wrong impression.

Response: The TriSonica has indeed not been reported in any of the datasets presented in this manuscript. However, as it is an instrument that will be used regularly in upcoming campaigns, we would prefer to keep it included in the instrument description so that the description remains applicable to future campaigns as well.

L238: The Wiedensohler inversion is for bipolar diffusion chargers e.g. „radioactive sources”. A different charge distribution has to be used for other neutralizers (see question above)

Response: Thank you for noticing this error, it has been corrected in the manuscript to “For the mSEMS, a manufacturer-provided inversion routine is applied to the raw particle counts...” (L239-240).

L266: The CPC 3007 provides inlet pressure readings.

Response: Retrieving the inlet pressure data requires a dedicated software and a connection to a computer, which were not available to us during the past campaigns. Consequently, our data processing routine was developed based on the CPC3007 data without pressure readings. For future campaigns, we will take this information into consideration.

L292: On which pressure sensor is the altitude calculation based?

Response: Thank you for pointing out that this was not specified in the manuscript. The pressure sensor used for the altitude calculation has been clarified by adding:

“The pressure used for this calculation is taken from the sensor designated as the reference, which is typically the flight computer (Bosch BME280 Humidity & Pressure Sensor). If pressure data from the flight computer are not available, pressure measurements recorded by another measurement instrument installed in MoMuCAMS can be used instead.” (L295-297)

L304 – 309: Please remove the sonic if it is not contained in any data set

As mentioned above, the sonic has indeed not been reported in any of the datasets presented in this manuscript. However, as it is an instrument that is an integral part of the payload and it will be used regularly in upcoming campaigns, we would prefer to keep it included in the instrument description so that the description remains applicable to future campaigns as well.

L318 – 322: The detection limit of the CPC 3007 is known to be sensitive to ambient temperatures (<https://doi.org/10.1016/j.jaerosci.2021.105877>). How did you ensure adequate working temperatures?

Response: Although the instrument is known to be sensitive to ambient temperature, this effect has primarily been reported at elevated temperatures. Bezantakos and Biskos (2021; <https://doi.org/10.1016/j.jaerosci.2021.105877>) showed that increasing temperature raises the minimum detectable particle diameter, which may affect new particle formation measurements in warm environments. In our studies, temperatures were predominantly below or close to 0°C, and we found no evidence that the performance of the CPC3007 was affected. For the most recent campaign, this is further supported by the good agreement in total particle number concentrations measured by the CPC3007, operated as the reference ground instrument, the CPC at the Neumayer Air Chemistry Observatory (SPUSO), and the mSEMS.

L321 – 322: How did you derive counting efficiencies, and how did you correct them?

Response: The procedure has been clarified by adding the following to the end of the sentence:

“Counting efficiency correction factors are applied at this step, if necessary and possible, based on intercomparison with a reference instrument.” (L324)

L328: The $dN/d\log D_p$ should be derived from the normalized bin width using the upper and lower bin limits.

Response: Thank you for noticing this imprecision. The sentence has been clarified in the manuscript to:

“The aerosol number size distribution in $dN/d\log D_p$ is derived by dividing the bin concentrations by the normalized bin width $d\log D_p$, calculated from the upper and lower diameter limits of each bin.” (L329-331)

L333 – 334: How did you differentiate larger aerosol particles from cloud droplets? Why is the LOAC data not mentioned here?

Response: Larger aerosol particles are generally expected to activate into cloud droplets under cloud conditions. In our measurements, this is reflected by a strong depletion of larger aerosol particles when entering a cloud, allowing the cloud droplet population to be distinguished from the aerosol population. This is clearly visible in Figure S3 of the Supplementary Material, where, within the cloud, the miniCDA measures substantially higher droplet number concentrations than the particle number concentrations measured by the POPS. If necessary, the number concentration of larger aerosol particles can be estimated from the POPS measurements and accounted for when interpreting the droplet counts. Typically, we did not or only rarely see particles > 500 nm. It was hence not necessary to make a correction for the datasets presented here. This has been clarified in the manuscript by adding:

“Within clouds, larger aerosol particles are generally expected to activate into cloud droplets, resulting in a strong depletion of the larger aerosol population. If necessary, the contribution of residual larger aerosol particles can be estimated from the POPS measurements.” (L341-343)

The LOAC is not mentioned here because it has so far only been used during the ARTofMELT campaign, for which the data were processed by the group responsible for the instrument and provided to us as processed data. We therefore do not currently have the processing steps for the LOAC within our data processing framework.

L355 – 357: Please consider general relative humidity effects on filter-based absorption photometers (<https://doi.org/10.5194/amt-12-5879-2019>). The spikes in humidity inversions must be excluded before processing with the manufacturer's software. Otherwise, artificial spikes at humidity inversions affect all values within the 2 min moving-average window, resulting in negative values (as seen in Figure A1). The STAP data probably needs reprocessing.

Response: Thank you for pointing out Düsing et al. (2019). We are aware of the impact of RH on the absorption readings. Our instruments are positioned behind heated inlets to reduce RH consistently to < 40% RH as suggested by ACTRIS. To exclude the impact of high ambient RH, we screened the microAeth data for coincidences with humidity inversions per the reviewer's suggestion. However, we commonly notice across our campaigns that negative absorption values, seen with STAP and microAeth, occur predominantly during periods of very low aerosol loading rather than during humidity inversions. For example, during the CHOPIN campaign the negative fraction at 625 nm goes from 85% when $N_{186-3370} < 5 \text{ cm}^{-3}$ to 0% when $N_{186-3370} > 100 \text{ cm}^{-3}$. This strong dependence on aerosol loading, together with the lack of consistent coincidence between negative absorption values and RH inversions, suggests that the negative values primarily reflect the instruments reaching their limit of quantification, where statistical noise in the signal produces small negative absorption values independent of any RH-driven artifact. For future campaigns, we will have a diffusion dryer connected to the heated inlet that will remove relative humidity from the sampled air.

L376 – 378: How did you treat pollution flagging for an mSEMS scan? Is the entire scan flagged when a part of the scan shows pollution influence identified with the POPS at a higher time resolution?

Response: In most cases, the pollution flags are derived from an instrument with a higher time resolution than the mSEMS. Therefore, a short pollution event identified by the higher-resolution instrument may occur during only part of an mSEMS scan. In some cases, a short pollution event may not be detected by the mSEMS, as the instrument may be measuring a different part of the particle size distribution at that time. During the flagging procedure, the mSEMS data are manually inspected to determine whether the pollution event is also evident in the single mSEMS scan. If so, the timestamp corresponding to the midpoint of the mSEMS scan is ensured to be flagged as polluted. Thus, the entire mSEMS scan is not automatically flagged based solely on a pollution event detected by the higher-resolution instrument.

L397: typo: multiplayer instead of multilayer

Corrected. Thank you for noticing.

Table 3:

- Pressure was taken from which sensor? Has been clarified earlier in the manuscript (L295-297).

- What are the correction factors for POPS, and where are they from?

Response: The correction factors for the POPS are campaign-specific. When needed, they can be derived from a comparison with a reference ground instrument or, when available, from the overlapping size ranges of another MoMuCAMS instrument. To clarify this in the table, all entries referring to a correction factor have been changed to “*Apply correction factors if needed.*”

- mSEMS range is inconsistent with the text and plots (8 to 280 nm)

Response: The reviewer correctly pointed out that the stated upper limit of 250 nm is inconsistent with the following text and figures, where an upper limit of 280 nm was used. However, the upper limit of the size distribution depends on the instrument settings used for each campaign. We therefore suggest using 300 nm as the general upper limit, as described in L131, while noting that the actual upper size limit may vary between campaigns. The corresponding value has been corrected in Table 3 to “*Total particle number concentration $N_{8-300max}$* ”.

- MA200: How do you derive 10s data from a raw file of 1 min resolution?

Response: This was a mistake in the stated time resolution. Thank you for noticing. It has been corrected in the manuscript to “*1 min resolution*”.

- Please add the measurement uncertainty for all variables! A new table providing the measurement uncertainty for each variable has been added into the Supplementary Material (Table S4).

- What about the LOAC, and how did you treat its 30s resolution?

Response: As mentioned above, the LOAC has so far been used only during the ARTofMELT campaign, and its data were processed by the group responsible for the instrument. As for the other instruments with lower time resolution, the LOAC measurements are reported at their native time resolution, with one measurement value reported for each 30 s timestamp. However,

to maintain consistency throughout the manuscript, we have included a corresponding line in Table 3 as a reference for future LOAC measurements within the MoMuCAMS setup.

L436 – 437: The sentence is slightly inaccurate. The temperature profile is insufficient to quantify vertical aerosol transport.

Thank you for pointing this out. The sentence has been clarified in the manuscript to “*The temperature profiles recorded during the Helikite flights allow for the determination of inversions and, when combined with the aerosol measurements, provide insight into aerosol transport across these inversion layers.*” (L445-447)

Section 4.2.2

- The plot and normalization, as well as their interpretation, go beyond the scope of a data descriptor.

Response: We agree that this example is relatively specific and goes beyond what would be required for a data descriptor alone. However, our intention in this section is to provide concrete examples of how the dataset can be used in different ways and to illustrate our approach to analyse the data. We therefore prefer to keep this example in the manuscript, as we believe that the more detailed example can provide ideas and demonstrate possible applications of the dataset to users.

- More background and accompanying material would be necessary to comprehensively.

Response: As mentioned above, additional background information has been added to each of the examples described in this section to provide better context and facilitate their interpretation.

L470: slightly incorrect: on 7 June, the accumulation mode is visible above and below the cloud, so it's not “additional”.

Response: Indeed, thank you for pointing this out. The word “*additional*” has been removed from the sentence.

Reviewer #2:

General Comments

The manuscript titled "Vertical observations of aerosol properties in the Arctic, Antarctic and Alpine atmospheric boundary layers with a tethered balloon" presents a multi-year observational effort to fill the data gap regarding vertical aerosol distributions in the lowermost kilometer of the atmosphere. Utilizing the Modular Multiplatform Compatible Atmospheric Measurement System (MoMuCAMS) aboard a Helikite platform, the authors provide a dataset from approximately 300 flights across nine polar and mountain regions between 2021 and 2025.

By documenting the "hElikite dAta proceSsing code" (EASE), the manuscript offers a traceable, standardized workflow for processing heterogeneous atmospheric data into quality-controlled products. The manuscript follows the Earth System Science Data (ESSD) technical descriptor format, logically transitioning from scientific motivation to campaign logistics and instrument specifications. The organization of campaigns into Arctic, Antarctic, and Alpine clusters provides a useful framework for comparative analysis. The technical workflow, particularly the clear definition of data levels (L0 to L2) according to EBAS standards, ensures high interoperability.

Minor Revisions

Page 7, Line 155 (Sampling Inlet Efficiency): At low flow rates, inertial losses for particles larger than 1 μm can be substantial, particularly if the platform is not perfectly aligned with the wind. The manuscript would benefit from a theoretical estimate or a discussion of the aspiration efficiency for the largest POPS and miniCDA bins under typical campaign wind speeds and orientation misalignments.

Response: As the tethered balloon orients with the wind, the orientation of the payload is determined by the balloon, such that the inlet is generally facing directly into the wind direction. The balloon and inlet realign with changes in wind direction within a few seconds. Only in the absence of wind or when the wind speed is below the detection limit of 1 m/s the balloon rotates slowly. In such cases, however, there is no well-defined wind direction relative to which the inlet could be misaligned. Such conditions are however relatively rare during the measurements (occurred for 7 out of 68 flights during the latest ORACLES campaign). The inlet sampling efficiency and transmission losses of the MoMuCAMS payload have been characterised and are described in detail by Pohorsky et al. (2024) for particles between 8 and 3000 nm, thus including the largest POPS bins. The transmission efficiency for particles between 50 and 1000 nm was found to be very close to 100%, while smaller particles are affected by diffusional losses and larger particles by gravitational deposition. However, these losses were typically less than 10%. This information has been clarified in the manuscript as follows:

"The instruments as well as the inlet sampling efficiency and transmission losses have previously been described and characterized in Pohorsky et al. (2024 and references therein)." (L115-116)

The miniCDA has a dedicated inlet oriented vertically as per manufacturer design and extending through the top of MoMuCAMS. To avoid the direct impaction of large, gravitationally settling droplets, a manufacturer-provided perforated inlet cap is installed at the end of the inlet. The inlet is relatively short (12 cm). However, neither the transmission losses of the inlet itself nor any additional losses associated with its vertical orientation have been characterized. To reflect this, we have included a note in the meta data of the data publication.

Table 1:

- CLAVIER Longitude: Correct the longitude for CLAVIER to 16.641° W.
Thank you for noticing, the coordinates of CLAVIER have been corrected in Table 1 of the manuscript; the longitude is now given as 16.641° W.
- Elevation Labeling: Clarify if "elevation" refers to the station's base height above sea level (a.s.l.) and specify if "maximum flight altitude" is recorded in a.s.l. or above ground level (a.g.l.).
Response: Thank you for pointing this out. The inconsistency has been corrected in Table 1. The station elevation is given as altitude above sea level (a.s.l.), and the incorrect "*above ground level*" designation has been removed. For the maximum flight altitude, "*above ground level (a.g.l.)*" has been added to the table header to clarify that this value is reported relative to the ground.

References

- DOIs: Where possible, please add a DOI to every reference. Specifically, provide more information for Patel and Divecha and Lenschow et al. to ensure these references are discoverable.
- Peer-Reviewed Sources: Please prioritize peer-reviewed publications where available. For example, update the citation for Pasquier et al. to the following: <https://doi.org/10.1175/BAMS-D-21-0034.1>.
Response: Thank you for noticing. The missing links have been added and the publications updated if needed.

- 1 *Supplement of*
- 2 **Vertical observations of aerosol properties in the Arctic, Antarctic**
3 **and Alpine atmospheric boundary layers with a tethered balloon**
- 4 **Yolanda Temel et al.**
- 5 *Correspondence to:* Julia Schmale (julia.schmale@epfl.ch)
- 6 The copyright of individual parts of the supplement might differ from the article license.

7 **Table S1: Overview of the different intercomparison and calibration methods for each type of measurement**
8 **(note results are shown in table S2).**

Measurement type	Description of intercomparison and calibration methods
Particle number concentration and size distribution instrumentation (mSEMS, aMCPC, CPC3007, Partector 2, POPS)	The laboratory Scanning Electrical Mobility Spectrometer (SEMS Model 2100, Brechtel Manufacturing Inc.) serves as the main reference instrument for verification of particle number concentration and size distribution measurements. The specific verification procedures performed depend on the instruments included in the campaign and may include: (a) Comparison of the different instruments with the SEMS reference to assess particle sizing and number concentration measurements. (b) Intercomparisons between multiple units of the same instrument type to assess number concentration measurements (mSEMS, aMCPC, CPC3007, Partector 2 and POPS). (c) Comparisons between different instrument types with overlapping size ranges to assess particle sizing (mSEMS, Partector 2 and POPS). (d) Size-selection experiments using Polystyrene Latex (PSL) or ammonium sulfate aerosols to assess size-resolving accuracy and number concentration measurements. (e) Zero tests using particle-free air downstream of a HEPA filter. If a particular unit shows deviating results (typically a difference exceeding 10%), the issue is addressed.
Droplet number concentration and size distribution instrumentation (miniCDA, LOAC)	As an initial verification, the miniCDA total number concentration and size distributions were checked using MonoDust measurements and compared with measurements from two similar optical aerosol spectrometers: a fine dust measurement device (Fidas® Frog, Palas) and the POPS. The two droplet counters (miniCDA and LOAC) have not yet been intercompared. More systematic work is required.
Aerosol absorption property instrumentation (STAP, microAeth)	Time series from simultaneous measurements of laboratory air are compared between the STAP and microAeth to assess the agreement between the two instruments.
Trace gas instrumentation (CO ₂ , Pico CO, Ozone)	The CO₂ sensor calibrations are performed in the laboratory with reference air mixtures with CO₂ mixing ratios of 400 and 800 ppm. A cap is inserted on the sensor's chamber and the air mixture is flushed through it for 10 minutes. After applying STP corrections based on the laboratory's environmental conditions, the measured values are compared to the true mixing ratios and a correction factor is applied if necessary. For the O₃ monitor, a zero check is performed at the beginning of each campaign with an O₃ scrubber. The instrument's parameters are then adjusted to recalibrate the true zero. If another O₃ monitor is available during a campaign, the instrument is installed on the same inlet before and after the campaign to assess its comparability. The CO monitor is operated on the ground before each flight in its "dual mode" (Pohorsky et al., 2024). In dual mode, the instrument alternates between two different inlets. A CO scrubber placed in front of the second inlet allows the instrument to maintain a continuous true zero

	measurement, avoiding any drift in the instrument's measurement. These baseline values are recorded before and after each flight to correct the data during flights. Additionally, whenever another CO monitor is available during a campaign, a direct comparison on the ground is performed at the beginning and the end of the campaign.
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10 Table S2: Overview of the intercomparison results available for each campaign. Both published and unpublished results are summarized below. Figures cited without
11 an external reference are provided in the following sections of this Supplementary Information.

Campaign	Ground based reference	Particle number concentration and size distribution	Droplet number concentration and size distribution	Aerosol absorption properties	Trace gases
Brigerbad	✗	Total number concentration comparison of aMCPCs with reference MCPC (agreement within 5%) and counting-efficiency test (Pohorsky et al., 2024, Figs. S4-S5). mSEMS sizing tests via size-selection experiment (deviation below 7%) and comparison with SEMS (Pohorsky et al., 2024, Fig. 4, Tables 2 and S1). POPS sizing tests via size-selection experiment and comparison of total number concentration with MCPC reference (deviation below 10%) (Pohorsky et al., 2024, Figs. S6-S7).	-	No intercomparison available.	CO baseline measurement checks (Pohorsky et al., 2024, Fig. 7). Ozone zero calibration performed before each campaign using an ozone scrubber.
ALPACA	✓	mSEMS, POPS and SEMS intercomparison (agreement within 10% for mSEMS-SEMS and 20% for POPS-SEMS; Pohorsky et al., 2024, Fig. 6). mSEMS, POPS and SEMS intercomparison using MoMuCAMS measurements averaged over the lowest 2 m of	-	No intercomparison available.	CO₂ sensor calibration for determination of the correction factor. CO baseline measurement checks (Pohorsky et al., 2024, Fig. 7) and ground-based intercomparison (agreement within 8%) (Fig. S5).

Campaign	Ground based reference	Particle number concentration and size distribution	Droplet number concentration and size distribution	Aerosol absorption properties	Trace gases
		each profile and corresponding ground-based measurements (agreement within 10%) (Pohorsky et al., 2025, Fig. S3).			Ozone zero calibration performed before each campaign using an ozone scrubber.
PaCE	✓	In-flight intercomparison of Helikite particle counters (mSEMS , CPC3007 , and Partector 2); pairwise comparisons show good agreement.	-	No intercomparison available.	CO₂ sensor calibration for determination of the correction factor.
ARTofMELT	✓	Ship-based Differential Mobility Particle Sizer (DMPS) and POPS intercomparison with applied correction factor (Pohorsky et al., 2026, Fig. S2, Table S2).	Side-by-side deployment of the LOAC and reference instrument (FM120, Droplet Measurement Technologies, USA) in two fog events for quantitative comparison (agreement within 20-30%) (Pohorsky et al., 2026, Fig. S3).	No intercomparison available.	-
GreenFjord	✓	mSEMS and SEMS intercomparison (Alden et al., 2026a, Fig. S3)	-	STAP and microAeth intercomparison (Alden et al., 2026a, Fig. S1)	CO₂ sensor calibration for determination of the correction factor. Ozone zero calibration performed before each campaign using an ozone scrubber.
Turtmann	✗	The two mSEMS units agree within 2% in total number concentration and within 3 nm in particle diameter; mSEMS particle sizing agrees within 4%	-	STAP and microAeth intercomparison showing good agreement (Fig. S4).	CO₂ sensor calibration for determination of the correction factor.

Campaign	Ground based reference	Particle number concentration and size distribution	Droplet number concentration and size distribution	Aerosol absorption properties	Trace gases
		with the size-selected measurements (Fig. S1). Good agreement between POPS101 and POPS105 ; strong undercounting (50-60%) in intercomparison with SEMS.			
CLAVIER	✓	mSEMS and ground-based Mobility Particle Size Spectrometer (MPSS) intercomparison (deviation of 15% for total number concentration over common size range and 18 nm for sizing; MPSS measuring broad diameter range).	miniCDA particle sizing agrees well with the selected particle diameter (1.28 μm).	STAP and microAeth intercomparison showing good agreement (Fig. S4).	-
CHOPIN	✓	The two mSEMS units agree within 2% in total number concentration and within 3 nm in particle diameter; mSEMS particle sizing agrees within 4% with the size-selected measurements (Fig. S1). mSEMS and POPS intercomparison show good agreement in non-cloud flights (slope = 0.78, $R^2 = 0.95$); POPS undercounting in cloud (slope = 0.67) (Alden et al., 2026b).	-	STAP and microAeth intercomparison showing good agreement (Fig. S4).	CO₂ sensor calibration for determination of the correction factor.

Campaign	Ground based reference	Particle number concentration and size distribution	Droplet number concentration and size distribution	Aerosol absorption properties	Trace gases
ORACLES	✓	Good agreement in total number concentration between CPC3007, mSEMS and ground-based CPC3775; mSEMS and ground-based SMPS intercomparison (deviation of 20% for total number concentration over common size range and 20 nm for sizing; SMPS measuring broad diameter range). Good agreement between POPS101 and POPS105; strong undercounting (50-60%) in intercomparison with SEMS. POPS462 and SEMS intercomparison (total number concentration agreement within ~10% over common size range) (Fig. S2). mSEMS and POPS462 agree within 2%.	Good agreement between POPS and miniCDA under cloud-free conditions with low concentrations of particles above 670 nm. Within the cloud, miniCDA droplet concentrations exceed POPS particle concentrations by at least one order of magnitude (Fig. S3).	-	-

For several campaigns, the intercomparisons have been published alongside the scientific results. The corresponding references can be found in Table S2. Below, we present additional intercomparisons that were not included in the publications, as well as intercomparisons for which the corresponding work has not yet been published.

mSEMS: The following provides an example of the intercomparison procedure used to characterize and validate the mSEMS deployed during the Turtmann, CHOPIN, CLAVIER, and ORACLES campaigns. The total particle number concentrations of laboratory air measured by the two mSEMS units on 5 June 2025 over 5 hours show good agreement, with a linear regression slope of 0.98 ($R^2 = 0.98$), corresponding to a 2% deviation from a 1:1 relationship (Figure S1a). When comparing the measured size distributions, the particle diameters agree within 3 nm based on the fitted lognormal curves (Figure S1c). For the size-selection experiments on 31 January 2024, Polystyrene Latex (PSL) aerosols were nebulized and size-selected using the Differential Mobility Analyser (DMA) of the laboratory reference Scanning Electrical Mobility Spectrometer (SEMS Model 2100, Brechtel Manufacturing Inc.). The particle sizing by the mSEMS agrees within 4% with the particle diameters selected by the reference DMA, with regression slopes of 0.96 and 0.97 ($R^2 = 1.00$) (Figure S1b).

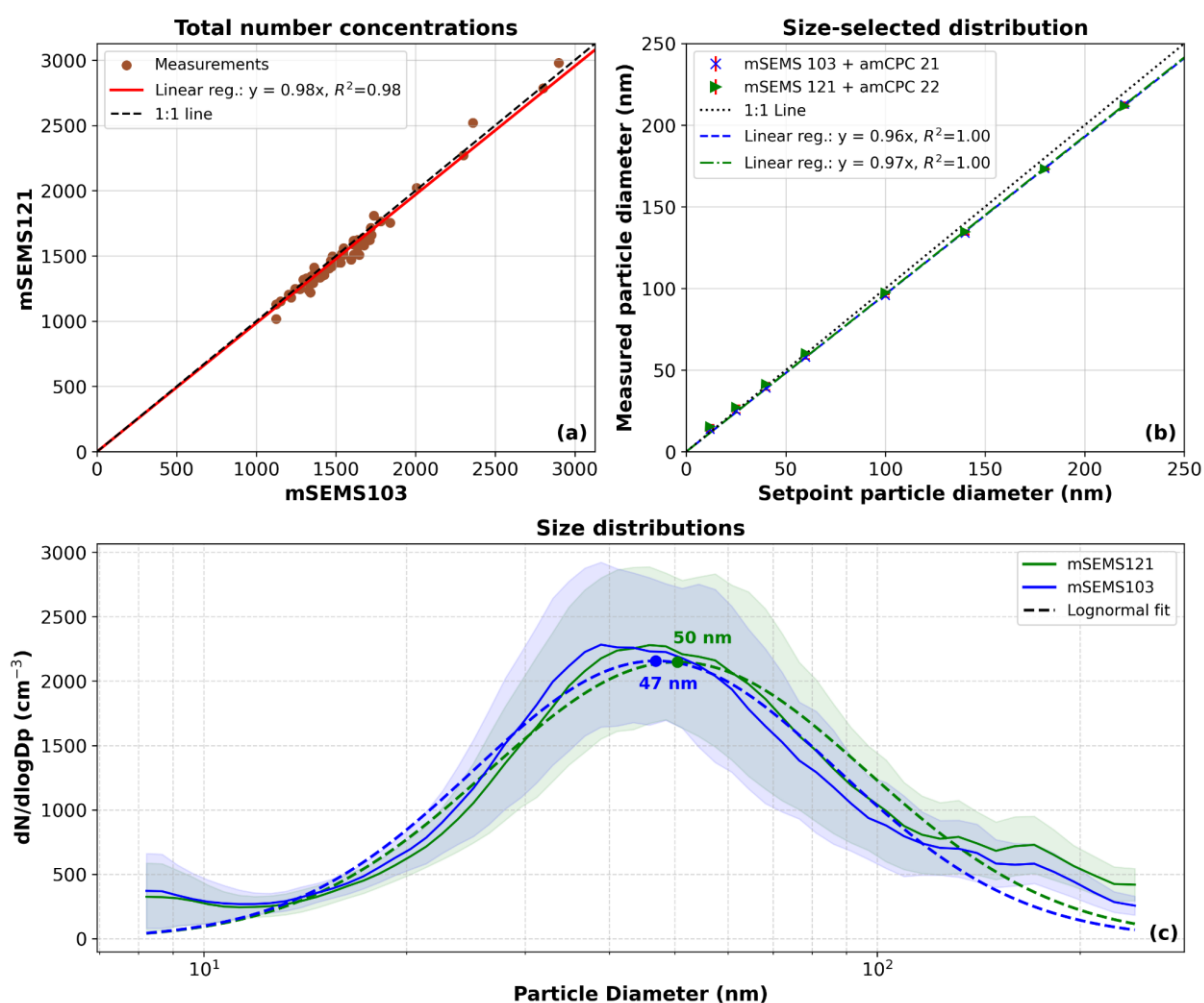


Figure S1: mSEMS intercomparison results. (a) Comparison of total particle number concentrations measured by two mSEMS units on 5 June 2025. (b) Comparison of mSEMS particle sizing with size-selected polystyrene latex (PSL) particles using the reference DMA on 31 January 2024. (c) Comparison of particle size distributions measured by the two mSEMS units on 5 June 2025.

POPS: The new POPS462 unit, used during the latest ORACLES campaign, shows good agreement with the SEMS for laboratory air measurements (22 January 2025), both in terms of total particle number concentration and size distributions. When integrating over the common size range, POPS462 is found to underestimate the total particle number concentration by around 10% compared with the SEMS (Figure S2).

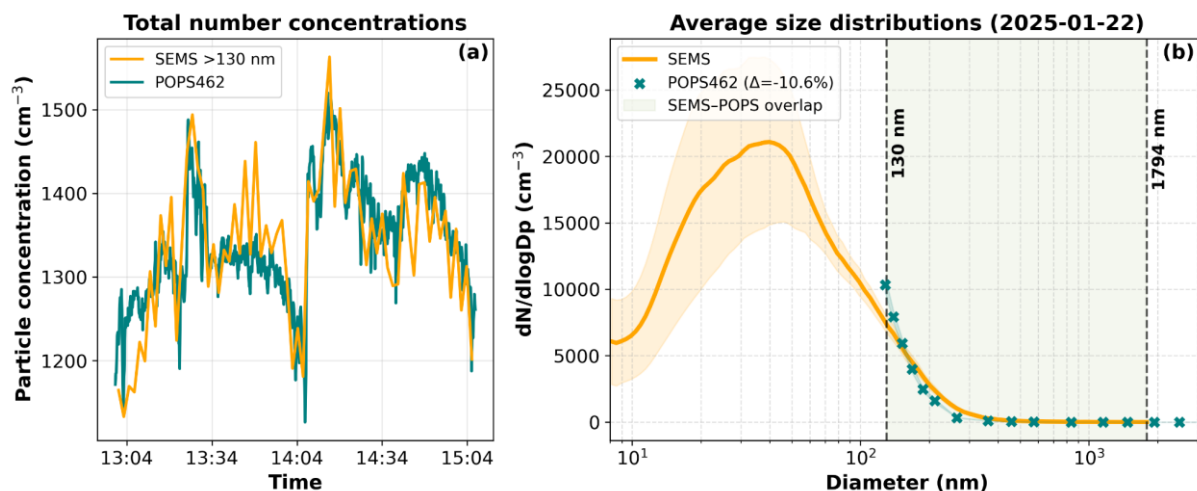


Figure S2: Intercomparison of the EERL SEMS and POPS462 showing (a) total particle number concentrations measured by both instruments above 130 nm and (b) particle size distributions and the corresponding differences in integrated total particle number concentrations over the overlapping diameter ranges.

The two older POPS units 101 and 105 also show good agreement with each other in terms of both total particle number concentrations and size distributions. However, when comparing them with the mSEMS in the field or the EERL SEMS, both POPS units appear to undercount particle total number concentrations by up to 50-60%. Any identified undercounting and, where applicable, the corresponding correction have been specified in the metadata of the respective datasets.

miniCDA: As an initial verification prior to the CLAVIER, CHOPIN and ORACLES campaigns, the miniCDA total particle number concentration and size distributions were assessed using MonoDust 1500 (silicon dioxide particles with mean diameter of 1.28 μm) measurements and compared with measurements from a fine dust measurement device (Fidas® Frog, Palas, same manufacturer as for the miniCDA) and the POPS in July 2024. As expected, a primary peak is visible around 1.3 μm , with a secondary peak between 1.8 and 1.9 μm , which is attributed to a small fraction of larger particles in the MonoDust. The high size resolution of the miniCDA measurements allows the different particle size modes to be clearly resolved. The miniCDA seems to provide a higher size resolution than the Fidas® Frog. A direct intercomparison with the POPS is more challenging because the POPS has a considerably lower size resolution, with only eight size bins, and cannot measure particles larger than 3 μm . Nevertheless, the POPS also shows a matching peak around 1.3 μm , as well as an additional peak around 0.74 μm , which is attributed to the ambient aerosol contribution. These results demonstrate that the miniCDA detects the expected particle size distribution and their relative changes.

In the absence of laboratory measurements of cloud droplets for the miniCDA, a suitable cloud measurement from the ORACLES 2024-2025 campaign is used for a field-based intercomparison. Below the cloud (Figure S3, yellow), the POPS and miniCDA show good agreement in both the measured size distributions and the total particle number concentration integrated over their overlapping size range. Although the miniCDA is a cloud droplet counter, it also detects particles larger than 670 nm. The integrated total particle number concentrations over the overlapping size range of the POPS and miniCDA are in very close agreement. Particles larger than 600 nm generally occur at relatively low concentrations at Neumayer III and are therefore not expected to significantly affect the cloud droplet number concentrations during cloud measurements if they are not activated into cloud droplets. Indeed, the droplet number concentrations measured within the cloud are at least an order of magnitude higher than the particle number concentrations for the overlapping size range (Figure S3, blue). The near-zero concentrations measured by the POPS are attributed to its near-interstitial inlet, which, as shown here, does not measure cloud droplets unless they are very small.

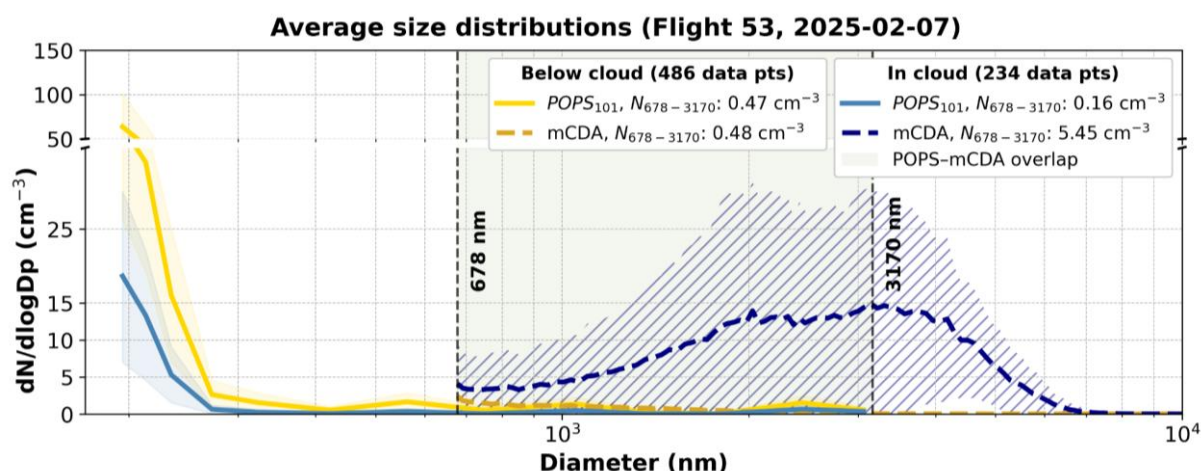
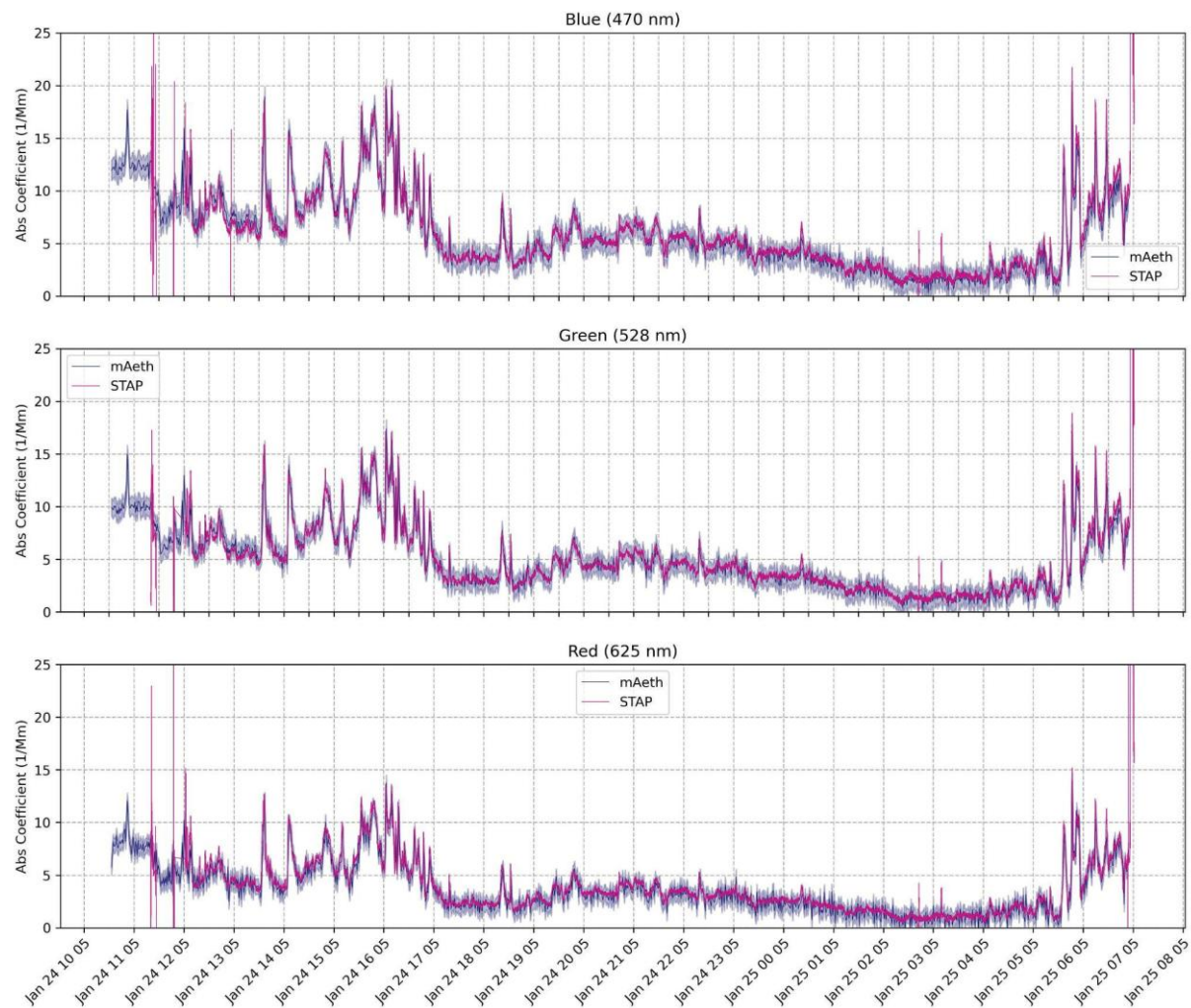


Figure S3: Intercomparison of the size distributions above 170 nm during Flight 53 (summer season 2024-2025, 2025-02-07), showing good agreement between the POPS and miniCDA in both the measured size distributions and total particle number concentrations below the cloud. As expected, within the cloud, the miniCDA measures substantially higher droplet number concentrations than the particle number concentrations measured by the POPS.

74 **STAP and microAeth:** Absorption coefficients at 470, 528, and 625 nm were measured by the microAeth and
75 STAP on 24-25 January 2024 over 22 hours using laboratory ambient air. The intercomparison shows very good
76 agreement for all three wavelengths (Figure S4).



77
78 **Figure S4:** Intercomparison of absorption coefficients at 470, 528, and 625 nm measured by the microAeth and STAP in
79 laboratory ambient air. The measurements show very good agreement.

CO₂ sensor: The CO₂ sensor is calibrated using gas mixtures with known CO₂ mixing ratios of 400 and 800 ppm. The raw sensor measurements are first corrected to STP conditions and then compared with the corresponding known mixing ratios. A linear regression is performed between the measured and reference CO₂ mixing ratios to characterize the sensor response. The resulting regression relationship is used to derive correction factors, which are subsequently applied to the measurements. The corrected CO₂ mixing ratios are then compared with the known values of the calibration gases to assess the performance of the sensor and the effectiveness of the applied correction.

CO sensor: During ALPACA, a ground-based comparison was performed between the Mira Pico CO monitor and a reference monitor deployed at one of the study's measurement site downtown (Community and Technical College). The two instruments sampled from the same Teflon inlet for a period of 40 hours between January 20 and January 22, 2022. The results of the intercomparison are presented in Figure S5. A linear regression between the two sets of measurements for a time resolution of 1 minute indicate a slope of 0.92 and an adjusted R² of 0.95, indicating a very good match, within the instruments' uncertainty.

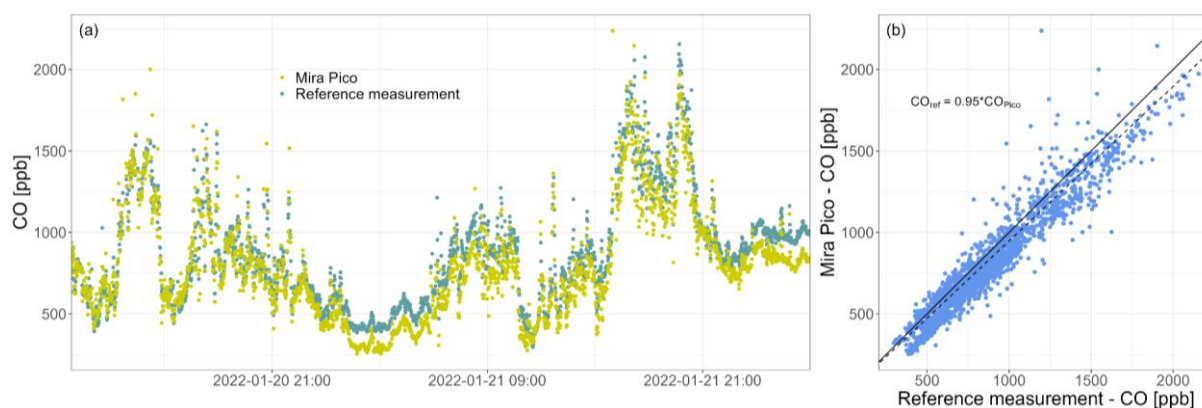


Figure S5: Intercomparison of the Mira Pico with a reference gas filter correlation analyzer at the CTC measurement site in Fairbanks between 2022-01-20 at 09:48 and 2022-01-22 at 03:00 AKST. (a) Time series of CO measurements at 1-min resolution for each instrument. (b) Scatter plot of Mira Pico measurements against the reference instrument. The solid black line represents the 1:1 line. The dashed line represents the slope of the linear regression.

Intercomparisons with near-by ground-based stations are valuable for assessing the consistency of the Helikite measurements with established long-term observations. However, their interpretation can be challenging because differences in instrumental configurations, operating settings, and particle size ranges covered by the instruments can contribute to discrepancies between the measurements, particularly when the instruments are not operated under identical conditions. In addition, the intercomparisons are not necessarily performed at exactly the same location, as the Helikite measurement position may be some distance from the ground station. Spatial variability in aerosol properties may therefore also contribute to differences between the measurements. Finally, the ground-station instruments are typically calibrated only once per year due to the remoteness of the stations, whereas our instruments deployed during the campaigns may have been calibrated and intercompared in the laboratory more recently. Differences in calibration age may therefore also contribute to the observed intercomparison differences.

PaCE: In-flight particle counter intercomparison and surface intercomparison with the ACTRIS Sammaltunturi station. The total particle number concentrations measured by the Helikite particle counters (mSEMS, CPC3007, and Partector 2) were intercompared throughout the PaCE campaign flights. No individual flight has enough data points recorded for all three instruments to allow a three-way comparison. Therefore, the instruments were compared in pairs. Overall, good agreement was found between the instruments.

The intercomparison was further divided into cloudless and cloudy data points. For the cloudless cases, each pair of instruments could be intercompared three times. The differences were below 10% in most cases, with the CPC3007 tending to show lower total particle number concentration in comparison to the mSEMS. The Partector 2 was generally found to slightly overestimate the total particle number concentration compared with both the mSEMS and CPC3007. However, for the cloudy cases, two of three intercomparisons between the Partector 2 and mSEMS showed overestimation of up to 50%, while one of the two intercomparisons with the CPC3007 showed overestimation of up to 100%. Therefore, the Partector 2 appears to significantly overestimate the total particle number concentration within clouds. This pronounced difference in Partector 2 performance between cloudless and cloudy conditions can be attributed to the high humidity within the cloud, which is consistent with the manufacturer's recommendation not to use the instrument under high-humidity conditions. In cases where the overestimation was particularly pronounced and the Partector 2 measurements were inconsistent with those of both the CPC3007 and mSEMS, the Partector 2 data were excluded from the dataset.

In addition, the Helikite payload was operated for 10 hours next to the Sammaltunturi station, which is equipped with a Differential Mobility Particle Sizer (DMPS, range 10-800 nm) and an Aerodynamic Particle Sizer (APS, 0.5-20.5 μm), to intercompare particle size distribution measurements. The Sammaltunturi setup and instruments are described by Beckman et al. (2025). The APS and POPS show good agreement, with particle number concentrations below 2 cm^{-3} per bin above 1 μm . The overall structure of the particle size distributions also agrees well between the DMPS and mSEMS, with a Hoppel minimum centered around 50 nm during the intercomparison period. However, the measured concentrations differ substantially, with a mean difference of up to 72% between the mSEMS and DMPS measurements, particularly in the central size range (28.8-112.2 nm). The two tails of the distributions show better agreement, with a difference of 2.3% at 10.0 nm and 33% (-12.8 cm^{-3}) at 238.8 nm. Since the total particle number concentrations measured by the mSEMS were found to agree with the CPC3007 and Partector 2 onboard the Helikite, the difference might be attributed to an overestimation by the DMPS at the Sammaltunturi station.

ORACLES: Intercomparison with instrumentation from the Neumayer III Air Chemistry Observatory (SPUSO). During surface filter sampling, the entire payload is operated together with the filter sampler to obtain aerosol and meteorological information for the collected samples. These surface measurements can also be used for intercomparisons with the ground-based aerosol instrumentation at the Neumayer III Air Chemistry Observatory (Jurányi and Weis, 2026 [10 min resolution]). In these cases, the payload is operated at the ORACLES site, which is approximately 1 km from the Air Chemistry Observatory. As there are no topographic features between the two measurement locations, the air mass is expected to be similar at both sites despite the 1 km distance between them.

When comparing the total particle number concentrations measured by the EERL instrumentation with those measured by the Neumayer III ground-based air chemistry observatory (SPUSO) over a 1 hour period on 28 December 2025, good agreement is observed between the EERL CPC3007 and mSEMS and the SPUSO CPC3775 (lower cut off diameter of 4 nm). However, the SPUSO SMPS shows higher total particle number concentrations. This difference is related to the fact that, as the SPUSO OPC is currently under repair, the measurement range of the SMPS has been extended up to 830 nm. For this extended size range, the diffusion correction applied to the SMPS may lead to an overestimation of the particle number concentration below 100 nm. Note that each instrument covers a different particle diameter range, which needs to be considered when comparing the measured total particle number concentrations.

Regarding the particle size distributions, the mSEMS and SPUSO SMPS show a 20 nm difference in the accumulation mode diameters based on the fitted lognormal curves. When integrating the size distributions over their common diameter range, the resulting total particle number concentrations differ by up to 20% between the mSEMS (263.2 cm^{-3}) and the SMPS_{SPUSO} (331.2 cm^{-3}). As mentioned above, these differences are related to the current size range settings of the SPUSO SMPS and the associated diffusion correction.

In addition, the mSEMS and POPS462 size distributions show good agreement. Integration over their common size range results in a difference of less than 2% in the calculated total particle number concentration (mSEMS: 64.3 cm^{-3} and POPS: 63.5 cm^{-3}), but when integrating the size distributions over the POPS (66.1 cm^{-3}) and SMPS_{SPUSO} (51.0 cm^{-3}) common diameter range, the resulting total particle number concentrations differ by up to 30%.

CLAVIER: Intercomparison with instrumentation from the ACTRIS Villum Research Station atmospheric monitoring setup. The EERL instrumentation was connected to the station inlet on 13-14 April 2024 for approximately 20 hours to perform an intercomparison with the ground-based on-site Mobility Particle Size Spectrometer (custom built Scanning Mobility Particle Sizer).

When comparing the total particle number concentrations measured by the EERL mSEMS with those measured by the Villum ground-based MPSS, good agreement is observed, with slightly higher concentrations measured by the MPSS. Regarding the particle size distributions, the mSEMS and MPSS show an 18 nm difference in the accumulation mode diameters based on the fitted lognormal curves. When integrating the size distributions over their common diameter range, the resulting total particle number concentration measured by the mSEMS (127.2 cm^{-3}) is 15% lower than that measured by the MPSS (150.5 cm^{-3}). This is similar to what was observed for the SMPS from the Neumayer III Air Chemistry Observatory described above. In this case, the MPSS also measures over a broad size range of 8.9 to 850 nm.

176 **Table S3: Information for each field campaign on the additional data available, the distance from the**
177 **helikite launch site to other measurement sites, and key references for accessing the data.**

Campaign	Additional data	Source	Distance from site	Key references
ALPACA	Surface meteorology and aerosol particles	Various campaign instrumentation at University of Alaska Fairbanks Farm site	On Site	https://arcticdata.io/catalog/view/doi:10.18739/A2CR5NF36
ARTofMELT	Surface meteorology and aerosol particles	Various campaign instrumentation onboard IB Oden	On site	Data section in: https://acp.copernicus.org/articles/26/10331/2026/
Brigerbad	-	-	-	-
CHOPIN	Profiles of meteorology and aerosol particles, and remote sensing	Various campaign instrumentation at Kalavrita ski resort	On site	https://projects.au.dk/cleancloud/the-cleancloud-meta-database Select Station: Helmos
	Aerosol particles	Temporary station HAC2 station	On Site 3 km	Contact: athanasios.nenes@epfl.ch
CLAVIER	Surface meteorology, aerosol particles, and remote sensing	Villum research station	On Site	https://projects.au.dk/cleancloud/the-cleancloud-meta-database Select Station: Villum
GreenFjord	Surface meteorology, aerosol particles, and gases	Various campaign instrumentation	On Site	Data section in: https://acp.copernicus.org/articles/26/7165/2026/
ORACLES	Surface meteorology and aerosol particles	Neumayer III station	On Site	https://pangaea.de/
PaCE	Surface meteorology and aerosol particles	Sammaltunturi station	5 km	https://data.icos-cp.eu/portal/#%7B%22filterCategories%22:%7B%22station%22:%5B%22iAS_PAL%22%5D%7D%7D
	Profiles of meteorology and aerosol particles	FMI tethered balloon	3 km	https://essd.copernicus.org/preprints/essd-2026-135/
Turtmann	Surface meteorology and radiation	Temporary weather station	On site	Contact: julia.schmale@epfl.ch

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179 **Table S4: Uncertainties related to each variable provided in the data products.**

Variable name	Uncertainties	Reference
Altitude	Increasing with altitude, up to 15 m at 1000 m	Propagated from pressure uncertainty
Latitude (Lat)	1 m	VectorNav VN-200 datasheet (https://www.vectornav.com/resources/detail/vn-200-gnss-ins?return=/resources/technical-documentation/datasheets&title=Datasheets)
Longitude (Long)	1 m	VectorNav VN-200 datasheet (https://www.vectornav.com/resources/detail/vn-200-gnss-ins?return=/resources/technical-documentation/datasheets&title=Datasheets)
Pressure (P)	1.7 hPa	BME280 datasheet (https://www.bosch-sensortec.com/media/boschsensortec/downloads/datasheets/bst-bme280-ds002.pdf)
Temperature (Temperature)	0.3°C	SHT85 datasheet (https://sensirion.com/media/documents/4B40CEF3/640B2346/Sensirion_Humidity_Sensors_SHT85_Datasheet.pdf)
Relative humidity (RH)	3%	SHT85 datasheet (https://sensirion.com/media/documents/4B40CEF3/640B2346/Sensirion_Humidity_Sensors_SHT85_Datasheet.pdf)
Wind speed (WindSpeed)	0.1 m/s	Pohorsky et al. (2024)
Wind direction (WindDir)	± 2 deg	Pohorsky et al. (2024)
CO ₂ mixing ratio (CO ₂)	3 ppm + 1% of value	Pohorsky et al. (2024)
Total particle number concentration N ₁₈₆₋₃₃₇₀ (POPS_total_N)	± 10%	Pohorsky et al. (2024)
POPS bin concentration (POPS_bX)	± 10% up to 800 nm, 30% above	Pohorsky et al. (2024)
Total droplet number concentration N _{0.66-43} (mCDA_total_N)	Not available	-
mCDA bin concentration (mCDA_dataBX)	Not available	-
Filter sampling (Filter_position)	-	-
Filter flow (Filter_flow)	± 5%	FILT datasheet (https://www.brechtel.com/product/filter-sampler/)
Total particle number concentration N ₁₀₋₁₀₀₀ (CPC_total_N)	± 20%	CPC3007 datasheet (https://tsi.com/getmedia/8c0677b4-6cda-43b6-9a74-1c96acc30d4e/3007_5001117_A4?ext=.pdf)
Total particle number concentration N ₁₀₋₃₀₀ (Partector_total_N)	± 30%	Partector 2 datasheet (https://www.naneos.ch/pdf/naneos_partector2_operation_manual.pdf)
Total particle number concentration N _{8-300max} (mSEMS_total_N)	± 10%	Estimation from intercomparisons during field campaigns
mSEMS bin concentration (mSEMS_Bin_ConcX)	Up to +40% below 30 nm, ± 5% above	Pohorsky et al. (2024)
Aerosol light absorption coeff.	± 0.2 Mm ⁻¹	Pohorsky et al. (2024)

450 nm (STAP_siglab_smth)		
Aerosol light absorption coeff. 525 nm (STAP_sigmag_smth)	$\pm 0.2 \text{ Mm}^{-1}$	Pohorsky et al. (2024)
Aerosol light absorption coeff. 624 nm (STAP_sigmar_smth)	$\pm 0.2 \text{ Mm}^{-1}$	Pohorsky et al. (2024)
MicroAeth eBC mass value for 375, 470, 528, 625, 880 nm (MA_eBC_X)	Minimum $\pm 25\%$	Salo et al. (2024)
MicroAeth light absorption coeff. 375, 470, 528, 625, 880 nm (MA_Abs_Coeff_X)	Minimum $\pm 25\%$	Salo et al. (2024)
O ₃ mixing ratio (O3)	Greater of 1 ppb or 2% of value	Pohorsky et al. (2024)
CO mixing ratio (CO)	1 ppb	Pohorsky et al. (2024)
Total particle number concentration N _{0.2-50} (LOAC_total_N)	$\pm 30\%$	Supplement of Pohorsky et al. (2026)
LOAC bin concentration (LOAC_dataBX)	$\pm 30\%$	Supplement of Pohorsky et al. (2026)

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