

Author Response

Vertically-resolved source contributions to climate relevant aerosol properties in Southern Greenlandic fjord systems

Joanna E. Alden (nee Dyson)^{1, a}, Nora Bergner¹, Benjamin Heutte¹, Lionel Favre¹, Mihnea Surdu¹, Julian Weng², Marta Augugliaro², Patrik Winiger², Berkay Dönmez^{1,3}, Roman Pohorsky¹, Radianca Calmer¹, Carmelle Chatterjee¹, Kalliopi Violaki³, Athanasios Nenes^{3,4}, Luke Gregor⁵, Silvia Henning⁶, and Julia Schmale¹

¹Extreme Environments Research Laboratory, Ecole Polytechnique Fédérale de Lausanne, Sion, Switzerland

²PSI Center for Energy and Environmental Sciences, Paul Scherrer Institute, Villigen PSI, Switzerland

³Laboratory of Atmospheric Processes and their Impacts, Ecole Polytechnique Fédérale de Lausanne, Switzerland

⁴Center for the Study of Air Quality and Climate Change, Foundation for Research and Technology Hellas, Patras, Greece

⁵Swiss Data Science Center, ETH Zürich and EPFL, Zurich, Switzerland

⁶Institut für Troposphärenforschung, Leipzig, Germany

^anow at: Université Savoie Mont Blanc, INRAE, CAARTEL, Thonons-Les-Bains, 74200, France

Correspondence to: Julia Schmale (julia.schmale@epfl.ch)

Reply to referee #1

This study presents a very good analysis of aerosol properties measured at different heights in Southern Greenland fjords. By combining ground-based instruments with tethered balloon measurements, the authors provide a very good dataset that shows how aerosols vary up to several hundred meters above the surface. They identify four main source types: local pollution, aged marine background air, biomass burning plumes, and new particle formation events. The results show that both local human activities and long-range transported wildfire smoke can strongly increase CCN concentrations and influence aerosol optical properties. The study highlights how aerosol properties change with altitude and show why vertical measurements are important for understanding aerosol–cloud interactions. The analysis is well supported, and the connection between aerosol sources and climate-relevant properties is convincing. Also, the manuscript is clearly written and well structured. Overall, I recommend the manuscript for publication.

We thank the reviewer for the positive and thoughtful evaluation of our manuscript. We appreciate the reviewer's recognition of the value of the combined ground-based and tethered balloon measurements and the importance of the vertical aerosol observations in Southern Greenland fjords. We are glad that the reviewer finds the analysis well supported and the manuscript clearly written and structured.

Reply to referee #2

We thank the referee for their comments, addressed individually in red below. Changes to the manuscript are also noted below.

General scientific comment:

The manuscript presents interesting and valid results on aerosol properties measured at a Southern Greenlandic location. The manuscript is marked by a high quality of written English language. Major efforts were carried out in doing an extensive field campaign also establishing a ground-based station including even remote sensing techniques at the remote site. In addition, tethered balloon measurements were performed. The work presents a rare and comprehensive dataset worth publishing while investigating different air mass types that were classified according to the prevailing major observations.

Minor revisions are suggested to improve the manuscript addressing the comments below. One major point is that I recommend not to discuss figures and tables from the Supplementary Information in the main text. The manuscript should focus on major findings presented in the main manuscript. Readers that want to get e.g. more information about additional cases should go to the SI and find all relevant information there. One sentence referring to that information in the main manuscript should be enough.

We thank the referee for the careful review of our manuscript and for the positive assessment of the dataset and field campaign. We appreciate the recognition of the extensive measurement effort, including the combination of ground-based observations, remote sensing, and tethered balloon measurements at this remote Southern Greenlandic site.

We also thank the referee for the constructive suggestions for improving the clarity and structure of the manuscript. In particular, we have revised the manuscript to better separate the main findings from supporting material in the Supplementary Information, following the reviewer's recommendation. Several figures and detailed discussions have been moved to the SI or removed where appropriate to streamline the main text.

All comments have been addressed individually below, and the manuscript has been revised accordingly.

Detailed scientific comments (main manuscript):

2.1 Sampling site

Page 4, line 95:

Comment: Could you provide the distance of the mountains to Narsaq?

The distance from Narsaq to the summit of Qaqqarsuaq is approximately 2 km, 1.5 km to the summit of Tassiigaaq, less than 1 km from the centre of Narsaq to the base of mountains.

This information has been added to the manuscript.

2.2 Ground-based measurements

Page 5, line 109:

Comment: Please say what you mean with interstitial, does it mean the sizing (PM1/TSP) was decisive if interstitial or total was measured, then please say so.

The particle size cutoff for the interstitial and total inlets were 1 μm and 40 μm respectively. Added to the manuscript.

General comment:

For a better overview, I suggest to place Figure S1 from the supplementary information to the main manuscript.

Figure S1 moved to before Table 1.

2.2.1 Particle number concentration and size distribution

Page 6, line 118:

Comment: Has the SMPS been size calibrated or a comparison to total particle number has been checked against another particle counter? If so, please state.

In line 211, Section 2.3, we state that the mSEMS and SEMS intercomparison in the field show good agreement in total PNC and PNSD. This is shown in Figure S4, now Fig. S3.

2.2.2 Aerosol absorption and scattering coefficients

Page 8, line 151:

Comment: Can you say something about the calibration of the Aurora with span gas and zero measurements?

The following has been added to the manuscript to address this: “The Nephelometer was calibrated before the campaign and periodic zero tests were conducted throughout the campaign. A span gas check and calibration were performed on 11 July 2023 but with values that fell outside the recommended range of the manufacturer. Therefore, a flag is applied to data following the unsuccessful calibration although no discernible shift or step-change in the data was observed.”

2.2.3 Cloud condensation nuclei number concentrations

Page 8, line 156:

Comment: Has a calibration based on ammonium sulphate or similar salt been performed? If so, please state.

Line 156 edited to give clarity. The line now reads : “Calibration of the CCNC was performed following the ACTRIS procedure, detailed in Gysel and Stratmann, 2013 and Rose et al., 2008 with ammonium sulphate particles both before and after the campaign to ensure

supersaturation values remained consistent, with post-campaign results showing good agreement with the pre-campaign calibration ”

2.4 Data processing, pollution flagging, and environmental condition categorization

Page 10, line 225 - 240:

Comment: I am missing more hard criteria of category nr. 1 and nr. 3. I understand that category nr. 2 will be explained later in the manuscript and nr. 4 has a subjective character. But for category nr. 3 you must have some mathematical or other criteria to identify the events? Can you please elaborate more clearly on this?

We clarify that category 1 is defined by exclusion: days not identified as Category 2, 3, or 4 are classified as background. Growth events were identified following the classification scheme of Kulmala et al. (2012), in which events are categorised based on the characteristic appearance of a coherent, continuously growing nucleation mode in the PNSD time-evolution plot (commonly referred to as a 'banana' shape). Given the sub-Arctic maritime setting and the high growth rates observed ($1.7\text{--}7.6\text{ nm hr}^{-1}$; Table S8), which are at the upper end of or exceed typical Arctic NPF rates reported in the literature, all identified events were unambiguous — In the few borderline cases growth rates were still calculated and then discounted if negligible. Growth rates were subsequently calculated for all identified events as described in Text S4, confirming the coherent size evolution in each case. The GR calculation itself therefore serves as a post-hoc quantitative verification of the visual classification. We have added the following into the manuscript: "Growth rates were calculated for all identified events (Text S4, Table S8), providing quantitative confirmation of the coherent size evolution in each case."

Page 10, line 242:

Comment: Please check comment in German language in this line!

Removed from manuscript.

Page 11, Figure 2:

Comment: I recommend to place Figure 2 rather more in the supplementary information.

As Fig S5, now S4, makes Fig 2 somewhat redundant, we have removed Fig 2 from the manuscript and update the manuscript text to reference Fig S4 alone.

3.2 Background versus fresh pollution regimes

Page 19, line 362:

Comment: Do you mean that BG_{CI} includes regional particle formation and growth events that are not identified and classified as new particle growth events (GE)?

Yes, in the sense that the background category will include particles that originated from growth events but have grown beyond the 6-hour window used to define the GE category (Section 2.4). The 6-hour window was chosen to capture the active growth phase consistently

across all events, some of which had ceased growing within this period and others which had not. Data outside this window, including particles both above and within the nucleation mode sizes, are therefore retained within the background category, where they contribute to the background Aitken mode population and are no longer distinguishable from it.

Page 19, line 371:

Comment. I agree that pollution clusters are different in magnitude and variability, but that does also count for the middle cluster of the background. Do you have an explanation for that?

The variability within the background clusters is consistent with the definition of the background category itself (Section 2.4), which represents a mixture of marine aerosol, aged anthropogenic pollution, and potentially local waste-related emissions, with no expectation of homogeneity. The three background sub-clusters likely reflect natural day-to-day variability in the relative contributions of these sources, driven by changing meteorological conditions and air mass history. Unlike the pollution/background distinction — which could reflect a mechanistic difference in source type — the internal background variability does not warrant a separate physical interpretation beyond what is already acknowledged in the category definition. Furthermore, it is worth noting that while short-lived pollution spikes are excluded from the background category by definition, aged and processed anthropogenic aerosol remains present and will contribute to variability within the background population. The three background sub-clusters therefore likely reflect varying mixtures of marine aerosol and aged anthropogenic influence under different meteorological conditions, which is entirely consistent with the category definition and expected given the location.

Addition of the following after line 371: “The internal variability within the background clusters is consistent with the background category definition, which retains aged and processed anthropogenic aerosol alongside the marine signal, with only the short-lived fresh pollution spikes that are excluded, and reflects natural day-to-day variability in the relative contributions of these sources under changing meteorological conditions.”

Page 20, line 381:

Proposal: ... of specific aerosol properties ...

Suggested edit made to the manuscript.

3.3 Particle growth events

Page 21, line 398:

Comment: You mean (Fig. 3)!

Corrected in the manuscript.

Page 21, Figure 7:

Comment: What do you mean by normalised?

The term 'normalised PNSD' refers to the standard representation of particle number size distributions as $dN/d\log D_p$, in which the particle number concentration is normalised to the logarithmic width of each size bin.

Page 22, line 420:

Comment: Can you in general exclude any anthropogenic sources in this region which you calculate to be the start area of the event? The conclusion is quite fascinating that the fjord must be the source!

While anthropogenic sources within the source region cannot be entirely excluded, the upwind region to the west consists predominantly of the fjord while to the north lies the glacier terminus, an area with no significant settlements or industrial activity within the estimated nucleation distances of 5.5–31.8 km. The absence of a clear mode below 8 nm in the ground-based measurements — noting that GE c1 exhibits a shoulder around the 8 nm detection limit — is consistent with particles having already undergone initial growth prior to arrival at the measurement site, supporting transport from a remote nucleation source rather than local formation. However, we acknowledge that the 8 nm lower size cut-off of the SEMS precludes definitive conclusions about the presence or absence of freshly nucleated particles at the measurement site. A more detailed investigation of NPF source attribution in Southern Greenland fjord systems will be presented in a forthcoming paper based on ship-borne observations.

Page 23, line 440:

Comment: Please check comment in German language in this line!

Removed from manuscript

Page 23, line 442:

Comment: Honestly, I clearly see also a trimodal distribution for sGE c1 cluster. In any case, this does not change your conclusions on the evolution of the whole event! I recommend to refit the data.

Re-fitting the data does not change the small mode we can see above 100 nm. Line 442 has been changed to say : “sGE c2, a tri-modal distribution peaking at 21, 57 and 172 nm”

Page23/24, Figure9/10:

Comment: I feel it is a bit complicated to follow the figures and also your conclusions. I suggest to combine Figure 9 and Figure 10. It would be great to see the evolution of PNSD in terms of clusters or averaged distributions of the ground PNSD based on the vertical PNSD. I understand here that the clustering is done separately for both instruments. This makes in principle sense, but does not show all the time similar time periods when you compare the distributions. Can you only cluster vertical PNSDs and then show the median distributions of the ground-based PNSD for these cluster periods? I ask this as you want to identify connections between the distributions.

We thank the reviewer for this suggestion. However, given that the ground and vertical measurements are concurrent, calculating median ground PNSDs for the vertical cluster periods would yield distributions equivalent to those already shown, as the ground clusters (sGE c1 and c2) and vertical clusters (vGE_200 c1 and c2) correspond to the same time periods by construction. This correspondence is demonstrated directly in Fig. S13, now Fig. S11. To aid clarity, we have added dashed and dash-dot vertical lines to panel d to represent the periods where sGE c1 and c2 are present on the ground, for easier direct comparison with the vertical cluster time periods.

The additional text has been added to the figure caption : “Region between black dashed lines in panel d) represents time during which *sGE c1* was present on the ground, while region between black dash-dotted lines represents time during which *sGE c2* was present on the ground. Direct comparison of ground and vertical cluster distributions is provided in Fig. S11. ”

3.4 Biomass burning plume advection

Page 29, line 554 - 555:

Comment: Extend the sentence “... observed with the mSEMS at higher altitudes ... observed with the SEMS at ground level ...cc

Amended in the manuscript.

Page 30, line 562 - 570:

Comment: I do not agree with extensive discussions of details in the main manuscript while figures are found in the supplementary information. Either you include all in the main manuscript or you put the explanation to the supplementary information where the figures are found. Having scientific discussions in the main manuscript that need the detailed reading of the supplementary information are in my view not in the sense of the use of supplementary information, you here simply extend the main manuscript. Decide!

We have deleted the plume 2 and 3 summary from the main manuscript, leaving the full discussion in the SI. We feel that moving the full discussions into the main manuscript would considerably lengthen the paper.

3.5 Source contribution to climate relevant aerosol properties

Page 30, line 584 - 586:

Comment: Please check sense of sentence as you compare here the different categories to the background conditions!

Thank you for spotting this. The sentence should have started “We could clearly identify periods elevated over BG (Fig. 15a–c),...”

Page 31, line 602 - 607:

Same comment as above: (Repetition) I do not agree with extensive discussions of details in the main manuscript while figures are found in the supplementary information. Either you include all in the main manuscript or you put the explanation to the supplementary information where the figures are found. Having scientific discussions in the main manuscript that need the detailed reading of the supplementary information are in my view not in the sense of the use of supplementary information, you here simply extend the main manuscript. Decide!

We agree with the reviewer. Following the same approach applied to the vertical case study, the detailed discussion of wavelength-dependent absorption has been removed from the main text and relocated to the supplementary information alongside Fig. S23, now Fig. S21.

4 Conclusions

Page 33, line 632:

Comment: "... with enhanced optical properties ..." - Please be more specific here!

Clarification made in the text.

Page 33, line 649:

Comment: With view on the values, are you referring to median values? Please state!

These values indeed refer to medians. Clarification made in the text.

Detailed scientific comments (supplementary information):

Figure S2:

Comment: Does the RH show the ambient RH outside or is it the RH after drying. In this case it would be quite high especially for filter-based measurements? Please comment!

This shows the ambient RH outside, not the RH after drying. The following was added to the figure caption : "Note this is the outside RH, not the RH after drying in the inlet."

Figure S7:

Comment: From the colour coding I cannot see the two PNSDs for both size ranges in each plot. Can you simply only show the mean cluster PNSD for both size ranges in each plot?

Thank you for this suggestion. As the two PNSDs are nearly identical (with average differences of less than ~5%), we have removed this figure from the Supplementary Information to streamline the SI and instead note in the text that the average PNSDs for the two size ranges differ by less than 5%.

Added text: "Ground clustering using size ranges of 8–1000 nm and 8–240 nm produced nearly identical particle number size distributions (PNSDs), differing on average by less than 5%."

Figure S14:

Comment: To make the figure caption complete you need to label also a), b), and c).

Panel letters added to the figure caption.

Page 30, line 251:

Comment: Please check comment in German language in this line!

Removed from manuscript

Figure S22:

Comment: For my understanding Figure S22 d)e)f) should be the same as Figure 15 a)b)c) in main manuscript. You use different x-scaling, but also look at Figure S22d) which looks different from Figure 15a)?

We thank the reviewer for spotting this mistake. Re-plotting and re-scaling has fixed this issue and the redone plot is replaced in the SI.

Specific language comments (main manuscript):

All language comments taken into account and corrections integrated into manuscript.

Abstract

Page 1, line 19:

... aerosol number size distribution ...

3.1 Campaign overview

Page 16, line 323:

... peaked ...

3.2 Background versus fresh pollution regimes

Page 20, line 382:

.... This data ...

3.4 Biomass burning plume advection

Page 25, line 480:

..... arrived about 300 m above the ground with a ...

Page 28, line 532:

Check language “between” and following of that line!

Page 28, Figure 13:

... clustered PNSD at ground level covering ...

Page 29, line 546:

... (blue, vP1_100 c2) ...

Page 30, line 577:

... clustered particle number size distributions ...

Specific language comments (supplementary information):

Page 18, line 136:

... Table S6 ...

Page 20, line 170:

... is ...

Page 32, line 274:

... a first broadened ...

Page 32, line 275:

... a second ...

Page 32, line 276:

... a third ...

Page 32, line 277:

... 39.8 ...