Review

General

The paper presents results and analyses of aerosol measurements conducted at two observatories Gruvebadet (GAL) at 61 m a.s.l and Zeppelin (ZEP) at 472 m a.s.l in Ny Ålesund, Svalbard. The data consists of aerosol optical properties, concentrations of ions, trace elements and lead isotopes as well as meteorological data and air mass backtrajectories from February 2022 to March 2023. Simultaneous measurements at the two altitudes have been used to shed light to the layering of aerosols in the Arctic atmospheric boundary layer (ABL). The paper is important as it presents the most comprehensive comparison of aerosols at GAL and ZEP to date and analyses of the variability at both of them.

However, it is fairly heavy to read and it has several deficiencies, see below. I can recommend publishing the paper after making the respective corrections and modifications. Some of the detailed comments are just suggestions that I think would improve the paper, it is up to you decide.

A: We thank the Reviewer for their positive and constructive evaluation of our manuscript, and for recognizing the importance of this study comparing the aerosol measurements at Gruvebadet and Zeppelin observatories. We have completely revised the manuscript to improve clarity and flow, addressing all the specific comments and suggestions provided. Below, we provide point-by-point responses to each comment and describe the corresponding revisions made to the text.

Major deficiencies

1) There is no proper evaluation of local contamination at GAL. Now the word "contamination" appears in the paper only three times: twice dealing with sample analysis pretreatment and once in the title of a paper in the references. For me it gives the impression that it has been tried to avoid to discuss the whole issue. But that makes no sense, all field stations suffer from it, everybody needs to face the same problem. Proper analysis would be very valuable not only for the present paper but also for all future short campaigns, longterm monitoring and data a the whole period or different seasons. The WD boxplot that is now as an inset in Fig. S11 is definitely not informative enough, just delete it. Then plot absorption coefficient and single-scattering albedo SSA as a function of WD and WS in some method, for instance as a pollution wind rose or some other polar plot or simplest as absorption vs. WD at different WS. I recommend using log scale for the absorption as the range will strech over several orders of magnitude. Typically at field stations, clear contamination sector analyses and possibly for designing sampling sector control. This paper would be very suitable for presenting it, it has all the necessary data. This analysis would be most useful at the beginning of the main paper or the attachment, it could be referred to in the other data analyses in the rest of the paper. Plot standard wind roses showing the probability distribution of wind direction (WD) and wind speed (WS) in sectors. For can be found this way: high absorption coefficients (and eBC) and low SSA. It will also show some minimum WS below which contaminated air is stagnant and aerosol concentrations rise.

A: We thank the reviewer for this constructive and valuable suggestion. We fully agree that evaluating potential local contamination is crucial for remote field stations like GAL and provides important context for data interpretation.

To thoroughly address this point, we have generated a new pollution rose plot (now included as Figure S3 in the Supplementary Material) that shows the aerosol absorption coefficient (σ_{abs}) and Single Scattering Albedo (SSA) as a function of wind direction and wind speed. For what concern the wind direction boxplot included in Fig. S11, we delete it as suggested by the referee. However, the polar plot of the wind patterns is included in the Fig. S1.

In agreement with the reviewer's recommendation, we have introduced a dedicated discussion on local contamination in Section S1.1 of the revised supplementary material (referencing the new figure in the SI):

The polar plot reported in Fig. S3 shows that the highest aerosol absorption coefficient values (σabs > 2.0 Mm-1, dark red markers) are predominantly associated with winds coming from the North (345° - 360°) and East/South-East (90° - 110°) sectors at moderate to high wind speeds (5 - 12 m/s). These high-absorption events also correspond to smaller marker sizes, which signify lower Single Scattering Albedo values (SSA ≅ 0.85) and point to the presence of strongly absorbing anthropogenic aerosols such as black carbon. Interestingly, the North-East sector (45°)—where the human settlement is located just 1 km away—exhibits low data density and significantly lower absorption levels, indicating that direct pollution transport from this nearby settlement is not the primary driver of contamination at the measurement site; instead, the main sources of contamination appear to originate from the Northern and Eastern/South-Eastern sectors or are transported from farther upwind sources.

2) The optical properties have been discussed only very briefly in section 3.2 and without any proper quantitative connections to the filter samples, concentrations of the chemical constituents and total aerosol mass. This is definitely a deficiency, you are not using the 2 full potential of your data. The simplest approach would be to make scatter plots and linear regressions of scattering vs aerosol mass concentrations. And absorption vs EC. Those would first give a simple quality check, they should be positively correlated. Secondly, they would yield you mass scattering coefficient MSC of total mass and mass absorption coefficient MAC of EC. But much better would be to use the concentrations of major chemical constituents and calculate multiple linear regression MLR of scattering and the constituent concentrations the way that has been done in the US national parks within the IMPROVE network for decades. See, e.g., Malm and Hand, Atm.Env. 41, 3407-3427 (2007) and newer papers citing that. MLR is fast and simple to do even with Excel. You would get MSC (±sterr) of the major constituents into a table to be directly compared with those published elsewhere. You would thus have a site-specific equation for visibility estimation at GAL and ZEP, telling how much the variability of a selected constituent affects the variability of visibiity. Just at the core of haze research and of your paper, right?

A: We sincerely thank the reviewer for the constructive suggestion to apply multilinear fitting to isolate the optical properties of individual chemical components. While we agree this would yield valuable insights, the primary scope of this manuscript is to compare aerosol dynamics between two Arctic sites at different elevations to better understand boundary layer processes. Adding an in-depth analysis of component-specific optical properties would significantly broaden the scope and potentially obscure the central narrative of the paper. To address the reviewer's underlying point while preserving the study's primary focus, we have expanded the revised manuscript to include the analysis and discussion of the overall Mass Absorption Cross-section (MAC). We plan to pursue a detailed investigation into the Mass scattering cross section as a function of aerosol size distribution and as a function of aerosol components and sources, after proper source apportionment analysis in a dedicated follow-up study. The manuscript has been modified adding the following paragraph to section 3.6:

"To investigate the optical properties of black carbon at both sites, Fig. 9 shows the variability of the aerosol absorption coefficient at 660 nm versus the EC concentration. Absorption at 660 nm was selected to minimize potential interferences from other light-absorbing species, such as brown carbon and dust. EC concentration was determined using the EUSAAR_2 protocol. At both sites, EC and the absorption coefficients correlated strongly, with Pearson correlation coefficients (r) of 0.95 at Grivebadet (n=18) and

0.95 at Zeppelin (n=17). The BC Mass Absorption Cross sections (MAC), representing the ability of EC to absorb light at 660 nm, are calculated using the York regression, to take into account the uncertainties of both the EC concentrations and the absorption coefficients. The resulting values were 13.2 m2 g-1 (95% CI: 10.6 - 15.7 m2 g-1) at Zeppelin and 10.5 m2 g-1 (95% CI: 7.9 - 13.1 m2 g-1) at Gruvebadet, indicating no statistically significance difference within their uncertainty ranges. The observed MAC values are comparable to coefficients reported previously for Zeppelin and other Arctic sites (Sing et al., 2024, Ohata et al., 2021)"

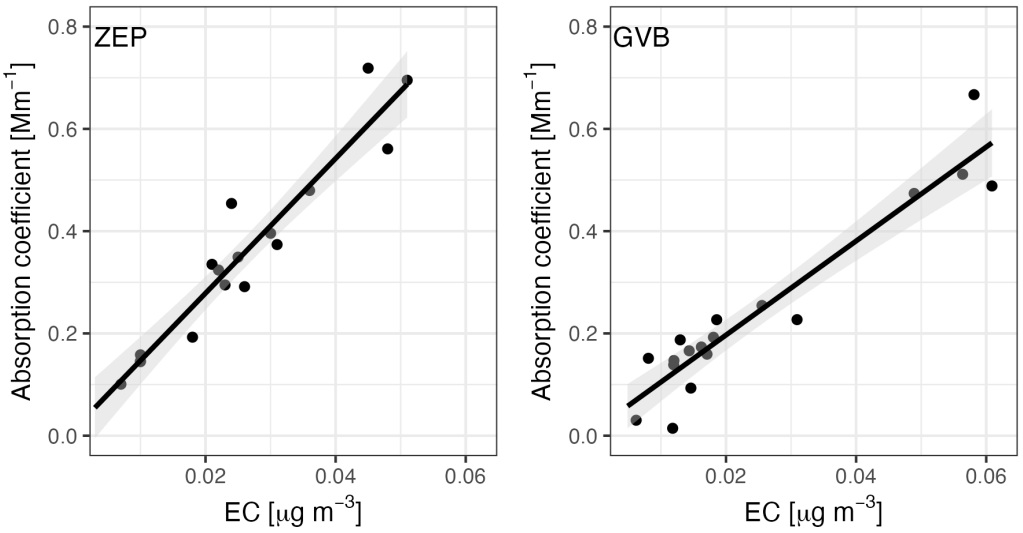


Figure 9. Comparison between absorption coefficients at 660 nm and EC concentrations at Zeppelin and Gruvebadet.

Detailed comments

L53: A suggestion: to be consistent with the introduction of ZEP in the next paragraph you could also give the altitude of GAL here even though both are presented later.

A: We added (61 m asl) to be consistent with the introduction of ZEP.

Fig 1: A suggestion: take the map of the present Fig. S11 and put it below the photographs. The photographs would be 1a, the map 1b. The map is such an important picture that it should be at the beginning of the whole paper. Or as Fig. S1.

A: Following the reviewer's recommendation, we have updated Figure 1 in the main text: the map is now presented in panel 1a and the photographs in panel 1b.

L115: "... Absorption data were corrected for multiple scattering using a correction coefficient (C) ..." Give the value of C. Another question is that the AE33 reports actual BC concentrations even if it actually measures attenuation. Which MAC values were used for calculating absorption coefficients? Many are available in the literature. If you used the manual's values, fine, just write it here.

A: We thank the referee for highlighting this important point. To avoid the uncertainty derived from the use of a non site-specific MAC, we compared absorption coefficients at the two sites, instead of BC concentrations. In the original manuscript, the absorption coefficient at Gruvebadet was not converted into BC mass.

L118: How and how often was the nephelometer calibrated and zero checked?

A: The following text has been added to the manuscript: "At GAL, two identical nephelometers are swapped roughly every year during spring. The nephelometer not currently in use is sent to Italy for calibration before being shipped back to the Arctic to replace the unit that has been collecting measurements in the meanwhile. The dataset used in this article was obtained using the instrument calibrated approximately one year earlier (data of February–April 2022) and one calibrated just before to be shipped (April 2022–March 2023)."

L114-118: What were the flows of the AE33 and the nephelometer? Inlet cutoff diameter?

A: The following text has been added to the manuscript: "Both nephelometer and AE33 at GAL operate with an internal air flow equal to 5 lpm, under a isokinetic TSP main inlet."

L128-129: " Ion composition was measured by two Ion Chromatographic systems performing the analysis of inorganic anions and inorganic cations. " Give the list of anions and cations analyzed.

A: The complete list was added to the main text and here it is: (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}, $C_2O_4^{2-}$)

L150: "...inductively coupled plasma atomic emission spectrometry ..." You hve given acronyms for all other ICP spectrometers, why not this?

A: As suggested by the referee, we used the acronym ICP-MS instead of "inductively coupled plasma mass spectrometry".

Section 2.1: All filter samplers: was there any sector control for eliminating local contamination? I don't find any discussion on clean and dirty sectors. Add that.

A: As suggested by the referee, we included a discussion about local contamination in the supplementary information Section S1.a.

L252: "3.2 Aerosol optical properties" There is no section 3.1 so shouldn't this be 3.1?

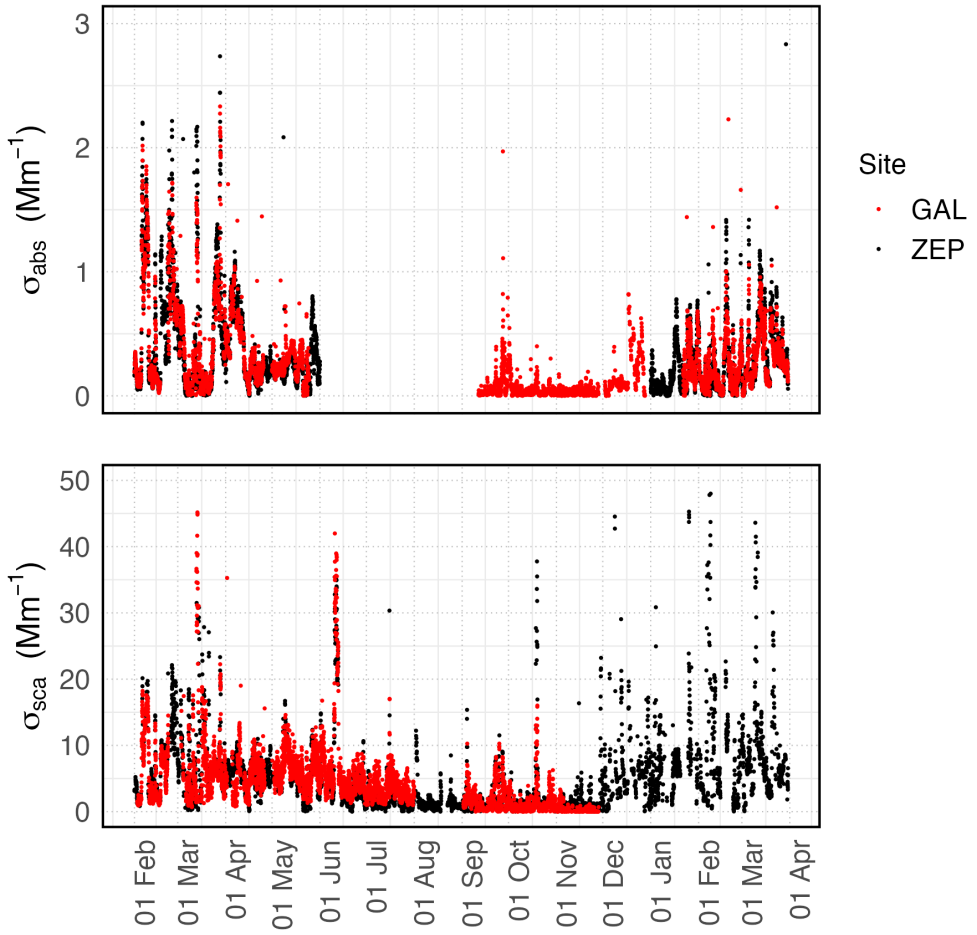
A: Thanks, we completely agree with the referee.

Fig. 2: change the x axis time scale to the same as in Fig 3, always starting on the first day of each month. That makes it easier to compare data. Harmonize the figure time scales as much as possible. And on the contents of the section "3.2 Aerosol optical properties". As I wrote above about major deficiencies of the paper, this section could possibly be the one for presenting connections between aerosol optical properties

and concentrations of aerosol mass and chemical constituents. Or it could be even more logical after all chemical analyses have been presented, just before section 3.9 But wherever, it could consist of two figures. The first could be a two-panel figure as in Fig 2: time series of EC mass concentrations and aerosol absorption coefficients averaged over the filter sampling periods and aerosol mass concentration and scattering coefficient averaged over the filter sampling periods at the two sites. The second could be the scatter plot with linear regressions to yield MSC and MAC as I suggested above. Then the IMPROVE-type MLR, its results in a table and some discussion.

A: In agreement with the previous comment, we modified section 3.6 by adding the discussion of the mass absorption cross section at the two sites after comparing aerosol absorption coefficients and EC concentration. We decided to move the discussion on the variability of scattering coefficients in a dedicated paper.

Figure 2 has been updated as follows to make it comparable with the other figures. We also modified the color of markers to make it consistent with the other parts of the manuscript.



L266-278: This comparison of the scattering and absorption coefficients at the two sites is far from satisfactory. It is clear that the differences will be centered around zero but it hides important information. Instead, I suggest you make a matrix of loglog-scale scatter plots of abs(GAL) vs abs(ZEP) and scat(GAL) vs scat(ZEP) and a linear-scale SSA(GAL) vs SSA(ZEP) scatter plot. Log scale is good in these because the values vary several orders of magnitude and it is relevant to see how they agree over the range. If you colorcode them with WD and WS you could possibly find explanations of the differences and agreements.

We thank the reviewer for this insightful comment. We agree that the original plot was not optimal for investigating the differences in optical properties between the two sites. Consequently, we have replaced Figure S4 with a new figure incorporating the suggested scatter plots of absorption coefficients, scattering coefficients, and single scattering albedo (SSA).The updated figure confirms a satisfactory comparison, with discrepancies falling within measurement uncertainty. The absorption and scattering coefficients at both sites align closely along the 1:1 line, with larger relative differences occurring at lower values near the instruments' analytical detection limits. Similarly, the SSA comparison demonstrates good agreement between the sites when the average extinction coefficient is larger than 3 Mm^{-1}.

The largest discrepancies are observed in the scattering coefficients. Higher scattering at Gruvebadet is generally observed during the warm season, likely due to inputs of local marine aerosols retained in the lower troposphere, or efficient cloud scavenging of inorganic particles at Zeppelin during cloudy periods. Conversely, higher scattering at Zeppelin occurs mainly during the cold season, which may be attributed to effective stratification of the lower troposphere and more efficient long-range transport at higher elevations. It is important to note that such discrepancies are observed for relatively low scattering coefficients (values around 1 Mm^{-1}). The seasonal differences will be evaluated in a more detailed future study.

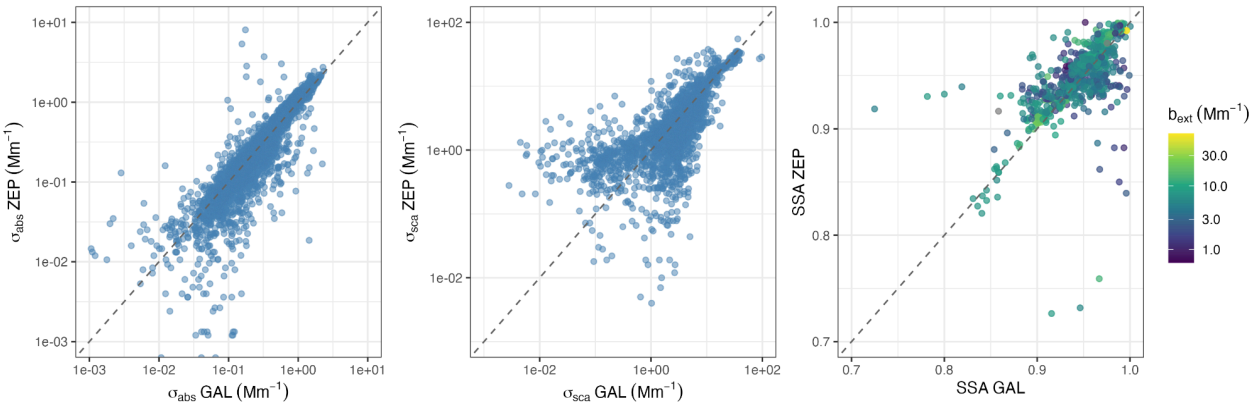


Figure S4. Comparison between absorption coefficients, scattering coefficients, and single scattering albedo at the two sites. SSA markers are color-coded as a function of the mean extinction coefficients.

Following the referee's suggestion, we also evaluated the potential influence of local wind direction, particularly regarding local contamination from the nearby village and harbor at Gruvebadet. Nevertheless these conditions accounted for less than 1% of the total measurement time and had no significant impact on the overall conclusions.

The text is modified as follows in red:

The comparison of the absorption coefficients, scattering coefficients and Single Scattering Albedo (SSA) at the two sites during the study period indicate that optical properties are comparable for most of the time and the discrepancies become evident when measurements are close to the analytical detection limits (Fig. S4). The accuracy of the absorption coefficient measurements, as reported by the manufacturers, is 0.05 Mm-1 for AE33 data (1-hour averages) and 0.1 Mm-1 for MAAP measurements (30 min averages). Asmi et al. (2021) observed that in ambient conditions the two instruments can measure coefficients as low as 0.012 Mm-1. Considering this value as the uncertainty level, the absolute value of the difference between the measurements would be significant if larger than 0.036. About 45% of the time, the difference of the absorption coefficients between the two sites was below this value.

For the integrating nephelometer, a systematic uncertainty below 10% is generally considered (Anderson et al. 1998). Assuming an uncertainty of 5% to be conservative, this would correspond to about 0.1 – 0.25 Mm-1, depending on the period. During the cold and the warm season, the difference between the scattering coefficient was not significant (lower than 3 times the combined uncertainties) for 61% of 40% of the time, respectively.

The comparison of the optical properties reported indicates that the two sites observed similar air masses and/or experienced similar processes for about half of the time during the investigated period.

L279: 3.3 Comparison of seasonal trends of sulphate and ammonium Why not nitrate?

A: We agree with the referee that nitrate would have been another important species to study, being nitrate mostly in acidic form in polar aerosol and being highly capable of being neutralized by ammonia. Nevertheless, given the broad view of the paper, we focused on the main neutralization reaction occurring in the atmosphere. Ammonia is the dominant trace gas capable of neutralizing the acid gases produced by the oxidation of SO_2 and NO_2 in the atmosphere and the soluble ammonium aerosol salts of sulfuric acid and nitric acid produced by neutralization processes with atmospheric ammonia become atmospheric aerosol particles in the sub-micron size range (McMurry et al., 1983). Gaseous ammonia can react with gaseous nitric acid and hydrochloric acid to produce NH_4NO_3 and NH_4Cl, respectively, through a process depending on different variables, but especially relative humidity in the atmosphere. However, ammonium sulphate aerosol has a low vapor pressure, which allows it to condense easily on particle and droplets surfaces even at low relative humidity. Hence, the chemical reaction between sulfuric acid and ammonia is mostly favoured among the possible chemical reactions in the atmosphere (McMurry et al., 1983; Warneck, 2000).

Moreover, nitrate concentration is always lower than sulphate one along the whole observation period. Namely, nitrate concentration is about 15% of sulphate concentration as an average through the whole year. During winter the nitrate /sulphate ratio is particularly low (around 12%) while in summer it reaches its maximum (about 21%) but sulphate still stays as the dominant acidic species.

McMurry, P.H., Takano, H., Anderson, G.R., 1983. Study of the ammonia(gas)-sulphuric acid(aerosol). Environmental Science and Technology 17, 347–352.

Warneck, P., 2000. Chemistry of the Natural Atmosphere, second ed. Academic Press, San Diego.

Fig. 3: In Feb 2022 there were very big GAL-ZEP differences for both SO4 and NH4. All month. Was WS low? Cold temp? Strong inversion? Interestingly, I don't find the word inversion anywhere in the ms. Such a topography would be ideal for the formation of inversions and pollution layers. Analyze that more (WS, WD, T time series, maybe also trajectories or footprints) , it would be very enlightening and maybe important. More about Fig. 3: It would be most useful to combine fig S5 here because they are so connected. As separate subfigs c and d. In Fig. S5, use more different colors for the two reference lines ammonium sulfate and ammonium bisulfate.

A: The reviewer is right in spotting a marked discrepancy for ammonium and sulphate between GAL and ZEP in February 2022. Such a difference, along with a general agreement of values between the two sites is not completely surprising. In fact, a comparison study of aerosol physical properties and sulphate concentration at GAL and ZEP (Rader et al., 2021) showed that sulphate concentration erratically changed even by a factor of 25 although aerosol load showed much lower variations (3-fold at maximum)

Rader, F.; Traversi, R.; Severi, M.; Becagli, S.; Müller, K.-J.; Nakoudi, K.; Ritter, C. Overview of Aerosol Properties in the European Arctic in Spring 2019 Based on In Situ Measurements and Lidar Data. Atmosphere 2021, 12, 271. https://doi.org/10.3390/atmos12020271

L343-344: "... we calculated crustal (CEF, Fig. S8) and Marine (MEF, Fig. S9) enrichment factors, following Barberi et al. (2016)." This is not detailed enough. I checked the paper of Barbieri (2016). It is written there this: "The Enrichment Factor is expressed as follow: (1) EF = (Metal/RE)soil/(Metal/RE)background Where, RE is the value of metal, adopted as Reference Element" 4 But there are no tables or numbers for element concentrations in background soil. So, where did you get all the background element concentrations and what did you use as the reference element? Probably Al? The same applies to the marine EF calculations. I suppose Na was your ref element for MEF. Show the equations for both EFs and the correct citations of the tables of background concentrations.

A: Thanks for this note. We have modified the text reporting the reference for the average concentration of elements in the upper continental crust and in the seawater, for the calculation of CEF and MEF, respectively, in substitution to the reference of Barbieri (2016). The new lines are: "To identify the dominant source categories, namely crustal, marine, and anthropogenic, we calculated crustal (CEF, Fig. S8) and mMarine (MEF, Fig. S9) enrichment factors, according to equation (1), using the mean element concentration in the upper continental crust (Wedepohl, 1995) and in the ocean (Goldberg, 1965) were used for the calculation of the terms at the denominator. Al and Na were used as reference elements for the calculation of CEF and MEF, respectively."

L350: "...The temporal variability normalized time series ..." I don't understand what you have done, see my question on Fig. S10 caption.

A: In these lines and in Fig. S10, we aimed to focus on the concentration profiles throughout the year. To facilitate the comparison of temporal trends among elements with very different concentration levels, and to highlight common patterns over time (e.g., Na and Sr), we decided to report, for each element, not the absolute concentration in the samples, but the ratio between its concentration and the maximum value observed overall. In this way, each element's line ranges from 0 to 100, making the comparison of the profiles easier.

To better explain this concept we modified the text as reported below:
"Figure S10 shows the normalized temporal profiles of the analysed elements. For each element, concentrations were divided by the maximum value observed for that element over the study period and expressed as a percentage. This approach enables comparison of the temporal variability among elements with markedly different absolute concentrations. The profiles highlight a clear seasonality, with generally higher relative concentrations in winter and lower values in summer. "

L379-380:" A comparison of selected elemental concentrations measured at GAL and at ZEP (Heavy metals at Zeppelin mountain (Ny Ãlesund), 2025) shows..." Add here the text "(Table 2)" because it is not written here where the comparison can be found.

A: According to your suggestion the text was modified as reported below:

“A comparison of selected elemental concentrations measured at GAL and at ZEP (Heavy metals at Zeppelin mountain (Ny-Ålesund), 2025) is reported in Table 2. It shows generally higher values at ZEP are generally observed, except from Pb and As, for which concentrations were similar at both sites.”

L391-392:"... vertical processing causes MD, even when present in the fine fraction, to be preferentially depleted..." The acronym MD will be defined later, in section 3.6. S either write it as mineral dust or give definition here.

A: according to your suggestion, the acronym MD was changed with “ crustal-derived aerosol material”

L410-411: Explain with a few sentences, what these isotope ratios tell, why they were analyzed. You present two different lead isotope ratios without any reasoning. Explain that for both of them.

A: An explanation of the rationale and meaning of the use of Pb isotopes in source assessment has been added at the beginning of paragraph 3.4.

L429: Explain the concept of the end member.

A: When samples are arranged in a straight line in the three-isotope plot, their isotopic composition can be explained with a binary model from the mixing of two sources (end-members, or EMs). The points close to the lower left corner of the plot are more influenced by the end-member A than B and vice versa. The sentences have been modified in order to clarify this concept.

Fig 5: Do the symbol colors mean something? What is diamictite debris? It is not mentioned anywhere in the text.

A: The colors simply help distinguish the symbols without any specific meaning. Diamictite debris indicates glacial-derived material (we added this information in the text).

L452: "... four approaches ..." I like this, it shows how difficult it actually is to estimate mineral dust concentrations.

A: We thank the referee for this comment

L465: Why not also nssCa?

A: We thank the referee for this question, as it correctly pointed out a discrepancy between the text and the formula. nssCa is not used as measured Ca is not used: Ca contribution is estimated from Al one using the crustal element-to-Al ratios reported by Henderson and Henderson (2009). As reported in the text, the 3rd method extends the 2nd one, thus correcting K, Ca and Fe using the crustal composition from literature. The formula in the paper has been corrected accordingly.

Fig 6: Please make the lines and symbols a bit more easily distinguishable. Remember also red-green color blindness.

A: We have modified Fig. 6 by replacing the red and green colours with more easily distinguishable colours. We hope that this modification improves the clarity of the figure. We did not use a stacked graph because our aim is to highlight the similarity of the individual trends.

L490-491: "A notable exception occurred in August 2022, when elevated CEFs were observed for multiple elements." I guess you should mention that the time series plot of Si EF is in Fig. S14.

A: We added the suggested mention in the manuscript.

L505-510: Lines 505-510 and Fig S15 show and discuss the mass balance. It is in a very unlogical section, under the title "3.6 Dust aerosol component" and before "3.7 Carbonaceous components". A new mass balance section would be important, it would combine all the chemical and gravimetric analyses, all other sections are explaining the 5 details. I would strongly suggest you give the mass balance discussion its own section and move it so that it is before the pure statistical section 3.9.

A: We added the mass balance section as suggested by the referee: it is now section 3.8.

Fig. S15 should be in the main text in this mass balance section. It is an important figure, it combines all chemistry and gravimetry. And add a subfigure that shows the monthly fractional contributions to gravimetric mass of each component as percents or fractions of one using log scale y-axis. If possible, it would be a boxplot of each component's contribution. But one very weird thing: there appears to be no sulphates! And this even if, according to Fig 3, sulfate concentration is often > 1000 ng m-3. Explain and recalculate the mass balance. Change the colors in the mass balance stacked bars if possible. EC could be black. Add also a table similar to Table 2. It would be a table that shows the gravimetric mass, major ionic compounds (sulfate, ammonium, (nitrate?,) sea salt), mineral dust, POM and EC in different seasons.

A: We moved Fig S15 to the main text and re-coloured it with a more logical palette. We also corrected it to show the ionic components.
This plot shows the gravimetric mass, and the absolute contributions of sulfate, ammonium, nitrate, sea salt, mineral dust, POM and EC in different months and thus seasons, so we think that adding the same numbers as a table is repetitive.

We also inserted the monthly fractional contributions to gravimetric mass of each component as percentage, as suggested by the referee.

Section 3.7. Carbonaceous components Explain somewhere in this section, how the concentrations of POMare calculated from OC.

A: Particulate organic matter (POM) was obtained by multiplying OC for a factor 2.1 as suggested by Turpin and Lim (2001) for non-urban sites.
Turpin, B. J. and Lim, H. J.: Species Contributions to PM2.5 Mass Concentrations: Revisiting Common Assumptions for Estimating Organic Mass, Aerosol Sci. Technol., 35, 602–610, 2001.

SUPPLEMENT Figure S5. Use more different colors and line types for the two reference lines ammonium sulfate and ammonium bisulfate Figure S10.

A: Figure S5 was removed as suggested by the referee and the data is now inserted in Figure 3, with the similar line and type of other Figures.

"Temporal trend of the elemental concentration normalized with respect to the maximum value." What does " normalized with respect to the maximum value " actually mean? Maximum of what?

A: As suggested by the referee, we clarified this concept as follows: " Figure S9 shows the normalized temporal profiles of the analysed elements. For each element, concentrations were divided by the maximum value observed for that element over the study period and expressed as a percentage. This approach enables comparison of the temporal variability among elements with markedly different absolute concentrations. The profiles highlight a clear seasonality, with generally higher relative concentrations in winter and lower values in summer." In the same way, we have changed the label of Figure S9 as follows: "Normalized temporal profiles of the analysed elements. For each element, concentrations were divided by the maximum value observed for that element over the study period and expressed as a percentage. The elements are grouped according to their value of CEF and MEF, which define the specific sources: a) marine, b) and c) crustal, and d) anthropogenic sources."

For instance, I don't really understand how the Sr percentage is higher than that of Na in (a). Give the equation in the text. Why don't you simply give the percentages of the concentration of selected element of the sum of the concentrations of elements? That would be much more relevant.

A:We have modified the main text and the label of Figure S9 to clarify the concept. The goal of these graphs was to show the temporal trends of different elements grouped according to their potential common origin and to allow their comparison even if they had very different concentrations (e.g., Fe was a thousand times more concentrated than Co, and in Figure S9 it is possible to observe the similarities and differences in their time trends). The reviewer's suggestion to report the percentage of each element relative to the sum of all element concentrations does not resolve the comparability issue for elements with very different concentrations. On the other hand, this approach would emphasize only the elements with higher concentrations. The fact that, for a specific sample, one element (e.g., Sr) had a higher percentage value than another (e.g., Na) means that the first element shows a concentration for that sample more similar to the higher concentration value that we found for that element in all samples. This suggests, for example, that the second element shows a higher dispersion of concentration values compared to the first one.

In many FigS10 sublots the percentages go up and down up and down almost in a wavelike pattern. It looks like some artefact. Is there some explanation?

A: The samples have been processed and analyzed randomly with respect to the sampling sequence in order to avoid external influences introducing significant artefacts in the time trends. The spike pattern that can be observed in Figure S9 can be ascribed to the inter-sample variability.

Referee 2

This manuscript reports on a 1-year study of aerosol chemical composition measured at the observatories Gruvebadet (GAL), located at an altitude of 61 metres above sea level, and Zeppelin (ZEP), located at an altitude of 472 metres above sea level, both in Ny Ålesund, Svalbard. Additional meteorological information on boundary layer properties such as turbulence or stratification, were provided by the Amundsen-Nobile Climate Change Tower. The rich data set contains information on aerosol optical properties, ion composition, trace elements, lead isotopes, meteorological data, and air mass back trajectories from February 2022 to March 2023. The analysis of the simultaneously taken measurements focused on the identification of the main drivers of aerosol variability in the Arctic atmospheric boundary layer (ABL). In that context the study makes a significant contribution to this field of research, because it presents the most comprehensive analysis of aerosol chemical and physical properties at the two Ny Ålesund observatories, combined with detailed information on atmospheric dynamics.From its scientific topic, significance and quality, the study deserves publication, and it fits well into the scope of ACP. However, the presentation of results needs major revisions before being acceptable for publication. Detailed comments are given in the following section.

Of general concern is the selection of figures shown in the main text, and those shown in the supplementary material. In the current version, the main text contains mainly presentations of time series or occurrence of elements while the plots showing information from statistical analysis are shifted to the supplementary material. This distribution of information makes the manuscript very difficult to read. Re-thinking the places where the figures are shown, is strongly recommended.

Another major difficulty is related to the use of language. There are so many places where the language needs to be improved that I can only give a few examples. In summary, manuscript revision by a native English-speaking person is strongly recommended.

A: We thank the Reviewer for their positive assessment of the scientific value, scope, and significance of our work, as well as for recognizing the comprehensive nature of our dataset combining aerosol physics, chemistry, and atmospheric dynamics at Ny-Ålesund. As suggested, we have revised the distribution of figures between the main manuscript and the Supplementary Material to improve the manuscript's structure and readability. We have reviewed and polished the entire text to improve language, sentence structure, and overall readability.

SPECIFIC COMMENTS

1| Section 2 on Materials and methods is well written and contains all necessary information to assess the quality of the performed analyses and the corresponding results. In contrast, the results section is very descriptive with lots of text which makes it difficult for the reader to get the important points. Furthermore, mostly time series and plots like a sequence of occurrences of various elements (e.g. Figure 4) are shown. However, from these types of plots no conclusions can be drawn which support the important results of the paper on the study shown in Figure 10. Compared to other figures, Figure 10 is really relevant and should be presented in a more illustrative manner.

A: We thank the reviewer for these helpful comments and for the positive evaluation of Section 2. We would like to underline that this study brings together diverse chemical and physical datasets collected by several universities and research institutes. We organized the "Results section" into specific subsections for each type of measurement so that each dataset could be described and interpreted individually first.

These individual observations and time series provide the foundation for the final integrated analysis. All datasets were subsequently combined using a Partial Least Squares Discriminant Analysis (PLS-DA) model, selected as the best chemometric approach, to address our primary objective: understanding how atmospheric layering influences aerosol variability in the Ny-Ålesund area.

In response to your suggestions, we have improved the description of Figure 10 as follows:
"The discrimination performance and variable contributions were evaluated by combining the coefficient plot (Fig. 10A), which indicates the directional association with each site, and the Variable Importance in Projection (VIP) plot (Fig. 10B), which quantifies the overall importance of each variable.

As shown in Fig. 10A, the two sites present distinct chemical signatures. GAL is characterized by positive regression coefficients, strongly associated with Cd, Cr, and V, which act as the primary discriminating tracers. In contrast, ZEP shows negative coefficients, strongly associated with trace elements such as As, Co, Cu, along with biogenic markers (arabitol and mannitol) and some trace elements (Pb, Mn, and Ni).

Considering VIP scores (Fig. 10B), several variables exceed the significance threshold (VIP>0.8). Notably, major ions such as K (VIP>1.8) and Na (VIP>1.3) show the highest importance in the model. Even though their regression coefficients (Fig. 10A) are close to zero. This suggests that major ions are crucial for the general discrimination between GAL and ZEP because these species are abundantly present at both altitudes but their overall concentration variations establish the core baseline of the aerosol matrix, allowing the model to effectively distinguish between the two atmospheric regimes. Specific trace elements and biogenic markers serve as site-specific tracers that reflect the vertical stratification between GAL and ZEP."

2| The panels of Figure 11 also contain important results of the study. However, this is hidden by naming the axes as "Factor 1" and "Factor 2" This naming is meaningless, and even worse, the title axis descriptions have been used for both panels but with a different meaning of Factor 1 and Factor 2. This needs to be changed by giving the axes meaningful titles.

A: In PLS-DA, "Factor 1" and "Factor 2" represent the latent variables computed mathematically by the model to maximize discrimination between sites, rather than individual physical parameters. Because each factor integrates contributions from the entire multi-variable dataset, assigning qualitative or conceptual titles to these axes would be speculative and scientifically inaccurate. However, to make the factors immediately meaningful to the reader without altering the axes, we have updated the caption of Figure 11 to explicitly include the percentage of explained variance for Factor 1 and Factor 2 in each panel. Additionally, the "Percent of variance" obtained by Partial Least Squares Discriminant Analysis (PLS-DA) remains available in Table S2 of the Supplementary Information.

3| The paragraph starting on line 389 presents an important result of the study, but this is neither supported by a graph another piece of information providing the evidence of the drawn conclusions. This is only one example of the fair presentation quality of the manuscript. There are more examples of that kind.

A: *We thank the Reviewer for this comment. We would like to clarify that the interpretations presented in the paragraph indicated by the Reviewer are directly supported by the results reported in the Supplementary Information and explicitly cited in the same paragraph. In particular, Figure S10 shows the temporal trends of elemental concentrations, Figure S8 reports the crustal enrichment factors, and Figure S7 reports the Spearman's ρ correlation coefficients between elemental concentrations. Moreover, the correlation between Lu and V is explicitly reported in the text (Spearman's $\rho = 0.48$, $p < 0.05$), while the interpretation of V as a tracer of fuel-oil combustion and the possible association of Lu with refinery emissions are supported by the*

cited literature. We therefore believe that the evidence supporting the discussion is already provided and explicitly linked to the corresponding interpretations in the text.

4| The Conclusions section is not in line with the current requirements of ACP. The guidelines for authors (https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html) state that the concluding section needs to contain a summary of the main results, synthesis and interpretation, comparison and context, caveats and limitations and finally a section on implications where the authors should discuss what the results mean for our understanding of the state and/or behaviour of the atmosphere and climate, which is the main requirement for publication in ACP. Please rework the conclusions section accordingly.

A: We thank the reviewer for pointing this out and highlighting the ACP structural guidelines. We fully agree with this recommendation and have thoroughly restructured and expanded the Conclusions section to strictly adhere to the five required pillars outlined by the journal reported to the suggested link. Furthermore, we removed redundant sentences throughout the section.

The revised section now is as follows (here we reported also the title of each pillar only to be clear):

1. Summary of Main Results: "The BETHA-NyÅ project successfully established a harmonized, multi-altitude aerosol monitoring framework in Ny-Ålesund, linking ground-level observations at the GAL Observatory, elevated measurements at the ZEP Observatory (ZEP), and meteorological profiles from the Amundsen-Nobile Climate Change Tower (CCT). We observed a high degree of seasonal coherence between GAL and ZEP for anthropogenic markers associated with Arctic Haze. Sulphate and ammonium exhibited nearly identical temporal trends, peaking in March–April with remarkably consistent median sulphate concentrations (203 ng m^{-3} at GAL and 220 ng m^{-3} at ZEP). Optical properties similarly aligned during the cold season (median scattering coefficients of 4.2 Mm^{-1} at GAL and 5.5 Mm^{-1} at ZEP). Conversely, biogenic tracers (arabitol and mannitol) displayed pronounced vertical gradients, with significantly higher concentrations at GAL compared to ZEP. Lead (Pb) concentrations at GAL averaged 174 pg m^{-3}, with isotopic ratios ($^{208}Pb/^{206}Pb \approx 2.10$ and $^{207}Pb/^{206}Pb \approx 0.86$) pinpointing persistent Eurasian and Russian origins."

2. Synthesis and Interpretation: "These findings demonstrate that long-range transport of anthropogenic pollution routinely overrides local vertical thermal stratification during haze episodes, resulting in a well-mixed lower atmosphere across different elevations. In contrast, primary biological aerosol particles (PBAPs) originate predominantly from local terrestrial or marine surface sources and remain trapped within the shallow, stable ABL, rarely reaching the elevated intake at ZEP. Thus, vertical distribution in Svalbard is governed by a dual regime: strong vertical homogenization for long-range transported fine aerosols versus boundary-layer confinement for local surface emissions."

3. Comparison and Context: "Our observed lead concentrations and isotopic signatures are in direct agreement with long-term monitoring records in Svalbard spanning 2010–2020. This continuity confirms that the dominant atmospheric pathways and regional source allocations transporting heavy metals into the High Arctic have remained stable over the past decade, despite shifting geopolitical and industrial dynamics in Eurasia. Furthermore, our cross-site comparison validates the robust performance of the BETHA-NyÅ harmonization protocol for intercalibrating distinct observational platforms in polar environments."

4. Caveats and limitations: "While this study demonstrates the value of coordinated multi-altitude sampling, specific observational and meteorological constraints should be noted. Offline chemical characterization inherently limits the capability to resolve short-lived atmospheric events or rapid boundary layer turnover occurring on sub-daily timescales. Vertical meteorological profiles were not continuously coupled with

aerosol sampling across all transitional phases, leaving minor uncertainties regarding the exact height and strength of atmospheric inversions relative to the upper site elevation."

5. Implications: "These results have important implications for our understanding of Arctic atmospheric dynamics and climate feedbacks linked to Arctic Amplification. The accumulation of biogenic aerosols within the lower ABL directly influences the concentration of ice nucleating particles (INPs) and the formation of low-level cloud droplets, which alters the local surface energy balance. At the same time, the uniform vertical distribution of anthropogenic pollution suggests that light absorbing particles can be deposited at different altitudes. Accounting for this vertical contribution helps to reduce uncertainties in climate models and to predict snow-albedo feedback in the High Arctic Environment."

MINOR ISSUES

Section 2: sometimes company names include a country of origin, sometimes not. This should be harmonised.

A: We added country of origin into section 2.

Line 153: What does "(2.195 10-4) (Hans Wedepohl, 1995)" mean? Furthermore, the reference is not included in the list.

A: Sorry for the mistake. "-4" has to be superscript. 0.0002195 corresponds to the Pb to Al ratio in the upper continental crust. We also added the reference.

Line 168: Were the quartz fibre filters used for aerosol carbonacous matter analysis pre-treated before being exposed, e.g. by a thermal pre-treatment?

A: Quartz fibre filters were not pre-treated, as thermal pre-treatment indeed cleans them but may also activate them enhancing absorption. Nevertheless, after investigating the blank uniformity on a batch of filters, we decided to use an innovative procedure for blank estimation and correction: sampling is performed on a 25 mm diameter filter obtained as a round punch from a larger 47 mm diameter filter. This allows us to analyse a punch from both the sampled filter and the un-sampled remaining part of the larger filter, thus correcting every sample with its own blank. The 25 mm diameter filter is cut just before the sampling, and the remaining part is stored in a petri that travels together with the corresponding sample: thus, this correction takes into account also for possible contamination occurring during handling and travel to/from the Arctic.
Further, the concentration of the flow on a smaller area (25 mm instead of 47 mm diameter) increases the sensitivity of the technique, as Thermo-Optical Analysis is sensitive to the areal density and not to the total amount of carbon on the filter.

Line 227: Do the authors mean PM2.6 or is this a typo and it should be PM2.5?

A: We corrected the mistake.

Results section: The colours for data from GAL and ZEP should be similar. Actually GAL data are mostly shown in red, while ZEP data are sometimes shown in grey, and sometimes in black.

A: Thanks for the suggestion. We corrected figures into the manuscript.

Line 370: in the sentence "Among the REE (Fig. S10c), Lu showed …" is the term "Lu" referring to a person or to the element Lutetium. I am sure the latter is the case, but then it should be made clear, e.g., by adding "the element" before "Lu".

A: As suggested by the referee, we added "the element" before "Lu".

TYPOS AND LANGUAGE USE (not comprehensive)

Line 42: The sentence “In particular, the Arctic near-surface temperature is increasing about 2-3 times faster than the global average and it is called “Arctic Amplification” … sounds unusual and might be changed to “ …. faster than the global average. This phenomenon is called “Arctic Amplification” …”.

A: Done

Line 46: “… because ABL can impact on the transport of …” needs to be rephrased. One suggestion is:” … because processes in the ABL can affect the transport of …”.

A: Thanks for the suggestion.

Line 68: The sentence “ZEP is less likely to be affected by local anthropogenic sources than GAL due to its location and by local air flow phenomena such as katabatic winds” is difficult to understand. One easy-to-understand option is: “Due to its location, ZEP is less likely to be affected by local anthropogenic sources than GAL and by local air flow phenomena such as katabatic winds.” It is only changing the structure of the sentence to make it easier to understand.

A: We modified it as suggested by the referee 2.

Line 74: “The sequence “… plays a relevant role on climate …” needs to be rephrased. One suggestion is “Aerosols play an important role in the climate …”.

A: Thanks. We modified it.

Line 78/79: TH term “but limited information … were produced” sounds unusual. It should be rephrased as, e.g., “but only limited information … is available.”

A: Done.

Line 88: The phrase “to assess how vertical gradient effects aerosol composition in the Arctic” is difficult to understand. It should probably be “to assess how vertical gradients of boundary layer properties driving horizontal and vertical transport may affect aerosol composition in the Arctic.”

A: Thanks for the suggestion.

Line 216: remove “so”.

A: Done.

Line 226: Correct as “Ion Chromatography”.

A: Done.

I stop here but recommend a careful language check.

A: Thanks for improving the manuscript.