

Barbaro et al.: Drivers of aerosol variability in the high Arctic: insights from integrated observations at Gruebadet and Zeppelin (Ny-Ålesund)
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Review

General

The paper presents results and analyses of aerosol measurements conducted at two observatories Gruebadet (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.

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 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 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 stretch over several orders of magnitude. Typically at field stations, clear contamination sectors 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.

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

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 (\pm 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 visibility. Just at the core of haze research and of your paper, right?

Detailed comments

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

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.

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 actually BC concentration 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.

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

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

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.

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

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.

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

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.

L266-278: This comparison of the of scattering and absorption coefficients at 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 $\text{abs}(\text{GAL})$ vs $\text{abs}(\text{ZEP})$ and $\text{scat}(\text{GAL})$ vs $\text{scat}(\text{ZEP})$ and a linear-scale $\text{SSA}(\text{GAL})$ vs $\text{SSA}(\text{ZEP})$ 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.

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

Fig. 3: In Feb 2022 there were very big GAL-ZEP differences for both SO_4 and NH_4 . 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.

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) $\text{EF} = (\text{Metal}/\text{RE})_{\text{soil}} / (\text{Metal}/\text{RE})_{\text{background}}$

Where, RE is the value of metal, adopted as Reference Element"

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.

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

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

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. So either write it as mineral dust or give definition here.

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.

L429: Explain the concept of end member.

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

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

L465: Why not also nssCa?

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

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.

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

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.

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 $> 1000 \text{ ng m}^{-3}$. 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.

Section 3.7. Carbonaceous components

Explain somewhere in this section, how the concentrations of POM are calculated from OC.

SUPPLEMENT

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

Figure S10. "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? 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.

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