



1 Drivers of aerosol variability in the high Arctic: insights from 2 integrated observations at Gruvebadet and Zeppelin (Ny-Ålesund)

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24 **Abstract.** This paper summarizes the main results from the scientific project “Boundary layer Evolution Through
25 Harmonization of Aerosol measurements at Ny-Ålesund research stations” (BETHA-NyÅ), in which aerosol measurements
26 of two Arctic atmospheric observatories located near Ny-Ålesund (Svalbard) at different elevations were harmonized: at the
27 Gruvebadet atmospheric laboratory (61 m a.s.l.) and Zeppelin observatory (472 m a.s.l.). This approach allows for a better
28 understanding of how atmospheric layering may affect the variability of aerosol observations in the Ny-Ålesund area. From
29 February 2022 to March 2023, a coordinated sampling campaign enabled a direct comparison of optical, chemical, and physical
30 aerosol properties, integrated with meteorological data from the Amundsen-Nobile Climate Change Tower. Results reveal a
31 strong seasonal coherence between the two sites for two topical markers such as sulfate and ammonium, with clear evidence
32 of the winter–spring Arctic Haze phenomenon. Local differences emerged mainly for biogenic tracers (e.g., arabinol and
33 mannitol) which were detected at higher concentrations at Gruvebadet compared to Zeppelin observatory, highlighting the role
34 of near-surface sources and aerosol stratification. The analysis of trace elements, lead isotopic ratios, and organic markers
35 helped us to distinguish natural from anthropogenic contributions, confirming the dominant role of long-range transport and
36 the persistence of isotopic signatures consistent with Eurasian sources. The systematic comparison across the two observatories



37 demonstrates the robustness of the harmonized protocol and emphasizes the importance of an integrated monitoring network
38 for evaluating the evolution of atmospheric processes in the Arctic.

39 **1 Introduction**

40 The Arctic is undergoing the most severe effects of the current climate change, amplifying and driving changes elsewhere
41 in the Earth system. In particular, the Arctic near-surface temperature is increasing about 2-3 times faster than the global
42 average and it is called “Arctic Amplification” (Serreze and Barry, 2011). The peculiar feedbacks between climate forcings
43 and environmental responses especially involve large changes in surface albedo, over land, sea and long-range transport
44 patterns of air pollutants (IPCC, 2013). A detailed understanding of atmospheric processes at different scales can help to define
45 the main causes of “Arctic Amplification”. In this scenario, knowledge of the vertical structure of the Arctic Boundary Layer
46 (ABL), which is the turbulent lowest part of the Arctic atmosphere, is essential because ABL can impact on the transport of
47 different type of aerosols and consequent deposition on the glaciers and sea ice surfaces (Jozef et al., 2024).

48 The Svalbard archipelago is Norway’s northernmost region and one of the northernmost land areas in the world. The Ny-
49 Ålesund research settlement is located on the Brøgger Peninsula on the shore of Kongsfjorden, in western Spitsbergen, the
50 largest island at Svalbard, and hosts several national and international research institutions with long-term monitoring programs
51 and research projects. This site offers an excellent opportunity to investigate the ABL due to the co-location of three key
52 facilities: the Gruvebadet Atmospheric Laboratory (GAL), the Zeppelin Observatory (ZEP), and the Amundsen-Nobile
53 Climate Change Tower (CCT). GAL is located 800 m south-west of Ny-Ålesund, dedicated to the chemical and physical
54 characterization of atmospheric aerosols, with continuous monitoring activities beginning in 2010, and are still ongoing
55 (Barbaro et al., 2024; Becagli et al., 2019; Conca et al., 2019; Feltracco et al., 2020, 2021; Giardi et al., 2016; Grotti et al.,
56 2024; Udisti et al., 2016). In addition, GAL has been the location for release of tethered balloons equipped with an optical
57 particle counter (OPC), a black carbon (BC) monitor, and meteorological sensors to investigate size-resolved particle
58 properties within and above the boundary layer (Mazzola et al., 2016).

59 ZEP is located at the top of Zeppelin Mountain (474 m asl), approximately 3 km from the coast of the fjord and 1 km
60 from GAL. The monitoring activity is linked to several different international programmes and research infrastructures, such
61 as EMEP (the European Monitoring and Evaluation Programme under LRTAP, UNECE), AMAP (Arctic Monitoring and
62 Assessment Programme), ACTRIS (European Research Infrastructure for the observation of Aerosol, Clouds, and Trace
63 gases), SIOS (Svalbard Integrated Arctic Earth Observing System), ICOS (Integrated Carbon Observation System) and
64 WMO/GAW (World Meteorological Organization/Global Atmosphere Watch). Continuous monitoring has been carried out
65 since 1989, although the first aerosol measurements in Ny-Ålesund date back to 1974 (Platt et al. 2022). The daily operation
66 at the station is performed by the Norwegian Polar Institute (NPI), while NILU and Stockholm university (SU) are responsible
67 for the scientific activities, depending on the type of measurement (Table 1). ZEP is less likely to be affected by local



68 anthropogenic sources than GAL due to its location and by local air flow phenomena such as katabatic winds (Ström, et al.,
69 2009).

70 The CCT was installed in 2009, about 2 km west of Ny-Ålesund on the southern coast of Kongsfjorden. The 34 m high
71 tower hosts standard meteorological sensors at different heights, sonic anemometers coupled with fast gas sensors, radiation
72 sensors, as well as snow and ground sensors at its base. The CCT was conceived to host scientific experiments and instruments
73 devoted to studying ABL dynamics under different atmospheric conditions (Mazzola et al., 2016).

74 Aerosol plays a relevant role on climate by scattering and absorbing the solar radiation and by influencing cloud formation
75 (i.e. cloud condensation nuclei). Aerosol particles are transported from the middle latitude influencing the composition of the
76 Arctic atmosphere, with consequent effects on cloud formation, albedo or sea ice. Several aspects remain poorly known,
77 representing the challenge of the recent aerosol research. For example, organic compounds have notable consequences for
78 atmospheric chemistry and cloud formation, but limited information about the sources of key compound classes such as sugars
79 were produced. On the other hand, the positive forcing of black carbon (BC) is well known because it enhances light absorption
80 processes in the atmosphere, especially in the Arctic, and after its deposition over the glaciers, where it triggers and accelerates
81 melting processes. However, the dynamics of BC entertainment in the ABL are still a poorly understood process and may have
82 a wide variability depending on local conditions (Gilardoni et al., 2020). Studying the ABL is crucial as even small changes
83 in its thermodynamic structure or stability can strongly influence aerosol dispersion, cloud formation, and surface radiative
84 balance. It plays a key role in regulating the exchange of energy, moisture, and aerosols between the surface and the free
85 troposphere. These processes ultimately determine how aerosols affect Arctic amplification and ice–albedo feedback
86 mechanisms (Gilardoni et al., 2020).

87 The key objective of BETHA-NyÅ project was to set up an aerosol measurement inter-comparison experiment between
88 the GAL and ZEP observatories to assess how vertical gradient effects aerosol composition in the Arctic. Composition data
89 from the two observatories were integrated with meteorological information from the CCT and radio soundings. Long-term
90 harmonization of sampling strategies is highly beneficial for assessing how atmospheric layering affects the chemical
91 signatures observed at different heights in the Arctic.

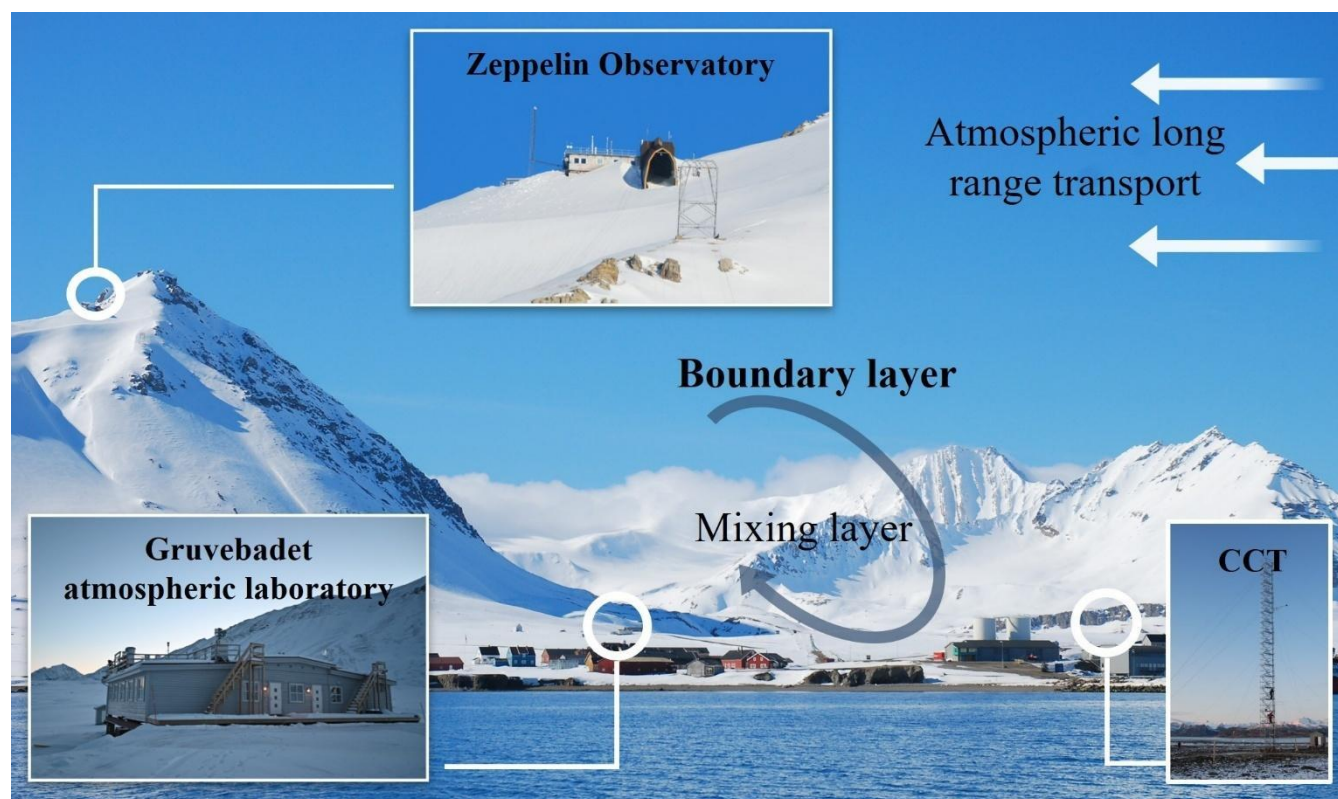
92 **2 Material and methods**

93 Ny-Ålesund is a unique location to investigate ABL processes thanks to the availability of two facilities at the same place but
94 at different altitudes: GAL (78.918°N, 11.895°E) at 61 m above sea level and ZEP Observatory station (78.908°N, 11.881°E)
95 at 472 m above sea level (Fig. 1) less than 2 km from each other. Moreover, data from the CCT enabled us to assess how local
96 near surface processes influence boundary layer dynamics, thermal stratification, and surface-atmosphere interactions affect
97 aerosol concentration, composition, and transport between the two monitoring sites.

98 The instrumental set-up and sampling parameters at GAL were adjusted to match, as closely as possible, the procedures used
99 at ZEP. The harmonized protocol was implemented on 21 February 2022, and this paper presents results from the first sampling



100 period, which continued until March 2023; the protocol remains in use at present. Table 1 summarizes the harmonized
 101 measurement set-up at both sites.
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Figure 1. Sampling locations at Ny-Ålesund: Gruvebadet atmospheric laboratory (GAL), Zeppelin Observatory (ZEP) and Amundsen-Nobile Climate Change Tower (CCT).

Table 1. Summary of details of harmonized measurements in both sites: Gruvebadet atmospheric laboratory (GAL) and Zeppelin (ZEP) Observatory.

Measurements	GAL	ZEP
Absorption coefficient	Managing: ISP-CNR Sampler: aethalometer at seven wavelengths (AE33, Aerosol Magee Scientific), cut-off TSP Sampling resolution: 1 hour	Managing: SU Sampler: Multi Angle Absorption Photometer (MAAP, Magee Scientific Corporation), cut-off TSP Sampling resolution: 1 hour
Major inorganic ions	Managing: University of Florence	Managing: NILU Sampler: Three stage open filter face



	<p>Sampler: PM₁₀ low-volume sampler (TECORA Skypost), flow 38.3 L min⁻¹</p> <p>Filters: 47 mm diameter PTFE filters</p> <p>Sampling resolution: 24-48 hours</p>	<p>Filters: 47 mm diameter Teflon filters (Zefluor PTFE)</p> <p>Sampling resolution: 24 hours, change at 07:00 UTC.</p>
Trace elements, isotopic and elemental analysis	<p>Managing: University of Turin</p> <p>Sampler: PM₁₀ high volume (TECORA Skypost), flow 150 L min⁻¹</p> <p>Filters: 90 mm diameter PTFE filters</p> <p>Sampling resolution: 1 week</p> <p>Managing: INFN</p> <p>Sampler: PM₁₀ low-volume sampler (TECORA Skypost), 38.3 L min⁻¹</p> <p>Filters: 25 mm diameter ring-supported, stretched PTFE</p> <p>Sampling resolution: 1-2 days</p>	<p>Managing: NILU</p> <p>Sampler: PM_{2.6} high-volume (DIGITEL DH-77), flow 43.8 L min⁻¹</p> <p>Filters: 150 mm diameter Whatman 41</p> <p>Sampling resolution: 48 hours</p>
EC/OC	<p>Managing: INFN</p> <p>Sampler: PM₁₀ low-volume sampler (TECORA Echo-PM), flow 38.3 L min⁻¹</p> <p>Filters: 25 mm diameter quartz fiber filters</p> <p>Sampling resolution: 1-2 days</p> <p>Managing: ISP-CNR</p> <p>Sampler: PM₁₀ high-volume Andersen sampler (TE-6070 series, Tisch Environmental Inc.), flow 1140 L min⁻¹</p> <p>Filters: quartz fiber filters</p> <p>Sampling resolution: 1 week</p>	<p>Managing: NILU</p> <p>Sampler: PM₁₀ Digitel high-volume sampler (DH-77, DIGITEL), flow 666 L min⁻¹</p> <p>Filters: Quartz fiber filters (WHATMAN 40 QM-A)</p> <p>Sampling resolution: 1 week</p>
Organic tracers	<p>Managing: ISP-CNR</p> <p>Sampler: PM₁₀ high volume Andersen sampler (TE-6070 series, Tisch Environmental Inc.), flow 1140 L min⁻¹</p> <p>Filters: quartz fiber filters</p> <p>Sampling resolution: 1 week</p>	<p>Managing: NILU</p> <p>Sampler: PM₁₀ Digitel high-volume sampler (DH-77, DIGITEL)</p> <p>Filters: Quartz fiber filters (WHATMAN 40 QM-A)</p> <p>Sampling resolution: 1 week</p>



110 2.1. Sampling at Gruvebadet atmospheric laboratory and sample processing

111 Gruvebadet atmospheric laboratory (GAL) is located 800 m from Ny-Ålesund at an elevation of 61 m above sea level. The
112 aerosol observations are conducted by CNR-ISP together with several Italian universities (Ca' Foscari University, Universities
113 of Florence, Genoa, and Turin) and research institutions, such as National Institute of Nuclear Physics (INFN).

114 An aethalometer at seven wavelengths (AE33, Aerosol Magee Scientific) was installed to measure the absorption coefficient
115 (σ_{abs}) and it is managed by ISP-CNR. Absorption data were corrected for multiple scattering using a correction coefficient (C)
116 (Yus-Díez et al., 2021). Absorption coefficients at 530 nm were derived from the coefficient measured at 520 nm and using
117 the Ångström Absorption Exponent (AAE) derived from the seven measured wavelengths. The scattering coefficient (σ_{sca}) at
118 530 nm was measured using a single wavelength integrating nephelometers (M903, Radiance Research, USA). Both σ_{abs} and
119 σ_{sca} were averaged at one-hour time resolution.

120 A low-volume sampler (TECORA Skypost), managed by Florence University, collected one-to-two-day PM_{10} aerosol samples
121 on 47 mm diameter PTFE filters (Pall Corporation and Cobetter Filtration Group). These filters were used to define PM_{10} mass
122 and ionic composition. These samples were prepared under a laminar flow hood in Florence and shipped to Ny-Ålesund; after
123 sampling, the filters were stored in a freezer at “Dirigibile Italia” Station and then shipped back to Florence together with field
124 blanks. The filters were cut into two parts; one half is archived for further analysis while the other half is devoted to ion
125 chromatographic analysis. The PM_{10} mass was determined by weighing the filter before and after the sampling using a five-
126 digit microbalance (Sartorius ME235P). The filters were conditioned for 48 h (25 °C and 50% RH) before weighing. The
127 portion of the filter devoted to chemical analysis was diluted with 10 mL of ultrapure water (18 M Ω ·cm, Millipore MilliQ
128 grade) and extracted in an ultrasonic bath for 20 minutes. Ion composition was measured by two Ion Chromatographic systems
129 performing the analysis of inorganic anions and inorganic cations. The detailed procedure is described in Becagli et al. (2011).
130 For all the parameters reproducibility on real samples was better than 5%, and filter blanks were found to be lower than the
131 detection limit.

132 An Echo Hi-Vol sampler (TCR Tecora, 150 L min⁻¹ with a week resolution) was used to collect PM_{10} samples on hydrophilic
133 PTFE filters (H100A090C, 90-mm diameter, efficiency > 99% for 0.3 μm particles; Advantec MFS, Dublin, CA, USA). These
134 samples were split into two different sections. The University of Genoa performed the determination of lead content and
135 isotope ratios, while trace elements and rare earth elements were determined by the University of Turin.

136 A quarter of each PM_{10} high volume filter for trace elements analysis and Pb isotopic analysis underwent microwave-assisted
137 digestion using a vessel-inside-vessel method, following EU air quality monitoring standards. Filters were cut with ceramic
138 scissors pre-washed with acidic solution. The digestion was carried out at 220 °C with a mixture of HNO_3 , H_2O_2 , and high-
139 purity water (HPW). Post-digestion, solutions were filtered, cleaned, and diluted to 10 mL. All steps occurred in a clean Class-
140 100 laminar flow hood. Reagent blanks and sample blanks (from transported blank filters) were used to correct background
141 contamination. Instrumental blanks and calibration checks ensured analytical accuracy, with <5% relative standard deviation
142 per element. Certified Reference Material (NIST 1648a) confirmed recoveries of over 80% for most elements. All details about



143 the sample processing are reported by Conca et al. (2019). The elemental quantification was performed by an iCAP PRO XP
144 ICP-OES (Thermo Fisher Scientific, Waltham, MA, USA), an Element 2 SF-ICP-MS (Thermo Fisher Scientific) and an 8900
145 ICP-MS/MS (Agilent, Santa Clara, CA, USA) according to the element concentrations.

146 The PM₁₀ high volume samples for lead isotopes were analyzed as described in previous works (Bazzano et al., 2015; Grotti
147 et al., 2023). Specifically, after performing the digestion, the samples were diluted to 10 mL with ultrapure water (Milli-Q
148 from Merck Millipore) and analyzed by inductively coupled plasma mass spectrometry using a NexIon 2000 ICP-MS unit
149 (Perkin Elmer, Waltham, MA, USA) for the determination of lead content and isotope ratios.

150 The concentration of Al was measured by inductively coupled plasma atomic emission spectrometry (Vista PRO Varian,
151 Springvale, Australia) and used to calculate the enrichment factors (EFs) following the equation:

$$152 \quad EF = (Pb/Al)_s / (Pb/Al)_{UCC} \quad (1)$$

153 Here, (Pb/Al)_s is the ratio measured in each sample and (Pb/Al)_{UCC} is the mean ratio for the upper continental crust (2.195 10-
154 4) (Hans Wedepohl, 1995).

155 Another low-volume sampler (TECORA Skypost), managed by INFN, collected one day PM₁₀ aerosol samples to define the
156 elemental composition. The sampler was customized with a reducing sampling spot inlet, designed and realized at INFN
157 Florence in order to get a sample deposit of about 2.1 cm diameter. The use of this reducer enhances the sensitivity of the
158 analytical technique, namely Particle Induced X-ray Emission (PIXE), as it is sensitive to the areal density of the sample, thus
159 it benefits from an increased concentration of the sample.

160 PM₁₀ samples for elemental determination using PIXE were analyzed without any sample pre-treatment, thus minimizing any
161 contamination risk. They were bombarded with a 3 MeV proton beam extracted in air using the set-up dedicated to
162 environmental analyses at the INFN-LABEC 3MV Tandatron accelerator (Chiari et al., 2021). Spectra of the emitted x-rays
163 were analysed using the GUPIXWIN code (Campbell et al., 2021), and to get the areal concentrations of the elements with
164 Z>10. Atmospheric concentrations were then obtained multiplying the areal density for the deposit area and dividing for the
165 sampled air volume.

166 A sampling strategy configuration with a flow-reducing sampling spot inlet was used for collecting samples for elemental (EC)
167 and organic carbon (OC) analysis (Caiazza et al., 2021). A low volume sampler (Tecora Echo-PM), equipped with reducers,
168 was used to collect PM₁₀ samples on 25 mm diameter quartz fiber filters. Before sampling, each 47 mm filter was punched to
169 obtain a 25 mm diameter filter, which was then placed in the sampler and used for aerosol collection. The remaining outer
170 portion of the 47 mm filter was kept and treated as a blank corresponding to each sample. After sampling, a 1.5 cm² punch was
171 taken from the exposed 25 mm filter for analysis. An equivalent 1.5 cm² punch was also taken from the corresponding 47 mm
172 blank corona. Both punches were analyzed by thermal-optical analysis (Sunset Laboratory Inc, USA), using the NIOSH-870
173 (Quartz of the National Institute for Occupational Safety and Health) protocol. It has been selected because this protocol
174 provides better agreement between EC measured on washed and untreated quartz filters compared to other available thermal
175 protocols (Giannoni et al., 2016, and references therein) and this is especially important for samples where the amount of EC



176 is very low. The value of every blank filter was subtracted from the corresponding sample, in order to get an accurate estimation
177 and subtraction of the blank values (EC was always below the Detection Limit in blank filters).

178 EC/OC measurements at ZEP were performed using pre-fired (850 °C; 3 h) quartz fiber filters (Pallflex Tissuquartz 2500QAT-
179 UP; 150 mm in diameter) weekly collected by Digitel PM10 high-volume sampler (Yttri et al., 2024). Thermal–optical analysis
180 (TOA) was carried out using the Sunset Laboratory, Inc., OC/ EC aerosol analyzer, operating the instrument according to the
181 EUSAAR-2 temperature program (Cavalli et al., 2010). To fully harmonise the monitoring procedures among ZEP and GAL,
182 the same analytical protocol was used on the weekly samples at GAL, and ECOC analyser by Sunset Lab. Inc. was used as
183 well. Samples collected according to the EC/OC monitoring ongoing since 2011 were measured according to the NIOSH-870
184 thermal protocol (i.e. according to the protocol in use since 2011), to compare and harmonise the data series.

185 A PM₁₀ high volume Andersen sampler (TE-6070 series, Tisch Environmental Inc.) was used at a flow rate of 68 m³ h⁻¹ to
186 collect aerosol samples on quartz fiber filters with a temporal resolution of 7 days. In these samples, ISP and Ca' Foscari
187 University usually determine water soluble organic compounds. In particular, we reported here the results of sugars
188 composition such as levoglucosan like biomass burning tracer or arabitol and mannitol, like fungal spores' markers. In addition,
189 EC and OC determination was also performed by INFN using these samples to obtain an intercomparison between two
190 instruments at GAL and also with ZEP samples. As EC and OC are quantified in ZEP samples using the EUSAAR2 thermal
191 protocol, this was also applied to these GAL samples. Blank values were evaluated as the average on different blank filters
192 (also in this case, EC was always below the Detection Limit in blank filters).

193 Monosaccharides (arabinose, fructose, galactose, glucose, mannose, ribose, xylose), sugar alcohols (erythritol, mannitol,
194 arabitol, sorbitol, xylitol, maltitol, galactitol) and mono saccharide anhydrides (levoglucosan, mannosan and galactosan) were
195 analyzed in weekly filter samples collected with Andersen high volume PM₁₀ sampler. A quarter of each filter was extracted
196 with 15 mL ultrapure water following the already validated procedure (Feltracco et al., 2020). The analytical determination
197 was performed using two published methods through ion chromatograph (Thermo Scientific™ Dionex™ ICS-5000, Waltham,
198 US) coupled to a single quadrupole mass spectrometer (MSQ Plus™, Thermo Scientific™, Bremen, Germany) (Barbaro et
199 al., 2015).

200 Anhydrosugars and sugar alcohols were also determined in the samples collected at ZEP using Digitel high volume PM₁₀
201 sampler. The analytical determination was carried out using ultra-performance liquid chromatography (UPLC) (Vanquish
202 UHPLC, Thermo Fisher Scientific) in combination with Q Exactive™ Plus Orbitrap (Thermo Fisher Scientific). Filter punches
203 (2×1.5 cm²) were extracted with 2 mL tetrahydrofuran (THF) following by evaporation and purification (Yttri et al., 2024).

204 **2.2 Zeppelin Observatory sampling and processing**

205 The ZEP Observatory is owned and managed by the Norwegian Polar Institute. It is located on Mount Zeppelin on the 20 km
206 long and 10 km wide Brøggerhalvøya, 2 km south of Ny-Ålesund.

207 The absorption coefficient (σ_{abs}) measurements reported were recorded using a Multi Angle Absorption Photometer (MAAP,
208 Thermo Fisher Scientific) at ZEP. The instrument is operated by Stockholm University (SU) at standard flow 1 m³ hour⁻¹ and



209 connected to a whole air inlet designed according to ACTRIS recommendations for sites often embedded in clouds
210 (<https://www.actris-ecac.eu/>). The MAAP of ZEP provides absorption coefficients at 637 nm, which were converted to 660
211 nm following the methodology described by Müller et al. (2011). Absorption at 530 nm was then estimated assuming an AAE
212 of 1. The eBC concentrations are calculated using instrument default MAC value $6.6 \text{ m}^2 \text{ g}^{-1}$. Major ions are determined upon
213 sampling with a three-stage filterpack using a devoted NILU filter holder system designed for sampling of particles and gaseous
214 compounds (Aas et al., 2025). The first stage in the air stream is a Zeflour ($2 \mu\text{m}$) filter used to collect the airborne particles
215 containing sulphate, ammonium, nitrate, sodium, calcium, potassium, chloride. The second stage is a filter (Whatman 40)
216 impregnated with KOH so to collect HNO_3 , SO_2 , HNO_2 , HCl, and other volatile acidic compounds. The third stage is an acid-
217 impregnated (oxalic acid) filter (Whatman 40), used to absorb alkaline air components such as ammonia. For the nitrogen
218 species, the sum of the concentration of the species in the gaseous and particulate phase is reported. Namely, for ammonium,
219 concentration reported in the database equals the sum of ammonium collected on the aerosol front filter and ammonia collected
220 on the acid-impregnated filter. Since the filterpack sampler is not equipped with an inertial impactor but a cylindrical vertical
221 air intake, the sampling efficiency for large particles (such as mineral dust, sea spray droplets) is reduced and size cut-off is
222 approximately PM_{10} except for strong sea salt episodes characterised by large particles. After exposure, the aerosol filters (first
223 stage of the pack) undergo an ultrasonic treatment with water in order to obtain a complete extraction before analysis. The
224 alkaline-impregnated filter is extracted by a hydrogen peroxide solution in order to oxidize any remaining sulphite to sulphate
225 while the third stage (acid-impregnated filter) is extracted by a HNO_3 solution. All the ions are analysed using Ion
226 Chromatograph.

227 Sampling of trace elements is done using a Digitel high-volume air sampler with a $\text{PM}_{2.6}$ cut-off, and samples are collected on
228 a weekly basis, using Whatman grade 41 filter papers for 48 hours with an airflow rate of $43.8 \text{ m}^{-3} \text{ hour}^{-1}$. The filters are
229 digested with nitric acid by Ultraclave, a microwave-based decomposition technique. Identification and quantification are
230 performed by ICP-MS, and In is used as an internal standard.

231 Details of the monitoring of at ZEP are found in the annual reports for the Norwegian Environmental agency; Aas et al. (2025)
232 includes inorganic and organic components, while Halvorsen et al. (2025) describe the monitoring programme of trace
233 elements.

234 **2.3 Air mass back trajectories analysis and source regions classification**

235 240 hours long back-trajectories reaching Ny-Ålesund on hourly basis were computed using the off-line, Linux based version
236 5.3.2 of the Hysplit code (Stein et al., 2015). For every hour, trajectories reaching the site at 50, 500, 1000 and 3000 m a.g.l.
237 were calculated in the days ranging from 1st January 2022 to 31st December 2023. This is a standard procedure in our lab which
238 allows us to build up a consistent database of BT in relevant receptor sites for successive uses of any type.

239 The meteorological data supplied to the program were Global Forecast System (GFS) at $0.25 \times 0.25^\circ$ resolution, downloaded
240 in the "ARL packed" format from the US National Oceanic and Atmospheric Administration (NOAA) Air Resources
241 Laboratory (ARL) database.



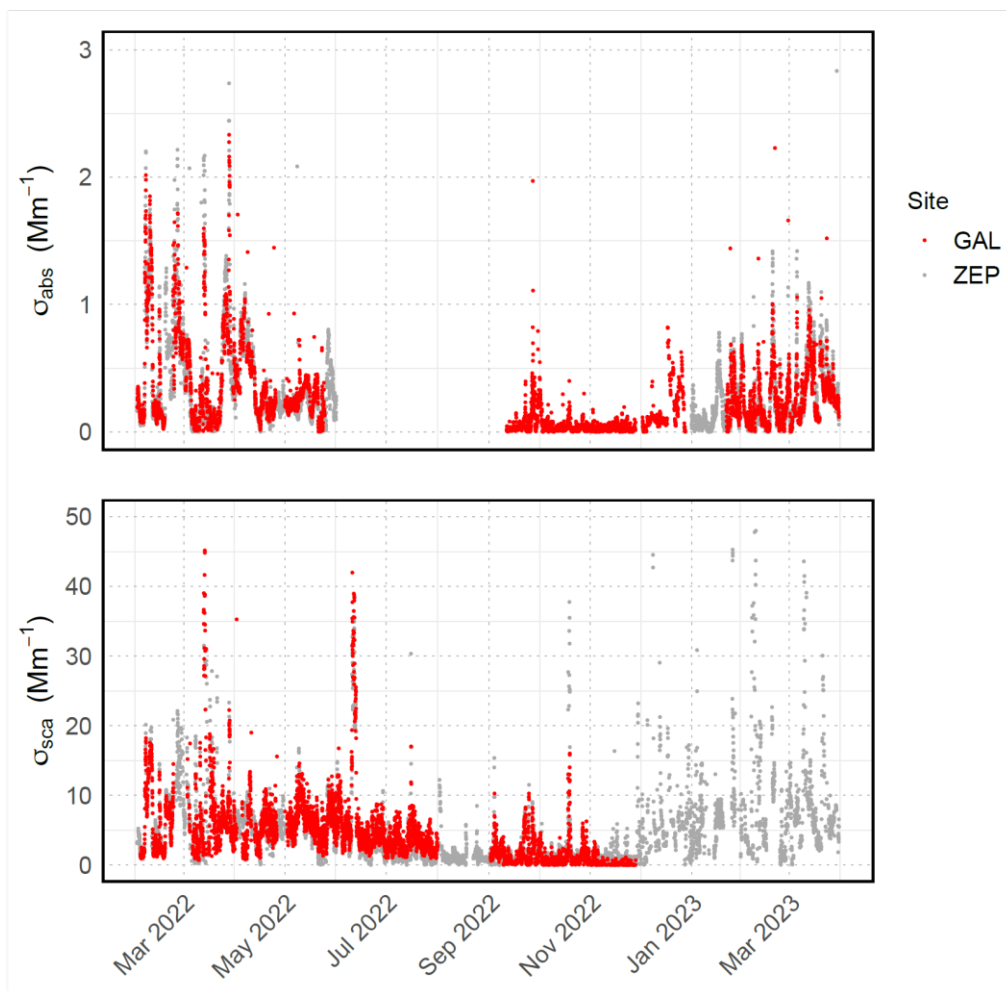
242 Trajectories frequencies, computed as the number of endpoints contained in each cell of a $1 \times 1^\circ$ lat/lon grid for each trajectory
243 reaching the receptor at 50 m a.g.l., were plotted for time ranges matching the sampling periods.
244 NASA FIRMS MODIS satellite (combined Aqua and Terra) data (NASA-FIRMS, 2025) were downloaded and projected onto
245 the frequencies plot as red circles whose diameter was proportional to the pixel fire radiative power. For each sample, fires
246 occurring on a time range including the sampling period and the preceding 10 days were considered.

247 **3. Results and discussion**

248 This section presents a comprehensive analysis of the atmospheric conditions and their variability in the Kongsfjorden region
249 of Svalbard, from February 2022 to March 2023 at the two observatories. An overview of the meteorological and radiative
250 conditions is reported in the Supplementary Material (Sec. S1.1) while the results for atmospheric composition is reported in
251 the following Subsections.



252 **3.2 Aerosol optical properties**



253

254 **Figure 2.** Aerosol absorption coefficients (top) and scattering coefficients (bottom) at 530 nm at GAL (in red) and at ZEP (in gray). A
 255 limited number of datapoints with absorption coefficient above 3 Mm⁻¹ and with scattering coefficient above 50 Mm⁻¹ are not shown in the
 256 figure.

257

258 Figure 2 shows the absorption and scattering coefficients at 530 nm measured at GAL and ZEP. In the cold season (November
 259 to April), the median scattering coefficients were 5.5 Mm⁻¹ (IQR: 2.6 – 8.5 Mm⁻¹) at ZEP and 4.2 Mm⁻¹ (IQR: 1.6-7.3 Mm⁻¹)
 260 at GAL, while in the warm season (May to October) they decreased to 1.6 Mm⁻¹ (IQR: 0.8 – 3.6 Mm⁻¹) at ZEP and to 3.1 Mm⁻¹
 261 (IQR: 1.2 – 5.3 Mm⁻¹) at GAL. The medians of the absorption coefficient were 0.22 Mm⁻¹ (IQR: 0.10 – 0.51 Mm⁻¹) at ZEP
 262 and 0.19 Mm⁻¹ (IQR: 0.09 – 0.44 Mm⁻¹) at GAL, and 0.22 Mm⁻¹ (IQR: 0.16 – 0.29 Mm⁻¹) at ZEP and 0.05 Mm⁻¹ (IQR: 0.02 –
 263 0.21 Mm⁻¹) at GAL, in the cold and warm season, respectively. The interquartile range reveals higher absorption coefficients



264 during the cold season at both sites, a seasonality that is attributed to air mass transport from mid-latitudes and the Arctic haze
265 phenomenon (Gilardoni et al., 2023; Stohl, 2006a).

266 The comparison of two observatories based on parallel measurements of absorption coefficients and scattering coefficients
267 conducted during the study period showed that the differences are centred around zero (Fig. S3).

268 The accuracy of the absorption coefficient measurements, as reported by the manufacturers, is 0.05 Mm^{-1} for AE33 data (1-
269 hour averages) and 0.1 Mm^{-1} for MAAP measurements (30 min averages). Asmi et al. (2021) observed that in ambient
270 conditions the two instruments can measure coefficients as low as 0.012 Mm^{-1} . Considering this value as the uncertainty level,
271 the absolute value of the difference between the measurements would be significant if larger than 0.036. About 45% of the
272 time, the difference of the absorption coefficients between the two sites was below this value.

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

277 The comparison of the optical properties reported (Fig. 2) indicates that the two sites observed similar air masses and/or
278 experienced similar processes for about half of the time during the investigated period.

279 **3.3 Comparison of seasonal trends of sulphate and ammonium between GAL and ZEP**

280 The presented data set covers the entire year 2022 and approximately half of 2023 (1st January 2022 to 27th June 2023). This
281 period includes the full BETHA-NyÅ project, as well as several months before and after, in order to provide an annual
282 overview of the temporal trends.

283 Sulphate and ammonium, as key components of Arctic Haze, exhibit a pronounced seasonal pattern, with a broad maximum
284 beginning in winter (most likely in December) and lasting until June or July, depending on the species (Fig. 3). Thereafter, a
285 relatively flat pattern is observed during summer and autumn.

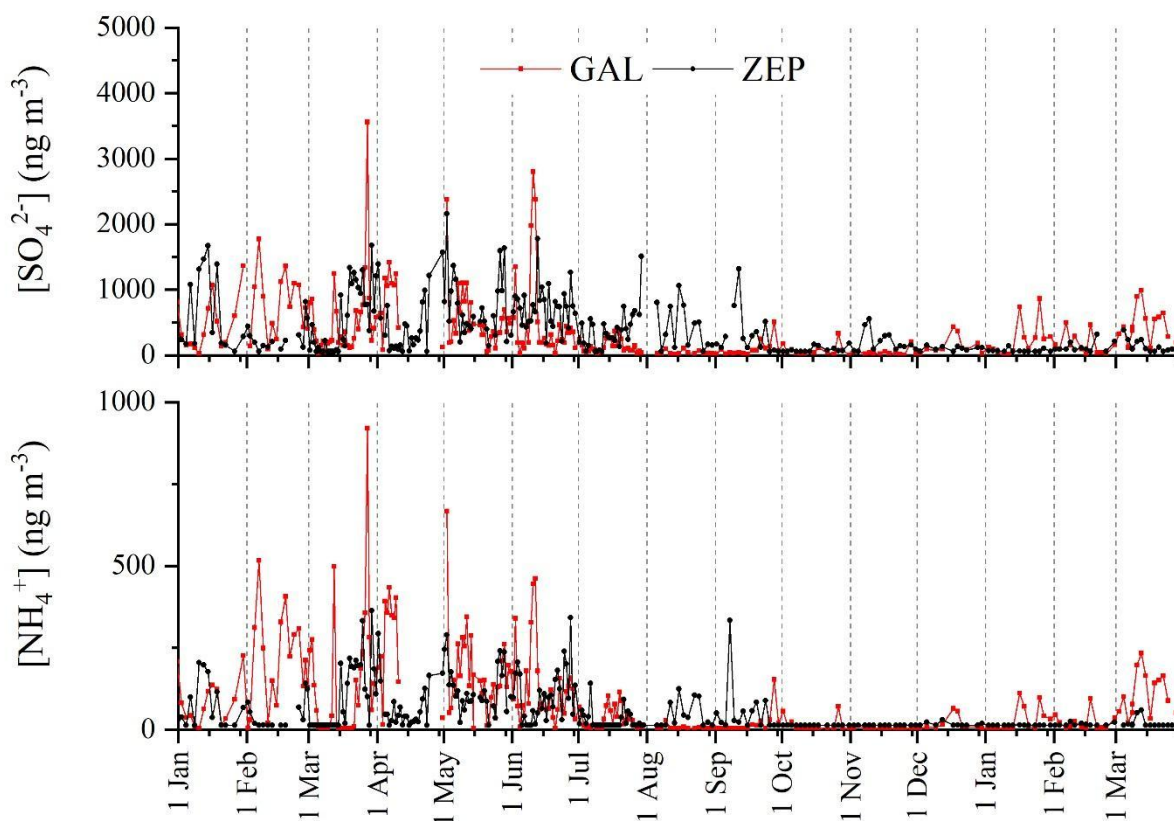
286 Sulphate, in particular, shows remarkably similar temporal behaviour and absolute concentrations at GAL and ZEP throughout
287 the study period (Fig. 3a). The Arctic Haze period is clearly identifiable at both sites. In 2022/2023, sulphate concentrations
288 peak in March-April and return to background levels (defined as the median over the entire study period) around mid-June at
289 GAL and approximately one month later at ZEP. Interestingly, the onset of the winter increase differs between the two sites,
290 beginning in early December at ZEP and in mid-January at GAL. Despite this offset, the highest peaks at both sites occur in
291 March and April, often on the same days.

292 The difference in the timing of the onset and decline of the sulphate maximum may be related to the sites' relative position
293 with respect to the boundary layer during early winter and early summer. While GAL is consistently located within the
294 boundary layer (Rader et al., 2021), ZEP may at times represent conditions of the lowermost free troposphere, although it is
295 most frequently influenced by the boundary layer air masses (Tunved et al., 2013). In terms of absolute concentrations, the
296 agreements between the two sites are striking, despite differences in sampling methodologies. This suggests a high degree of



297 robustness in the measurements. Median concentrations are very similar (203 ng m^{-3} at GAL and 220 ng m^{-3} at ZEP), as are
 298 the averages over the entire study period (381 ng m^{-3} at GAL and 395 ng m^{-3} at ZEP). In contrast, variability appears to be
 299 greater at ZEP.

300



301

302 **Figure 3.** Comparison of seasonal trends of sulphate (a) and ammonium (b) at GAL and ZEP from Jan 2022 to Jun 2023.

303

304 Another parameter that can serve as an indicator of anthropogenic influence, including Arctic Haze, is ammonium. The
 305 temporal profiles of ammonium at the two sites are similar in terms of seasonal behavior (Fig. 3b), with the highest
 306 concentrations observed between January and May-June in both years at both sites. A closer inspection of the plots shows that,
 307 unlike sulphate, the broad concentration maximum ends at approximately the same time at both sites, while the onset is delayed
 308 by about one month at GAL compared to ZEP (mid-January vs. early December).

309 The similar behavior of sulphate and ammonium is consistent with the neutralization of sulphuric acid by ammonia. The extent
 310 of this process is illustrated in Fig. S4, which shows the linear correlation between sulphate and ammonium at GAL and at



311 ZEP. Both correlations are statistically significant at the 99% confidence level, considering the correlation coefficients
312 ($R^2=0.838$ at GAL and $R^2=0.407$ at ZEP) and the large number of data points (> 300 at each site). While the data at ZEP appear
313 quite scattered, GAL exhibits a strong linear relationship between the two species. Notably, the slope of the regression line is
314 approximately 3.6 at both sites, indicating that they are influenced by similar dominant sources of sulphate and ammonium. This
315 also suggests that, for these secondary species, GAL is representative of long-range transport processes despite being located
316 at sea level.

317 The slope value lies between 2.66 and 5.39, corresponding to the sulphate-to-ammonium mass ratios expected for the formation
318 of NH_4HSO_4 and $(\text{NH}_4)_2\text{SO}_4$, respectively. This indicates that, on average, sulphuric acid is largely neutralized by ammonia,
319 forming a mixture of NH_4HSO_4 and $(\text{NH}_4)_2\text{SO}_4$. Previous studies in Ny-Ålesund (Giardi et al., 2016; Udisti et al., 2016, 2020)
320 showed that sulphate is mainly present as ammonium salt, particularly during spring and summer. Those studies, focused on
321 individual years (2013 and 2014), reported lower summer sulphate/ammonium ratios, approximately 2.66, consistent with
322 complete neutralization to $(\text{NH}_4)_2\text{SO}_4$. However, long term data (2010–2019) reported by Traversi et al. (2021) show generally
323 higher ratios at ZEP compared to GAL. Given that sulphate concentrations are similar at both sites while ammonium
324 concentrations are lower at ZEP, the difference in the ratio is primarily driven by ammonium. GAL may be more influenced
325 by local ammonia sources from surface emissions and seabird colonies, as suggested for the Canadian Arctic by Wentworth
326 et al. (2016).

327 Considering the $\text{SO}_4^{2-}/\text{NH}_4^+$ ratio (w/w) at GAL and ZEP over the investigated period (Fig. S5), average values are higher at
328 ZEP than at GAL (9.8 vs. 7.8), as are background values (6.6 vs. 4.1). This pattern is consistent with the long-term observations
329 reported by Traversi et al. (2021). At ZEP, the highest ratios occur during winter-spring period, likely associated with Arctic
330 Haze. In contrast, GAL shows a narrower maximum in March 2022 and April 2023, again, likely linked to Arctic Haze, but
331 also a frequent occurrence of elevated ratios in summer and autumn.

332 This summer-autumn enhancement at GAL cannot be attributed to additional biogenic sulphur sources, as MSA exhibits a
333 distinct seasonal pattern with a clear maximum from May to August (Becagli et al., 2019; Park et al., 2021), which does not
334 explain elevated values from July into winter. The observed difference may instead reflect local sulphuric acid sources
335 affecting GAL during summer and/or analytical artifacts related to differences in detection limits between the two sites.

336 3.4 Elements composition

337 34 elements (Na, K, Mg, Ca, Al, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Sr, Y, Nb, Mo, Cd, Ba, La, Ce, Pr, Nd, Sm, Dy,
338 Er, Tm, Yb, Hf, and Pb) were analysed in PM_{10} filter samples collected at GAL. Their concentrations (Fig. S6) range from a
339 few hundreds of fg m^{-3} for selected rare earth elements (REEs), such as Er, Tm, Yb, and Lu, to tenths of $\mu\text{g m}^{-3}$ for Na, spanning
340 approximately seven orders of magnitude. This wide concentration range reflects both their environmental abundance and the
341 characteristics of active sources in the vicinity of GAL. As expected, sea salt aerosol strongly influences the aerosol
342 composition at Ny-Ålesund, consistent with previous findings (Conca et al., 2019; Zhan et al., 2017).



343 To identify the dominant source categories, namely crustal, marine, and anthropogenic, we calculated crustal (CEF, Fig. S8)
344 and Marine (MEF, Fig. S9) enrichment factors, following Barberi et al. (2016). Elements with median CEF value < 10 and
345 median MEF value > 10 were attributed to crustal sources, whereas those with median MEF value < 10 and median CEF value
346 > 10 were attributed to marine sources. Elements for which both median CEF and MEF values exceeded 10 were classified as
347 anthropogenic.

348 According to this classification, Cr, Ni, Cu, Pb, Mo, Zn, As, and Cd were associated with anthropogenic sources. These
349 elements may originate from fossil fuel combustion, lubricants use, mining activities, and non-ferrous metal production
350 (Kyllönen et al., 2020; Wong et al., 2021). The temporal variability normalized time series (Fig. S10d) display a strong
351 seasonality, with elevated concentrations in winter and markedly lower in summer. This is typical for long-range transport of
352 air pollution into the Arctic from lower latitudes, favoured by a stable tropospheric stratification, stronger winds, and the
353 southward extension of the polar front in winter time (Platt et al., 2022; Stohl, 2006b).

354 Ni exhibited a distinct pattern, with an increase at the beginning of September followed by a gradual decline throughout the
355 month. This anomaly is reflected in a low correlation with other elements. Although Ni is often associated with ship emissions,
356 and typically highly correlated with V, the V/Ni ratio in September (0.3 ± 0.2 ; mean $\pm \sigma$) was far lower than the typical range
357 reported for ship emissions (2.5-4) (Becagli et al., 2020). In addition, the harbour traffic in Ny-Ålesund was relatively low in
358 September 2022 (27 vessels, 991 passengers) compared to August (103 vessels; 3975 passengers) (Statistics – Kings Bay —
359 Ny-Ålesund Harbour, 2025). Prevailing wind directions in September also do not support influence from the harbour to GAL
360 (Fig. S11). Although Ni-V-Cr enrichment has been reported in soils of south-west Greenland, this potential source does not
361 adequately explain the observed anomaly due to lack of coherent temporal variability among Ni, V, and Cr.

362 Concentrations of crustal elements (Figs. S10b and S10c) were generally higher in winter, when the surrounding terrain is
363 snow-covered, indicating substantial influence from long-range transport. However, two notable crustal events occurred in
364 summer, simultaneously with the increase of the concentration of sea salt elements. The first (17-23 August 2022) showed
365 elevated concentrations of Sr and Mo, along with spikes of Na, Mg, Ca, and K (Fig. S10a). A second event occurred on 21-27
366 September 2022, but without similar enrichment of Sr and Mo, highlighting the distinct signature of the first event. Both events
367 coincided with enhanced wind speed (Fig. S12), which likely promoted the resuspension of local soil and sea spray.

368 Higher concentrations of marine source elements in winter (Figure S10a) can be attributed to the higher burden of aerosol in
369 the atmosphere by a combination of enhanced long-range transport from lower latitudes and weaker atmospheric removal
370 processes (Spagnesi et al., 2025). Among the REE (Fig. S10c), Lu showed notable enrichment in early 2022 relative to other
371 lanthanides, consistent with its higher CEF (Fig. S8) and higher correlation with anthropogenic elements (Figure S7). This
372 enrichment may be related to refinery emissions at lower latitudes, where Lu serves as a catalyst in hydrocarbon processing
373 (Nayebare et al., 2018). A statistically significant small positive correlation is present between Lu and V (Spearman's ρ
374 correlation 0.48, p -value < 0.05 , $H_0 \rightarrow \rho = 0$), an element typically associated with the combustion of fuel oils (Arienzo et al.,
375 2021). The remaining REE follow concentration patterns previously reported for PM_{10} in the region (Conca et al., 2019). Ratios
376 between light and heavy REEs ($Pr/Yb = 0.9 \pm 0.5$ and $Pr/Er = 1.4 \pm 0.9$, mean $\pm \sigma$; normalized to upper crust continental values)

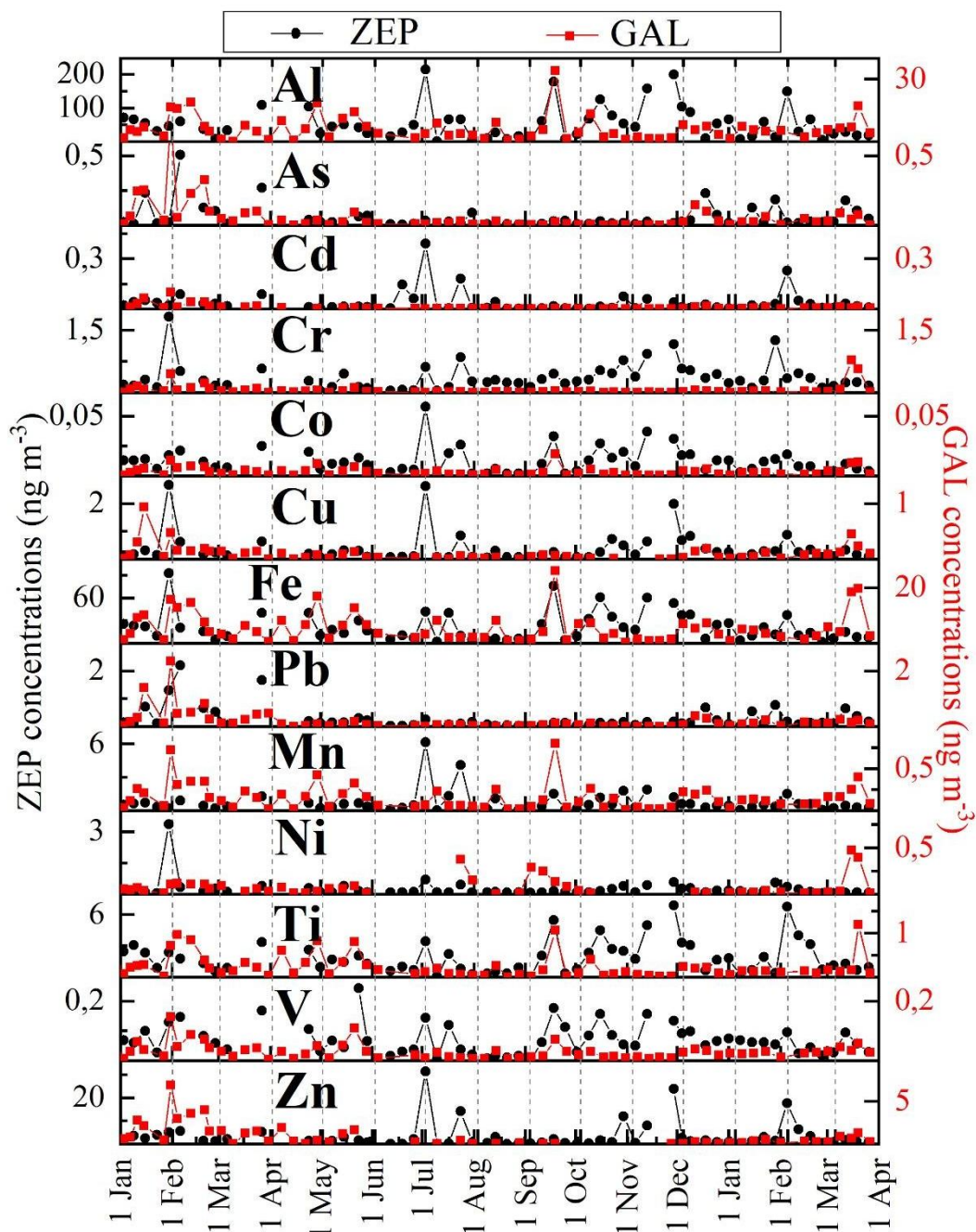


377 are close to unity, indicating little to no fractionation during transport. The Ce anomaly, averages 1.0 ± 0.2 (mean $\pm \sigma$), suggesting
378 no significant anthropogenic enrichment.

379 A comparison of selected elemental concentrations measured at GAL and at ZEP (Heavy metals at Zeppelin mountain (Ny-
380 Ålesund), 2025) shows generally higher values at ZEP, except from Pb and As, for which concentrations were similar at both
381 sites. The differences between the elemental concentrations in the time series of GAL and ZEP are statistically significant with
382 a Spearman's ρ correlation that is different from zero just for Pb (0.46, p-value < 0.05) and As (0.47, p-value < 0.05). Samples
383 from both observatories were digested using HNO_3 , ensuring methodological consistency and facilitating direct comparison.
384 However, the elemental data at ZEP refer to the fine fraction, whereas measurements at GAL were performed on PM_{10} .
385 Consequently, the observed differences between the two sites reflect not only vertical gradients and meteorological influences,
386 but also differences in the sampled size ranges. This implies that vertical contrasts in elemental concentrations may be even
387 more pronounced than suggested by Table 2. Notably, this effect is most evident for crustal elements (Al, Fe, Ti, and Mn) and
388 less so for anthropogenic species such as Pb and As.

389 Although both categories are predominantly influenced by long-range transport (Groot Zwaaftink et al., 2016), they exhibit
390 distinctly different vertical behaviour, as anthropogenic species are mainly associated with the accumulation-mode, whereas
391 crustal elements are mainly linked to coarser aerosol particles. This size-dependent vertical processing causes MD, even when
392 present in the fine fraction, to be preferentially depleted during downward mixing towards the surface through deposition,
393 whereas accumulation-mode particles remain well mixed. Consequently, air masses sampled at elevated sites such as ZEP are
394 less affected by near-surface removal processes than those measured at GAL. These results indicate that the observed
395 differences between the GAL and ZEP primarily reflect boundary-layer dynamics rather than differences in source origin,
396 underscoring the key role of ABL in shaping the vertical distribution of aerosol in the Arctic atmosphere.

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Figure 4. Comparison of the mass concentration in air for some elements between the samples collected at Gruebadet atmosphere laboratory (this work, red line and dots) and at Zeppelin observatory ((Heavy metals at Zeppelin Mountain (Ny-Ålesund), 2025), black line and dots) for the period January 2022 to March 2023. The concentrations are reported in a logarithmic scale.

402



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404

405 **Table 2.** Median and interquartile range for the season mass concentration for PM₁₀ samples collected from January 2022 to March 2023 at
406 Gruvebadet atmosphere laboratory and Zeppelin observatory (ng m⁻³).

		Winter	Spring	Summer	Autumn
Al	GAL	5.4 (3.6)	6.1 (8.0)	3.3 (3.0)	2.2 (2.9)
	ZEP	53.1(50.3)	31.4(22.4)	21.7 (44.6)	64.3(95.5)
As	GAL	0.05 (0.11)	0.03 (0.06)	0.01 (0.01)	0.003 (0.004)
	ZEP	0.03 (0.11)	0.04 (0.05)	0.007 (0.012)	0.02 (0.02)
Cd	GAL	0.01 (0.02)	0.007 (0.006)	0.001 (0.001)	0.0006 (0.0002)
	ZEP	0.03 (0.03)	0.012 (0.007)	0.01 (0.08)	0.005 (0.019)
Cr	GAL	0.04 (0.06)	0.04 (0.07)	0.02 (0.01)	0.01 (0.01)
	ZEP	0.31 (0.27)	0.18 (0.12)	0.24 (0.22)	0.41 (0.30)
Co	GAL	0.003 (0.004)	0.004 (0.003)	0.001 (0.001)	0.001 (0.002)
	ZEP	0.013 (0.007)	0.010(0.006)	0.004(0.00725)	0.014(0.021)
Cu	GAL	0.09 (0.12)	0.1 (0.1)	0.03 (0.01)	0.02 (0.04)
	ZEP	0.26 (0.20)	0.17 (0.20)	0.07 (0.08)	0.21 (0.46)
Fe	GAL	4.3 (5.1)	5.8 (5.5)	2.1 (2.3)	1.6 (3.0)
	ZEP	20.3 (15.5)	14.1 (12.0)	6.1 (5.6)	28.7 (38.9)
Pb	GAL	0.16 (0.28)	0.12 (0.15)	0.03 (0.03)	0.01 (0.04)
	ZEP	0.16 (0.54)	0.19 (0.18)	0.05 (0.05)	0.11 (0.10)
Mn	GAL	0.13 (0.15)	0.12 (0.13)	0.06 (0.05)	0.04 (0.09)
	ZEP	0.46 (0.30)	0.38 (0.34)	0.20 (1.05)	0.54 (1.02)
Ni	GAL	0.03 (0.05)	0.03 (0.05)	0.01 (0.14)	0.11 (0.17)
	ZEP	0.15 (0.19)	0.09 (0.11)	0.09 (0.02)	0.12 (0.19)
Ti	GAL	0.19 (0.14)	0.15 (0.25)	0.08 (0.13)	0.06 (0.09)
	ZEP	1.83 (2.345)	1.29 (0.81)	0.64 (0.58)	2.42 (3.14)
V	GAL	0.03 (0.02)	0.03 (0.02)	0.01 (0.01)	0.01 (0.01)
	ZEP	0.07 (0.04)	0.06 (0.06)	0.02 (0.02)	0.09 (0.09)
Zn	GAL	0.5 (1.8)	0.6 (1.0)	0.1 (0.2)	0.07 (0.04)
	ZEP	1.6 (2.3)	1.4 (1.1)	0.4 (2.2)	0.8 (3.2)

407

408 **3.5. Evaluation of natural and anthropogenic contributions using lead concentration, crustal enrichment, and isotopic**
409 **composition**

410 To better constrain the sources of PM₁₀, GAL samples were evaluated using Pb concentration, enrichment factors, and the
411 ²⁰⁷Pb/²⁰⁶Pb and ²⁰⁸Pb/²⁰⁶Pb isotope ratios (Table S1).



412 Lead concentrations (mean: 174 pg m⁻³; median: 80 pg m⁻³) agree well with values reported for PM₁₀ samples collected at
413 GAL from 2018 to 2020 (mean: 143 pg m⁻³; median: 55 pg m⁻³) (Grotti et al., 2024). Likewise, the crustal enrichment factors
414 (mean: 127; median 69) align with those previously reported (mean: 90; median 42) (Grotti et al., 2024), indicating a sustained
415 influence of anthropogenic sources on atmospheric Pb at GAL.

416 The ²⁰⁸Pb/²⁰⁶Pb and ²⁰⁷Pb/²⁰⁶Pb isotope ratios agree well with those reported for PM₁₀ samples collected at Ny-Ålesund during
417 2010 – 2020 (Bazzano et al., 2021; Grotti et al., 2024), and are similar to values observed at other High Arctic sites (Anon,
418 1989; Bory et al., 2014; Shotyky et al., 2005; Sturges et al., 1993).

419 Lead concentrations and CEFs exhibit the characteristic seasonal cycle for this region (Fig. S13), with higher values in samples
420 collected from December to April (Pb concentration: 290 ± 362 pg m⁻³; CEF: 198 ± 187, n=30) than in those collected from
421 May to November (Pb concentration: 50 ± 51 pg m⁻³; CEF: 52 ± 56, n=28). The difference between the two periods is
422 statistically significant (Mann–Whitney test, p < 0.001). In contrast, the Pb isotope composition does not differ significantly
423 between these two periods (Mann-Whitney test, p > 0.6): summer samples are characterized by isotope ratios (2.103 ± 0.012,
424 0.863 ± 0.008) that do not differ significantly from those in winter and spring (winter: 2.104 ± 0.007, 0.862 ± 0.002; spring:
425 2.105 ± 0.009, 0.861 ± 0.004). This feature was also observed in 2013 samples (Bazzano et al., 2016) and suggests a persistent
426 influence of the same dominant Pb source during both winter/spring and summer. The observed isotopic composition is
427 compatible with a Russian origin of atmospheric Pb at Ny-Ålesund (Ardini et al., 2020).

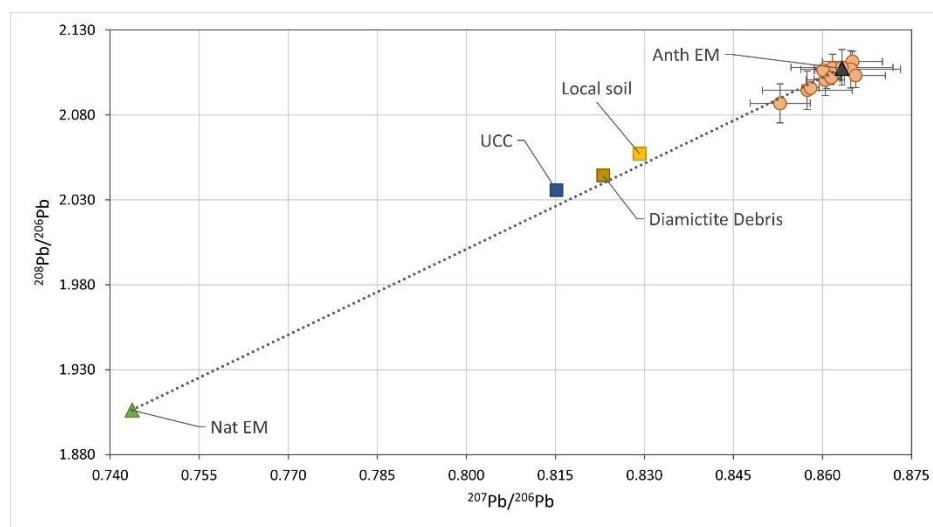
428 The relative natural and anthropogenic contributions to Pb the PM₁₀ samples were assessed by calculating the corresponding
429 end-members (EMs) following Grotti et al. (2024). Briefly, the isotopic data were grouped into six classes based on CEF (0–
430 20, 20–35, 35–50, 50–100, 100–150, 150). Each Pb isotope ratio was then modelled as a linear function of 1/CEF, and the
431 regression models were evaluated at CEF = 1 and CEF = ∞.

432 The estimated EMs (anthropogenic EM: ²⁰⁸Pb/²⁰⁶Pb = 2.108 ± 0.001, ²⁰⁷Pb/²⁰⁶Pb = 0.8633 ± 0.0002; natural EM: ²⁰⁸Pb/²⁰⁶Pb
433 = 1.906 ± 0.025, ²⁰⁷Pb/²⁰⁶Pb = 0.744 ± 0.007) are represented in a three-isotope plot along with the Pb isotopic compositions
434 of the PM₁₀ samples (monthly-averages) and some reference values from literature (Fig. 5). The mixing line between the
435 anthropogenic and natural end-members stretches towards the isotopic composition of the upper continental crust (UCC)
436 (Asmerom and Jacobsen, 1993) and passes through the area of Pb isotope ratio values of sediments collected in the
437 Kongsfjorden and at Ny-Ålesund (Bazzano et al., 2014). This finding suggests that the natural input to atmospheric Pb in this
438 area is most likely due to crustal material. However, given the Euclidean distances of the Pb isotope ratios of the samples from
439 the calculated EM signatures, the relative contribution of the natural origin appears to be negligible.

440 The ²⁰⁸Pb/²⁰⁶Pb and ²⁰⁷Pb/²⁰⁶Pb ratios of the anthropogenic EM are in good agreement with previous estimations (2.107 ± 0.002
441 and 0.864 ± 0.001) (Grotti et al., 2024) and compatible with the isotopic signature of Pb ores from the main mining sites for
442 Pb in East Kazakhstan and the Altai region (Zyranovsk: ²⁰⁸Pb/²⁰⁶Pb = 2.099, ²⁰⁷Pb/²⁰⁶Pb = 0.865 and Leninogorsk (now Ridder):
443 ²⁰⁸Pb/²⁰⁶Pb = 2.098, ²⁰⁷Pb/²⁰⁶Pb = 0.867) (Mukai et al., 2001). Since the majority of the Pb measured in the considered PM₁₀
444 samples is anthropogenic, this area can be identified as one of the main sources of this element at Ny-Ålesund. On the other



445 hand, emissions from Kola Peninsula smelters can be disregarded, as they appear to have low isotope ratios (Sturges and
 446 Barrie, 1989).
 447



448
 449 **Fig. 5.** Three-isotope plots $^{208}\text{Pb}/^{206}\text{Pb}$ vs $^{207}\text{Pb}/^{206}\text{Pb}$. Circles represent monthly averaged Pb isotope ratios in PM₁₀ samples collected at Ny-
 450 Ålesund from February 2022 to March 2023.

451 **3.6. Dust aerosol component**

452 Mineral dust (MD) concentrations were determined using four approaches, each based on the chemical composition of the
 453 collected aerosol, as described below:

- 454 1. Crustal oxides (Na₂O, MgO, SiO₂, Al₂O₃, TiO₂, K₂O, CaO, Fe₂O₃): MD concentration were estimated as the sum of
 455 the oxide-equivalent masses of major crustal elements (Na, Mg, Si, Al, Ti, K, Ca, and Fe). Elemental concentrations
 456 were converted to oxide weights using stoichiometric multipliers based on the crustal composition profile reported
 457 by Henderson and Henderson (2009).

458
$$\text{Mineral dust} = 1.35[\text{Na}] + 1.66[\text{Mg}] + 1.89[\text{Al}] + 2.14[\text{Si}] + 1.21[\text{K}] + 1.40[\text{Ca}] + 1.67[\text{Ti}] + 1.43[\text{Fe}]$$

- 459 2. Anthropogenically corrected oxide summation: This approach refines Method 1 by accounting for potential non-
 460 crustal (anthropogenic) contributions of K, Ca, and Fe. The crustal fractions of these elements are estimated from the
 461 measured aluminum (Al) concentration using the crustal element-to-Al ratios reported by Henderson and Henderson
 462 (2009).

463
$$\text{Dust} = 1.35[\text{Na}] + 1.66[\text{Mg}] + 3.79[\text{Al}] + 2.14[\text{Si}] + 1.67[\text{Ti}]$$

- 464 3. Sea-Salt and anthropogenically corrected oxide summation: This method extends Method 2 by additionally
 465 accounting for the sea salt contribution to sodium (Na) and magnesium (Mg). Non-sea-salt concentrations (nssNa
 466 and nssMg) were calculated as follows:

467
$$\text{Dust} = 1.35[\text{nssNa}] + 1.66[\text{nssMg}] + 1.89[\text{Al}] + 2.14[\text{Si}] + 1.21[\text{K}] + 1.40[\text{Ca}] + 1.67[\text{Ti}] + 1.43[\text{Fe}]$$



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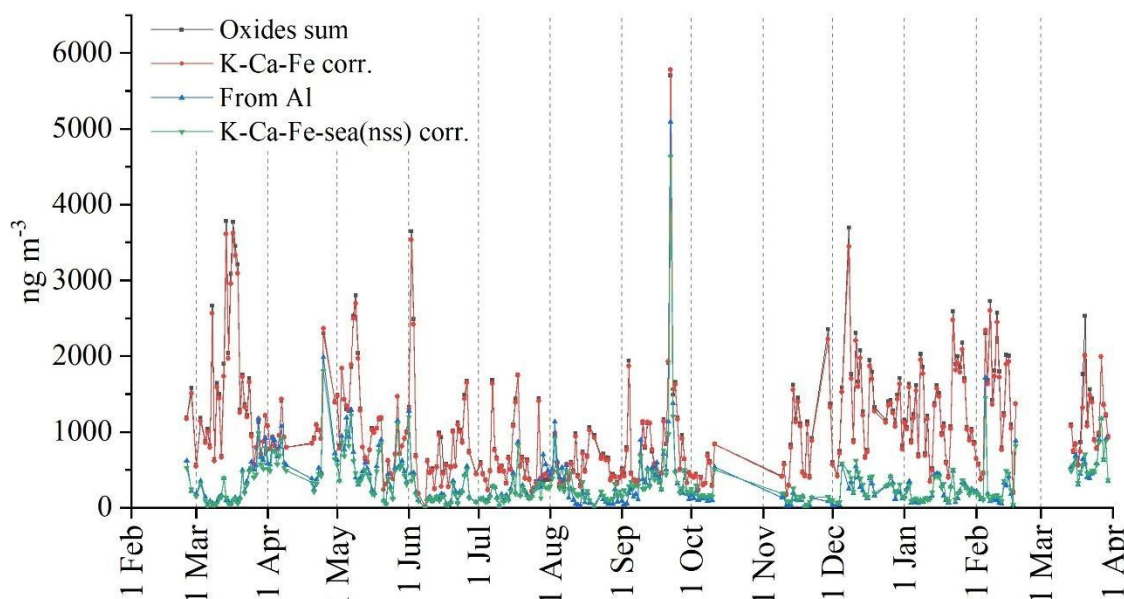
where

$$[\text{nssNa}] = 0.35[\text{Al}] \text{ and } [\text{nssMg}] = [\text{Mg}] - [\text{ssMg}] = [\text{Mg}] - 0.12[\text{ssNa}] = [\text{Mg}] - 0.12([\text{Na}] - [\text{nssNa}])$$

4. Aluminum-Based "Rule of Thumb": This simplified approach estimates mineral dust from the measured aluminium concentration, using aluminium's average mass fraction in the upper continental crust to provide a rapid proxy for dust concentration.

A comparison of the results from the four approaches is presented in Figure 6. The first two methods show a notable agreement, which suggests that the yearly average anthropogenic contributions of elements like K, Ca, and Fe are negligible compared to the dust's crustal contribution. However, these two methods consistently overestimate dust concentration relative to the other two approaches, which also show a reasonable agreement with each other.

Method 3, which corrects for both sea-salt and anthropogenic contributions, was ultimately selected as the most reliable approach for this study. Given the sampling site's location in Ny-Ålesund, a coastal environment within a fjord, the contribution of sea spray to Na and Mg concentrations is expected to be significant. Therefore, explicitly accounting for these sea-salt contributions provides a more accurate representation of the true crustal dust component. This exercise highlights the importance of selecting a methodology that is appropriate for the specific environmental context of the sampling location.



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486
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Fig. 6. Dust concentration as obtained following the approaches described in paragraph 3.5; i.e. as an oxides sum (appr. 1), K-Ca-Fe corr. (appr. 2), K-Ca-Fe sea(nss) corr. (appr. 3), or finally form the sole Al assessment following the approach 4.



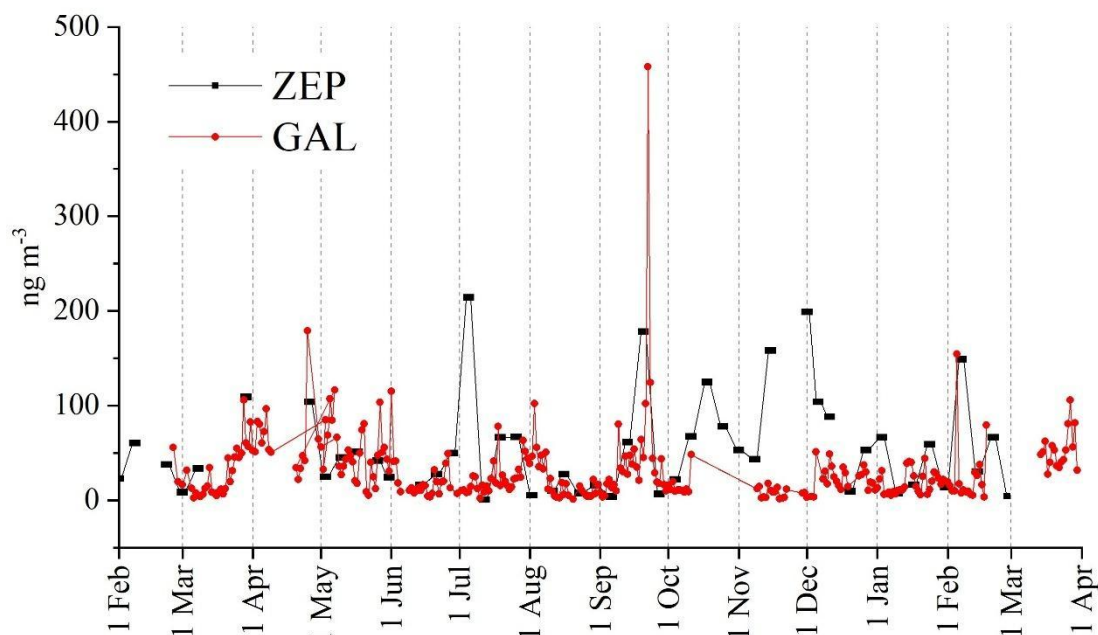
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489 Analysis of the CEFs for crustal elements showed that the CEF of Silicon remained consistently close to 1 throughout the year.
490 This suggests a stable silicon-to-aluminium ratio in the dust source. A notable exception occurred in August 2022, when
491 elevated CEFs were observed for multiple elements. Back-trajectory analysis for this period shows air masses traveling along
492 an Iceland-Greenland-Canada pathway where potential dust sources might be located (see Figure S17, right panel); however,
493 the back-trajectory analysis cannot rule out local sources.

494 Aluminium concentration has proven to be a reliable proxy for aerosol dust in Ny-Ålesund. Comparing Al concentration data
495 from two different sites (GAL and ZEP) can help differentiate between dust transport processes occurring above the Arctic
496 Boundary Layer and local dust resuspension events.

497 Figure 7 shows the Al concentration measured at both GAL and ZEP, with GAL data providing a higher temporal resolution.
498 A significant difference in the temporal trends was observed at the beginning of July 2022. During this time, Al concentrations
499 at ZEP were considerably higher than at GAL. This discrepancy suggests a dust transport event at a higher altitude, with the
500 elevated concentrations observed at the ZEP station not reaching the lower-lying GAL station.

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Fig. 7. Aluminium concentrations measured at both GVB and ZEP

505 The monthly average aerosol mass (Fig. S15), measured gravimetrically on filters before PIXE analysis, was compared with
506 a reconstructed mass based on PIXE and Thermo-Optical Analysis data. The difference between these two values, defined as



507 "unexplained mass", is primarily attributed to ionic components such as nitrates, which were not directly measured. While
508 sulfates were estimated based on the measured sulfur (S) concentration, other ionic species were not accounted for.
509 A clear seasonal pattern was observed in the PM loadings (Fig. S15). Concentrations were lower during the summer and
510 autumn months and higher during winter and spring. Similarly, dust contributions were at their minimum during the summer.
511

512 **3.7. Carbonaceous components**

513 Organic carbon (OC), elemental carbon (EC), and total carbon (TC) have been measured at GAL since 2011 (Caiazzo et al.,
514 2021) and at ZEP since 2017 (Yttri et al., 2021). The main differences compared to the ZEP protocol concern the sampling
515 duration (see Table 1) and the thermal protocol used for the thermo-optical analysis; at GAL, analyses have been performed
516 using the NIOSH-870 protocol).

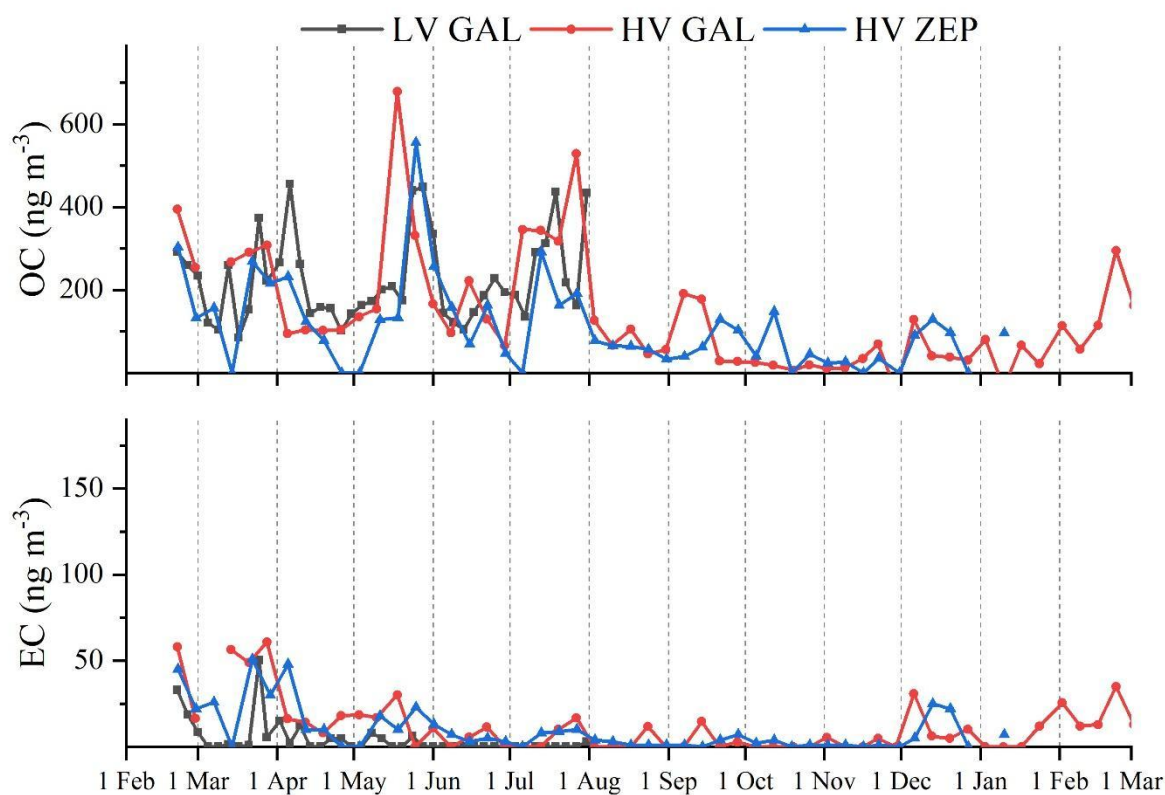
517 Within the framework of this project, sampling and analytical procedures at GAL and ZEP were harmonized for a one-year
518 period (Table 1). During this period, OC and EC were analyzed on PM₁₀ filter samples using the EUSAAR_2 thermal protocol
519 (Cavalli et al., 2010).

520 For a shorter overlapping period, GAL data obtained according to the procedure reported by Caiazzo et al. (2021) were
521 compared with GAL data generated using the standardized procedure to assess the comparability of the two data series (with
522 the purpose to avoid abrupt changes in the GAL data series). The results show a reasonable agreement between the two datasets
523 (Fig. 8), having different time resolution (4 and 7 days respectively), with monthly OC averages differing by approximately
524 10% (range: 4-17%). The comparison of EC data is more uncertain: besides the difficulty of comparing data for samples
525 collected with different time resolution, concentrations are frequently below the detection limit. These findings suggest that
526 the two thermal protocols (EUSAAR_2 and NIOSH-870) may produce comparable results for Arctic aerosol, although further
527 work focused on thermal protocols is needed..

528 GAL data for the observation period fall within the range previously observed (Caiazzo et al., 2021), although some lower
529 values were observed. Notably, these measurements are among the first obtained while the GAL observatory operated during
530 the Polar Night (October-February). Lower OC concentrations during winter may reflect reduced biogenic activity, more
531 extensive sea-ice coverage, decreased long-range transport from anthropogenic source regions, and a lower frequency of fire
532 events.

533 Comparing GAL and ZEP, the two observatories exhibit similar temporal behaviour for both OC and EC, although
534 concentrations are slightly lower at ZEP. This suggests that both sites are influenced by long-range transport of carbonaceous
535 aerosol, while GAL appears to possibly experience an additional contribution from local or regional sources, as further
536 suggested by the results shown in Fig.11 (see later).

537



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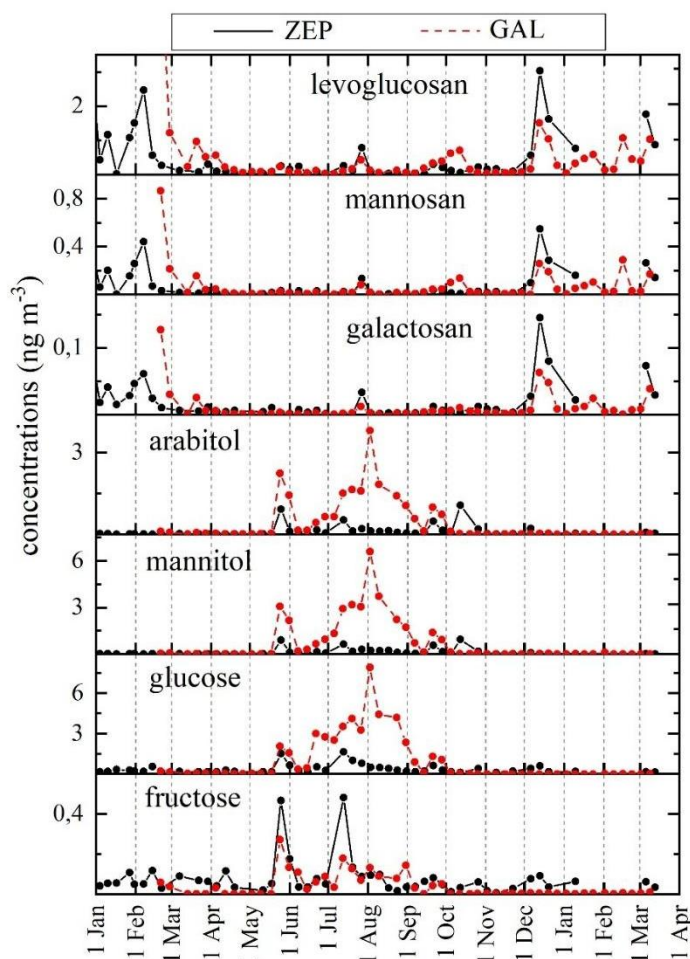
539 **Fig. 8.** OC (upper panel) and EC (lower panel) concentrations measured at both ZEP and GAL. ZEP results are obtained from
540 a HV sampler (7 days, EUSAAR2), while for GAL results as obtained from the HV sampler (7 days, EUSAAR2) harmonized
541 with ZEP and from the LV sampler (4 day-resolution, NIOSH-870) used in the past.

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543

544 **3.8. Biomass burning and biogenic organic tracers**

545 Figure 9 shows concentrations of the biomass-burning tracers levoglucosan, mannosan, and galactosan (Simoneit, 1999), the
546 fungal spore tracers arabitol and mannitol (Bauer et al., 2008), and the more generalized PBAP tracers glucose and fructose
547 measured at GAL and ZEP during BETHA-NyÅ. To verify laboratory performance comparability, four samples collected in
548 spring and summer at ZEP were additionally analyzed at the Venice Lab. No statistically significant differences were found
549 for any of the species between the Venice and NILU analyses (Wilcoxon test, $p > 0.05$; Fig. S16).



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Fig. 9. Levels of biomass burning tracers (levoglucosan, mannosan, and galactosan) and PBAP markers (arabitol, mannitol, glucose, and fructose) at GAL and ZEP from Jan 2022 to Apr 2023.

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The different tracers exhibit distinct behaviors. Arabitol and mannitol show pronounced summer maxima at GAL, whereas no comparable enhancement is observed at ZEP. These compounds are primarily associated with the coarse aerosol fraction ($> 1 \mu\text{m}$) (Feltracco et al., 2020) and indicate a local input from fungal spores near ground level. Due to their relatively large particle size, these particles are less efficiently transported upward within the mixing layer, which may explain the lower levels observed at the higher-altitude ZEP observatory.

559

560

Glucose is a less source-specific tracer, as it may originate from both biomass burning and PBAP sources (Feltracco et al., 2020). In this case, however, glucose follows the same pattern as the fungal spore tracers (Fig. 9), indicating a predominantly



561 biogenic origin. In contrast, fructose exhibits a different temporal behavior, although some peaks coincide with those of
562 glucose.

563 The observed levoglucosan concentrations are largely driven by long-range transport of emissions from residential wood
564 combustion (RWC) and wildfires, although contributions from local sources cannot be excluded. The isolated peak observed
565 at GAL at the beginning of the BETHA-NyÅ campaign is likely attributable to a local source. In contrast, the recurrent peaks
566 observed at ZEP during the heating season (Nov-Apr) are consistent with RWC, as previously demonstrated by Yttri et al.
567 (2024).

568 Not all levoglucosan maxima correspond to documented wildfires or RWC activity. Nevertheless, the relatively high
569 concentrations observed during the second half of March and the first half of April may be linked to events in central and
570 eastern Europe as well as in central Asia (see Fig. S17). Similarly, the slight increase in concentrations in late July and early
571 August may be associated with events in Europe, northern Asia, and North America (see Fig. S18). Comparable interpretations
572 apply to the maximum in early October 2022 (left panel of Fig. S19) and to one of the highest peaks detected in mid-December.
573 Elevated concentrations measured in early 2023 were likely influenced by fires primarily located in central and eastern Europe
574 (right panel of Fig. S19).

575 Owing to the relatively long sampling intervals and rapidly changing circulation patterns, the fire source influencing the
576 observed concentrations often extend over large geographical areas.

577 The isomeric ratio of levoglucosan to mannosan, proposed as a diagnostic indicator of the type of burned vegetation (Fabbri
578 et al., 2009), does not differ significantly between GAL (8 ± 4) and ZEP (8 ± 3). Although Yttri et al. (2024) reported differences
579 between the heating (Nov-May) and non-heating (Jun-Oct) periods at ZEP for 2017-2020, no such difference is observed for
580 the period considered here: the levoglucosan/mannosan ratios are 8 ± 4 (GAL) and 7 ± 1 (ZEP) during the heating season (7.5 ± 1.9
581 for Yttri et al. (2024)), and 8 ± 4 (GAL) and 8 ± 5 (ZEP) during the non-heating season (4.8 ± 1.2 for Yttri et al. (2024)). Given
582 that levoglucosan/mannosan ratios are typically higher for hardwood combustion (14–15) and lower (3–5) for softwood
583 combustion (Schmidl et al., 2008), the values observed at both sites suggest a mixture of hardwood and softwood combustion,
584 consistent with previous findings from the region (Feltracco et al., 2020).

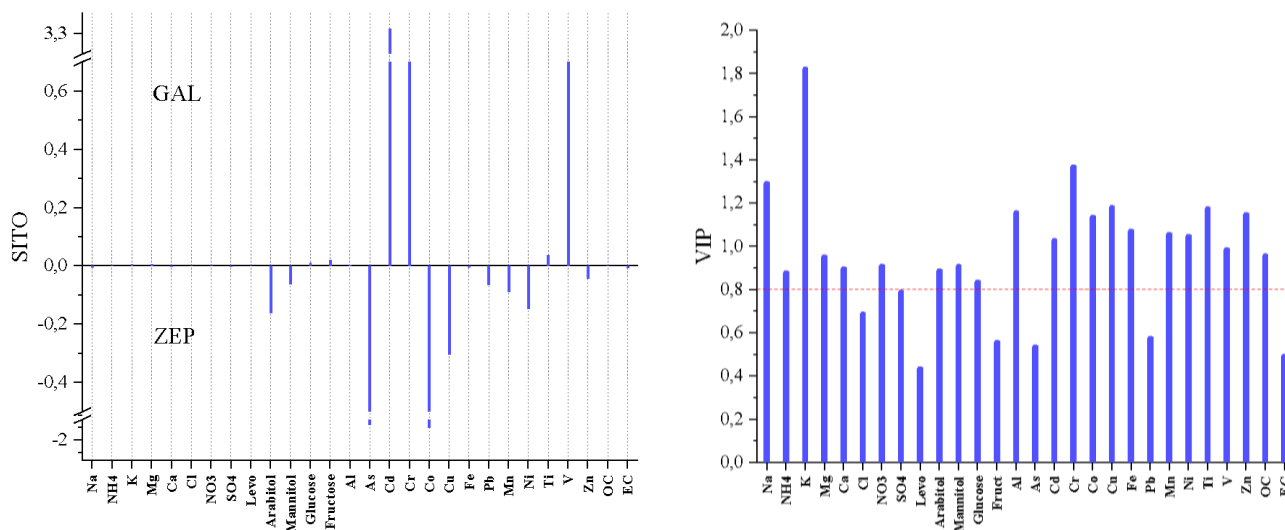
585

586 **3.9. Analysis of difference between GAL and ZEP and its main drivers**

587 We used multivariate statistical analysis to quantify the compound-specific differences between GAL and ZEP, highlighted in
588 the preceding sections. Preliminary analysis with the Shapiro-Wilk test demonstrated that the data were not normally
589 distributed and for this reason Linear Discriminant Analysis (LDA) was excluded as a chemometric approach. Instead, the
590 Kruskal-Wallis test confirmed significant site-related differences in the measured chemical variables, whereas Partial Least
591 Squares Discriminant Analysis (PLS-DA) was used to identify the species most responsible for discriminating between the
592 two sites.



593 The input matrix consisted of 30 chemical species and 106 observations, corresponding to 53 weekly samples simultaneously
 594 collected at GAL and ZEP. A large fraction of the variance was explained by the first two latent factors (Fig. S20 and Table
 595 S2), accounting for 38% and 61% of the cumulative variance in the predictor (X) and response (Y, sites classification). The
 596 high proportion of explained Y variance indicates that the chemical differences between two sites are statistically significant.
 597 The PLS-DA coefficient plot displays the estimated regression coefficients for each variable, providing insight into both the
 598 direction and magnitude of their contribution to site discrimination (Fig. 10A). The anthropogenic species Cd, Cr, and V are
 599 the most influential variables associated with GAL. In contrast, As, Co, Cu, Pb, Mn, and Ni emerge as the most relevant
 600 variables for ZEP, together with arabitol and mannitol.
 601 The Variable Importance in Projection (VIP) plot (Fig. 10B), interpreted in conjunction with the coefficient plot, summarizes
 602 the overall contribution of each variable to the PLS-DA model. While the coefficient plot highlights the direction and
 603 magnitude of individual effects, the VIP plot ranks variables according to their global relevance, thereby facilitating
 604 interpretation. The VIP analysis indicates that ionic species play a substantial role in differentiating the two sites, with Na and
 605 K showing particularly high importance.



606
 607 **Fig. 10.** A) Coefficients plot and B) Variable Importance in Projection (VIP) plot obtained with PLS-DA, explained which
 608 compound impacts on the difference of each site.
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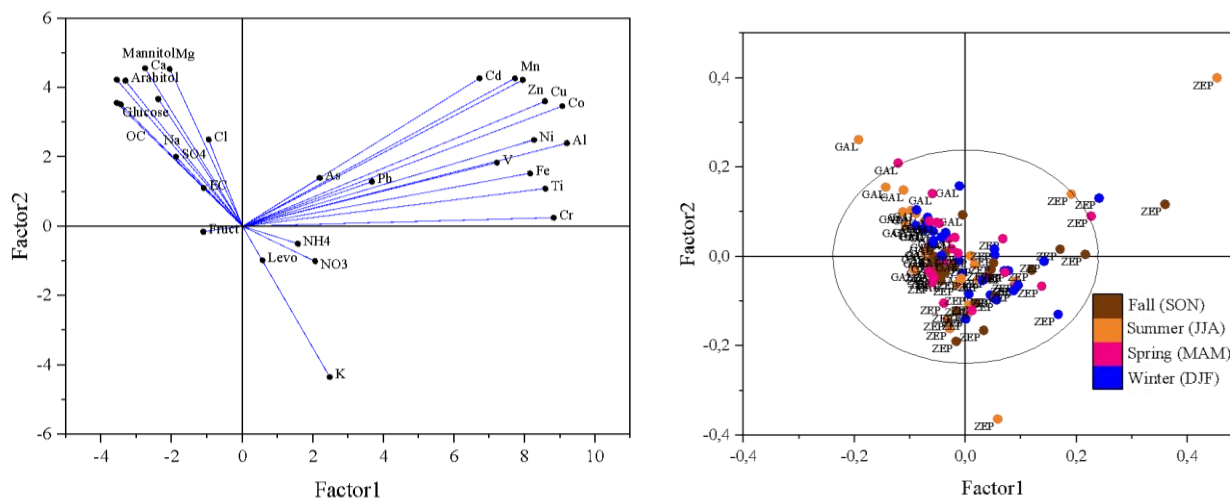


Fig. 11. A) Factor loading and B) scores loading obtained with PLS-DA.

The loading plot (Fig. 11A) illustrates how variables contribute to the latent components and identifies the chemical species most responsible for class separation. Factor 1 is associated with trace metals of predominantly anthropogenic (e.g., Cd, Co, Cu, Pb, As) and crustal origin. Factor 2 is primarily driven by markers of PBAP, such as arabinol, mannitol, and glucose, together with sea salt aerosol (Na^+ and Cl^-), combustion-related aerosol (e.g., EC) and the ubiquitous OC. The clustering of levoglucosan, K^+ , NH_4^+ , and NO_3^- reflects a mixed influence of biomass-burning emissions and secondary inorganic aerosol (NH_4^+ and NO_3^-) formation.

The score plot (Fig. 11B) shows the distribution of samples across seasons. In general, samples cluster close to the origin, indicating substantial overlap between sites and season and suggesting broadly similar chemical composition. Nevertheless, several samples deviate from the main cluster, pointing to episodic source influences. Some GAL samples collected during summer and spring show positive shifts along Factor 2, consistent with enhanced contributions from biogenic sources during the growing season. Conversely, isolated ZEP samples are shifted along Factor 1, likely reflecting episodes of long-range transport of anthropogenic aerosols.

Overall, the PLS-DA results indicate that, despite substantial overlap in chemical composition, seasonal variability is driven by a limited set of source-related variables. Biogenic species dominate the summer variability, whereas anthropogenic species contribute more consistently throughout the year at both sites. This indicates that PLS-DA captures subtle, yet systematic, differences in source influences across sites and seasons.

The Arctic boundary layer (ABL) is typically shallow (<200 m), stably stratified, and often capped by low clouds, although its depth varies strongly with season and surface type. Over continental regions, ABL deepens during summer (up to > 1.5 km), whereas over oceanic areas it is thickest in winter and spring due to strong air–sea temperature contrasts (Esau and



634 Sorokina, 2009). During summer, GAL and ZEP are therefore expected to experience similar air masses with broadly similar
635 chemical composition; however, local near-surface sources can still introduce differences between the two sites. In particular,
636 arabitol and mannitol, tracers of fungal spores, are emitted close to ground level and are mainly associated with coarse particles
637 (Feltracco et al., 2020). As ZEP is located well above these sources, such species are less likely to reach its altitude, explaining
638 the lower concentrations observed at ZEP.

639 During winter, stronger stratification and a shallower ABL enhance vertical gradients. This is clearly reflected in the score plot
640 (Fig. 11B), where winter samples (blue symbols) from GAL and ZEP are distinctly separated from one another along Factor
641 1. While ZEP shows a stronger influence of biomass burning species, consistent with its greater sensitivity to the long-range
642 transport of polluted air masses, GAL appears more influenced by SSA. This signature in GAL could be linked to an increased
643 frequency of regional cyclonic systems (Huang and Jaeglé, 2017) or, potentially, to local emissions originating from the fjord.

644 **4 Conclusions**

645 The BETHA-NyÅ project successfully demonstrated the feasibility and scientific value of coordinated aerosol measurements
646 at different altitudes in Ny-Ålesund. The coordinated effort between the GAL and ZEP observatories, combined with
647 meteorological observations from the Amundsen-Nobile Climate Change Tower, enabled a characterization of Ny-Ålesund
648 vertical distribution on aerosol composition.

649 The study achieved a high degree of seasonal coherence between the two sites, particularly for anthropogenic markers
650 associated with the Arctic Haze phenomenon. Sulphate and ammonium concentrations exhibited nearly identical temporal
651 trends, with sulphate levels peaking in March-April at both locations. Quantitatively, median sulphate concentrations were
652 remarkably consistent, recorded at 203 ng m⁻³ at GAL and 220 ng m⁻³ at Zeppelin. Optical properties also showed alignment
653 during the cold season (November to April), with median scattering coefficients of 5.5 Mm⁻¹ at ZEP and 4.2 Mm⁻¹ at GAL.
654 These results confirm that long-range transport of polluted air masses often overcomes local vertical gradients, resulting in a
655 well-mixed lower atmosphere during haze episodes.

656 The harmonization protocol proved robust, revealing that while long-range signals are uniform, local differences emerge due
657 to near-surface sources and boundary layer dynamics. The most significant site-specific variations were observed for biogenic
658 tracers like arabitol and mannitol, which showed higher concentrations at GAL. This suggests that primary biological aerosol
659 particles (PBAPs) are predominantly emitted from local terrestrial or marine sources near sea level and are trapped within a
660 shallow, stable ABL, often failing to reach the elevation of the Zeppelin observatory.

661 Our findings on lead (Pb) concentrations (mean: 174 pg m⁻³) at GAL align with previous long-term monitoring efforts (2010–
662 2020), indicating a stable and persistent anthropogenic influence. Lead isotopic ratios (²⁰⁸Pb/²⁰⁶Pb ~2.10 and ²⁰⁷Pb/²⁰⁶Pb ~0.86)
663 remain consistent with Eurasian and Russian sources, confirming that the transport pathways for heavy metals into the Svalbard
664 region have not significantly shifted in the last decade.



665 These results have direct implications for our understanding of Arctic Amplification. The persistent presence of Eurasian
666 anthropogenic aerosols and the stratification of biogenic particles influence the region's radiative balance and cloud formation
667 processes. Specifically, the trapping of aerosols within a shallow ABL can enhance local warming through surface-atmosphere
668 interactions. This study reinforces the necessity of integrated, multi-altitude monitoring networks to accurately model the
669 deposition of pollutants on snow and ice, a critical driver of the ice-albedo feedback mechanism in a rapidly changing Arctic
670 environment.

671 **Data availability**

672 Data about GAL measurements are available at IADC (DOI to be assigned). All data from ZEP are open access and are
673 available at <http://ebas.nilu.no/>. Different ZEP measurements are reported in different database
674 (<https://doi.org/10.48597/R38F-6XPS>; <https://doi.org/10.48597/ECGE-V26Y>; <https://doi.org/10.48597/6PXH-3ESD>).
675

676 **Competing interests**

677 At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

678 **Author contributions**

679 EB: Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Supervision,
680 Writing (original draft preparation); FA: Data curation, Investigation, Methodology, Writing (original draft preparation); SB:
681 Conceptualization, Data curation, Investigation, Methodology, Writing (original draft preparation); SB: Data curation,
682 Investigation, Methodology, Writing (original draft preparation); FB: Data curation, Investigation, Writing (review and
683 editing); GC: Conceptualization, Data curation, Investigation, Methodology, Writing (original draft preparation); AC: Data
684 curation, Writing (review and editing); DC: Conceptualization, Data curation, Investigation, Methodology, Writing (original
685 draft preparation); SC: Data curation, Investigation, Methodology, Writing (original draft preparation); MF: Data curation,
686 Investigation, Methodology, Writing (original draft preparation); AG: Writing (review and editing), Supervision; FG: Data
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688 Methodology, Writing (original draft preparation); MG: Data curation, Investigation, Methodology, Writing (review and
689 editing); DHR: Investigation, Methodology, Writing (review and editing); RK: Data curation, Investigation, Methodology,
690 Writing (review and editing); MM: Data curation, Investigation, Methodology, Writing (original draft preparation),
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692 Investigation, Writing (review and editing); MP: Investigation, Writing (review and editing); MR: Investigation, Writing



693 (review and editing); CR: Data curation, Writing (review and editing); MS: Data curation, Investigation, Methodology, Writing
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700

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720



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