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Pan-Arctic seasonal cycles and long-term trends of aerosol properties from ten observatories

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Abstract.

Even though the Arctic is remote, aerosol properties observed there are strongly influenced by
anthropogenic emissions from outside the Arctic. This is particularly true for the so-called Arctic haze
season (January through April). In summer (June through September), when atmospheric transport
patterns change, and precipitation is more frequent, local Arctic, i.e. natural sources of aerosols and
precursors, play an important role. Over the last decades, significant reductions in anthropogenic
emissions have taken place. At the same time a large body of literature shows evidence that the Arctic is
undergoing fundamental environmental changes due to climate forcing, leading to enhanced emissions
by natural processes that may impact aerosol properties.

In this study, we analyze nine aerosol chemical species and four particle optical properties from ten
Arctic observatories (Alert, Gruvebadet, Kevo, Pallas, Summit, Thule, Tiksi, Barrow, Villum, Zeppelin)
to understand changes in anthropogenic and natural aerosol contributions. Variables include equivalent
black carbon, particulate sulfate, nitrate, ammonium, methanesulfonic acid, sodium, iron, calcium and
potassium, as well as scattering and absorption coefficients, single scattering albedo and scattering
Ångström exponent.

First, annual cycles are investigated, which despite anthropogenic emission reductions still show the
Arctic haze phenomenon. Second, long-term trends are studied using the Mann-Kendall Theil-Sen slope
method. We find in total 28 significant trends over full station records, i.e. spanning more than a
decade, compared to 17 significant decadal trends. The majority of significantly declining trends is from
anthropogenic tracers and occurred during the haze period, driven by emission changes between 1990
and 2000. For the summer period, no uniform picture of trends has emerged. Twenty-one percent of
trends, i.e. eleven out of 57, are significant, and of those five are positive and six are negative. Negative
trends include not only anthropogenic tracers such as equivalent black carbon at Kevo, but also natural
indicators such as methanesulfonic acid and non-sea salt calcium at Alert. Positive trends are observed
for sulfate at Zeppelin and Gruvebadet.

No clear evidence of a significant change in the natural aerosol contribution can be observed yet.
However, testing the sensitivity of the Mann-Kendall Theil-Sen method, we find that monotonic
changes of around 5 % per year in an aerosol property are needed to detect a significant trend within
one decade. This highlights that long-term efforts well beyond a decade are needed to capture smaller
changes. It is particularly important to understand the ongoing natural changes in the Arctic, where
interannual variability can be high, such as with forest fire emissions and their influence on the aerosol
population.

To investigate the climate-change induced influence on the aerosol population and the resulting climate
feedback, long-term observations of tracers more specific to natural sources are needed, as well as of
particle microphysical properties such as size distributions, which can be used to identify changes in
particle populations which are not well captured by mass-oriented methods such as bulk chemical
composition.

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1 Introduction

Despite its remoteness, the Arctic is a main receptor of anthropogenic air pollutant emissions from the northern hemisphere (AMAP, 2015, p.2015; Barrie, 1986; Quinn et al., 2007; Shaw, 1995; Heidam, 1981). Arctic air pollution has been in focus for several decades, because of the detrimental effects on ecosystems, for example through acidification (AMAP, 2006) and deposition of pollutants that bio-accumulate especially in the marine food web (Rigét et al., 2019), and due to climate impacts (Sand et al., 2015). The Arctic Monitoring and Assessment Programme (AMAP) has issued a number of reports on the effects of short-lived climate forcers (AMAP, 2011, 2015), and found in the 2015 report that the Arctic equilibrium temperature response is in total +0.35 °C due to forcing from black carbon (BC) in the atmosphere (+ 0.40 °C), BC on snow (+0.22 °C), atmospheric organic carbon (-0.04 °C) and particulate sulfate (-0.23 °C).

Surface monitoring sites around the Arctic (Fig. 1) have been operated to determine air pollutants transported to the Arctic since as early as the 1960s at Kevo, Finland (Dutkiewicz et al., 2014), the 1970s at Barrow, USA (Bodhaine and Dutton, 1993), the 1980s at Alert, Canada (Sturges and Barrie, 1989), the 1990s at Zeppelin, Svalbard (Platt et al., 2021) and Villum Research Station, Greenland (Nguyen et al., 2016; Heidam et al., 1999), and more recently at a number of other observatories such as Summit, Greenland (Schmeisser et al., 2018), Tiksi, Russia (Asmi et al., 2016), Gruvebadet, Svalbard (Gilardoni, 2019; Traversi et al., 2021), Thule, Greenland (Becagli et al., 2019) and at Pallas, Finland (Aaltonen et al., 2006). Observations at these stations cover a wide range of aerosol properties including their chemical composition, i.e. particulate sulfate (SO_4^{-2}), nitrate (NO_3^-), ammonium (NH_4^+), mainly based on filter collection and subsequent laboratory analysis, and optical properties such as scattering and absorption coefficients and derived equivalent black carbon (EBC) concentrations. Using the Eclipse V4.0a emission inventory and state of the art models, AMAP et al. (2015) found that emission sources in East and South Asia and Russia are the largest contributors to black carbon burden in the Arctic, with particularly large contributions of domestic sources in East and South Asia, and emissions from wildfires and gas flaring in Russia. Depending on the location of the observations, different source regions become relevant (Backman et al., 2021). Based on Lagrangian transport model simulations, Hirdman et al. (2010b) showed

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110   for Zeppelin, Barrow and Alert that northern Eurasia is the main source region, whereas emissions from

North America are less influential. The Summit observatory located above 3000 m a.s.l. is more

susceptible to emissions of Southern Europe (Hirdman et al., 2010b), since those are uplifted and arrive

at the Arctic further aloft. From a near sea level surface observatory perspective, emissions in Asia and

particularly southern Asia are less important because they are transported through the higher troposphere

115   and do not significantly influence boundary layer measurements (AMAP, 2015; Shindell et al., 2008).

Seasonality of BC can be very different depending on the altitude. Mahmood et al. (2016) showed that,

for example, BC in the upper troposphere is only weakly influenced by wet deposition in stratiform

clouds, whereas lower tropospheric Arctic BC concentrations are highly sensitive to wet deposition.

The surface site observations have created a large data resource (for a history on monitoring sites see,

120   Platt et al., 2021), which has been analyzed for seasonal cycles. The seasonal cycle of aerosols in the

Arctic is generally driven by atmospheric transport patterns, which, during winter and early spring, favor

long-range transport from mid-latitudes and removal processes are minimal allowing for long aerosol

lifetimes and accumulation of pollutants (e.g., Stohl, 2006). This leads to the build-up of air pollution, the

so-called Arctic haze (Heidam, 1981; Barrie, 1986; Schnell, 1984; Radke et al., 1976). During summer,

125   the Arctic front is located far to the north and the Arctic dome largely prevents transport of particles from

sources at mid-latitude from reaching the high Arctic. The summertime Arctic atmosphere is much less

stratified and frequent precipitation removes aerosols efficiently, while simultaneously, conditions for

long-range transport of pollution are also less frequent. Hence, the summer boundary layer is

characterized by much lower aerosol concentrations, predominantly reflecting local and regional

130   emissions. Overall, the seasonality of aerosol properties at Arctic sites depends on the geographical

sectors, and on how the specific sites are connected to upstream pollution sources through atmospheric

transport. Sharma et al. (2019) analyzed annual cycles of 20 aerosol constituents at Alert. In brief, for all

anthropogenic tracers they find a winter peak and a summer minimum, as expected. For some natural

tracers, such as iodine, methanesulfonic acid (MSA) and mineral dust, they find two modes, one in spring

135   and the other in summer/fall. For mineral dust the spring peak represents long-range transported dust,

while the summer/fall peak reflects more local emissions when the snow cover is at a minimum. Similarly,

for MSA, an oxidation product from dimethyl sulfide (DMS), which is derived from marine microbial

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activity, the spring maximum reflects long-range transport, while the summer maximum is due to more local emissions. The reasons for the iodine pattern are not entirely clear but they are related likely to the

140   return of the sun in spring and freeze-up processes in fall (Baccarini et al., 2020). For other natural tracers, such as sodium (Na^+) and chloride (Cl^-), which mainly originate from sea salt, they find a winter maximum, which is driven by sublimating blowing snow leaving behind salt particles (Huang and Jaeglé, 2017; Fenger et al., 2013). Quinn et al. (2007) reported annual cycles of anthropogenic haze tracers for Barrow, Alert, Villum, Zeppelin and several other Eurasian stations. Eckhardt et al. (2015) compiled

145   annual BC and SO_4^{2-} data from the same four stations, as well as Tiksi and Pallas in addition for a model evaluation. Backman et al. (2017) systematically evaluated aethalometer measurements of black carbon from Barrow, Alert, Zeppelin, Summit, Pallas and Tiski, and Schmeisser et al. (2018) used the same data for a systematic analysis of seasonal cycles of aerosol optical parameters. While the seasonal patterns of the aerosol scattering and absorption coefficients follow the pattern of anthropogenic tracers in the Arctic

150   for four of the sites, Summit and Pallas show unique signals, where the maximum is in summer rather than winter. They attribute this difference to the higher load of natural aerosol emissions from the boreal forest around Pallas (Tunved et al., 2006, 2003) and the long-range transport of fire emissions to Summit during summer (Stohl et al., 2006).

The same or extended database used for the study of annual cycles has been used in a number of studies

155   for long-term trend analyses. The longest time series of black carbon measurements was collected at Kevo, Finland, where annual concentrations decreased by 73% between about 1970 and 2010 (Dutkiewicz et al., 2014). Sharma et al. (2006) and Quinn et al. (2007) found a 33 % decline between 1989 and 2003 for Barrow during the haze season. The analysis was updated by Sharma et al. (2013), who found a roughly 40 % decrease for Barrow, Alert and Zeppelin between 1990 and 2009. More recent work by

160   Sharma et al. (2017, 2019) extended the analysis until 2013, reporting a roughly 50 % decrease of black carbon since 1990 during the haze season. The authors ascribe the decline firstly to the economic changes in the former Soviet Union and secondly to air quality policies in Europe. Observations for SO_4^{2-} are analogous for Alert, also with a 50 % decline over the same time period (Sharma et al., 2019). Hirdman et al. (2010a) derived trends based on annual geometric mean data and found a 21.5 % decline at Zeppelin

165 between 1990 and 2008 in non-sea salt sulfate ($nssSO_4^{2-}$) and no trend for Barrow between 1997 and 2006. The absence of a trend at the latter station is potentially because of the short time period investigated. The results for Zeppelin are somewhat in contrast to Platt et al. (2021) who find a decline of 44 % between 1990 and 2019. EBC and $nssSO_4^{-2}$ are typical anthropogenic combustion tracers during the haze season, and therefore show significant negative trends at several stations during winter in the past

170 decades (Dutkiewicz et al., 2014; Sharma et al., 2019). For summer, a much smaller number of significantly decreasing concentrations has been reported. This is partly because during summer, anthropogenic emissions are transported much less effectively to the Arctic, and EBC as well as $nssSO_4^{2-}$ can also be emitted or derived from natural sources such as wildfires and marine organisms, respectively. The Arctic is undergoing rapid and unprecedented socio-economic as well as environmental changes. The

175 region is warming two to three times faster than the global average, a phenomenon called Arctic amplification (Serreze and Barry, 2011). Sea ice has declined by more than 30 % since the 1970s (Meier et al., 2014), marine microbial activity is changing significantly (Arrigo et al., 2012) and up to 50% increase in wild fire occurrence in the circumboreal region is expected by the end of the century (Flannigan et al., 2009). These changes may have important implications for natural aerosol emissions

180 and abundance (Schmale et al., 2021), particularly in summer, when e.g., BC and OC are emitted from wild fires. Despite the increase in fire emissions in higher latitudes in the past years, no increased contribution during the fire season has been shown so far. This is likely due to the fact that wild fires emit BC and OC further aloft into the atmosphere and in the absence of down-mixing to the boundary layer such aerosol layers are mostly not captured by surface observations. From carbon source apportionment

185 studies based on isotopic ratios, it is known that biomass burning dominates the summertime EBC signal (Winiger et al., 2019). However, there are no long-term isotopic observations, and thus, it is unknown whether the summertime dominance is a new phenomenon or not. In terms of socio-economic changes, impact of emissions from oil fields (e.g., Prudhoe Bay in Alaska), shipping (e.g., in Resolute Bay, Canada), flaring (e.g., in Siberia) and Arctic urban areas have been found to change aerosol composition

190 as well as cloud properties (Kolesar et al., 2017; Aliabadi et al., 2015; Schmale et al., 2018; Gunsch et al., 2017).

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$NssSO_4^{2-}$ can originate from the oxidation of DMS to SO_2 and further to sulfuric acid (Hoffmann et al., 2016), which can then condense onto preexisting particles and contribute to cloud condensation nuclei or form new ones (Hodshire et al., 2019; Beck et al., 2020; Schmale and Baccarini, 2021; Park et al., 2021).

195   DMS is also converted into MSA in the atmosphere (Hoffmann et al., 2016). It is hypothesized that retreating sea ice allows for more algal blooms in the Arctic Ocean, which would result in more DMS emissions and hence more particulate MSA, a unique tracer for this change. Gali et al. (2019) showed a significant increase in DMS emissions between 1998 and 2016 north of 70°N. A few studies have investigated the trends of particulate MSA and $nssSO_4^{-2}$ in the Arctic. Recently, Moffett et al. (2020)

200   found for Barrow that MSA and $nssSO_4^{-2}$ increased by 2.5 % and 2.1 % per year (July to September), respectively, in the period 1998 to 2017. The authors find a positive correlation with ambient temperature and hypothesize that the trend might result from a mixture of causes, which include retreating sea ice, warmer sea surface temperature and temperature dependent atmospheric chemical reactions. At Alert, Sharma et al. (2019) found no significant trend in MSA between 1998 and 2013, while there was a 4 %

205   increase per year (July to August) between 1998 and 2008 (Sharma et al., 2012), reflecting natural variability rather than the effect of sea ice retreat. A study of MSA concentrations at Gruvebadet, Svalbard, and Thule, Greenland between 2010 and 2017 showed that the relationship between retreating sea ice and particulate MSA is complex and likely not direct (Becagli et al., 2019). While concentrations seem to have a positive trend at Gruvebadet, they tend to decline at Thule, while sea ice is retreating in

210   both locations. The presence of algae responsible for the production of DMS precursors in the water is necessary for DMS release to the atmosphere and subsequent production of MSA. A modelling study has shown that even if more DMS is emitted, it does not mean that higher particulate MSA is observed at the surface stations, because MSA might be incorporated into cloud condensation nuclei (CCN) which are preferentially removed from the atmosphere through wet deposition further amplified by the fact that

215   there is an increase in precipitating clouds with retreating sea ice (Mahmood et al., 2019).

Another important natural aerosol component around the Arctic is sea spray, composed of sea salt and organics. Modeling studies have investigated the climatic effects following the hypothesis that retreating sea ice will lead to more sea spray production (Browse et al., 2014; Struthers et al., 2011). However,

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long-term observations that show an increased contribution of sea spray are scarce. Increasing trends in
220 sea salt produced by the sea spray were identified at Alert in 34 years of measurements of Na^+ and Cl^-
(Sharma et al., 2019). Recently, Heslin-Rees et al. (2020) have analyzed long-term trends of aerosol
optical properties from Zeppelin, Svalbard. They found that the scattering Ångström exponent (*SAE*)
became significantly smaller over time. A smaller SAE indicates in general larger particle diameters, and
hence the results provide evidence for a change in particle diameters towards larger sizes. Given that
225 coarse mode particle sources in the Arctic are limited, long-range transported mineral dust or local soil
dust are other potential sources, but sea spray is the main contributing factor. The authors find, however,
that the increased coarse mode is a result of changing atmospheric air mass advection patterns rather than
enhancement of sea spray production in regions with retreated sea ice. In winter, Na^+ is dominated by
blowing snow signals. Higher sea salt contributions at Alert from January to March (Sharma et al., 2019)
230 are found to be due to wind driven resuspension of sea salt originally deposited on the snowpack from
long range transport of sea salt from the North Atlantic Ocean. This contribution is more important than
that from frost flowers (Huang and Jaeglé, 2017) as supported by sulfur isotope analysis (Seguin et al.,
2014). Kirpes et al. (2018) investigated particulate Na^+ at Barrow in winter and found that sea spray
emissions most likely from open leads or open water were responsible for a considerable fraction of sea
235 salt, rather than blowing snow. This might be an indication for a shift in sea salt sources during winter.
However, no long-term analyses have been done.

Summarizing the large body of literature treating annual cycles and long-term trends at Arctic
observatories, we find three main approaches: (i) studies investigated a limited number of aerosol
properties across multiple stations for a short period of time (e.g. the systematic comparison of optical
240 properties (Schmeisser et al., 2018) or BC and SO_4^{2-} (Eckhardt et al., 2015), (ii) studies focused on one
station entirely reporting a large number of parameters for seasonal cycle and trend analyses (e.g., Sharma
et al., 2019), or (iii) studies discussed long-term trends of a small number of anthropogenic tracers across
multiple stations over different time periods (e.g., Quinn et al., 2007; Collaud Coen et al., 2020b). The
resulting gaps are a missing pan-Arctic synthesis of anthropogenic and natural seasonal and long-term
245 trend patterns over comparable time-periods, as well as a joint consideration of chemical and optical
aerosol properties and their seasonal and long-term co-evolution.

To close these gaps, here, we discuss seasonal cycles of nine chemical and four optical properties across ten stations averaged over the same time-period, and we derive decadal trends per variable and station, which can be compared to each other. We further include anthropogenic as well as natural aerosol tracers.

250 The purpose of this study is twofold: (i) we aim to reveal similarities and differences in seasonal patterns to highlight the regional nature of the Arctic aerosol regimes, and (ii) to contrast anthropogenic and natural aerosol long-term trends against decreasing anthropogenic emissions and discuss these trends in the context of environmental changes. This study follows chapter 5 of the recently released assessment report of short-lived climate forcers in the Arctic by the Arctic Monitoring and Assessment Programme (AMAP,

255 2021).

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2 Methods
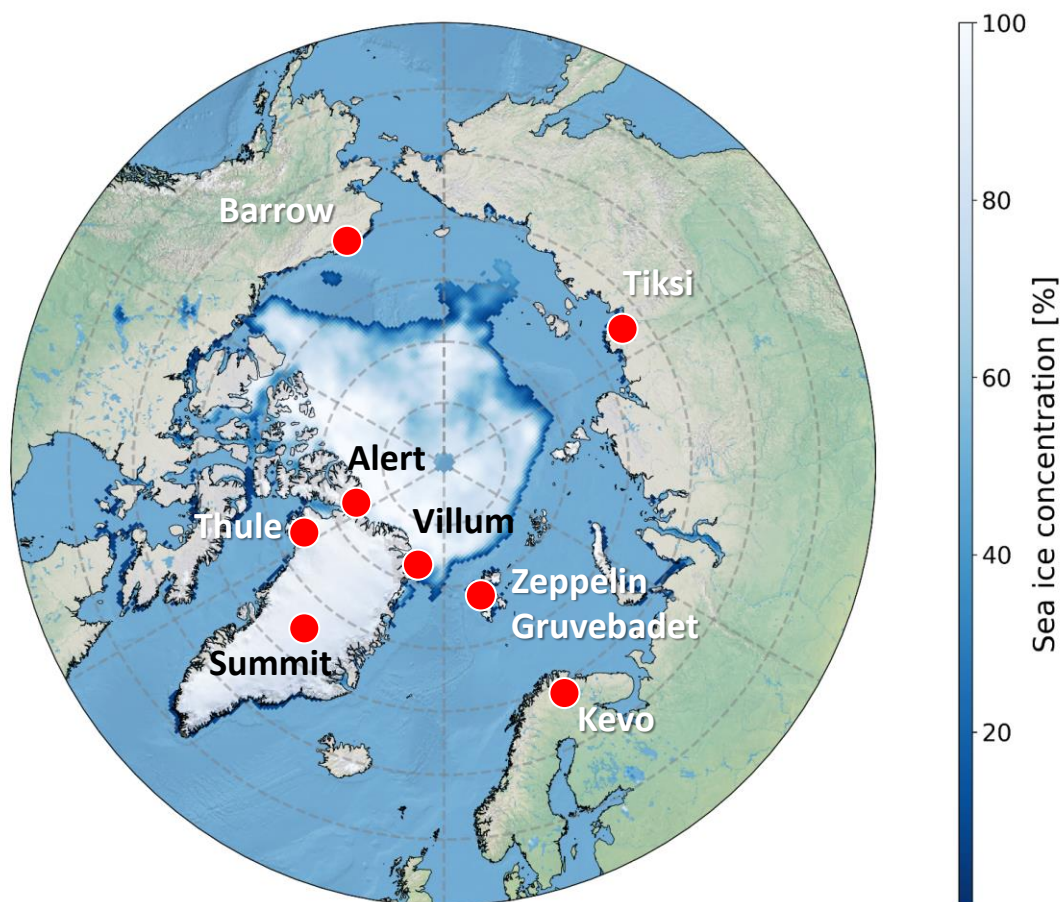
Figure 1: Location of observatories discussed in this study. The blue shading represents sea ice concentration (15 September 2018), reported as a fraction of covered surface (Maslanik and Stroeve, 1999). The base map is from Natural Earth II (https://www.naturalearthdata.com/).

2.1 Data and data treatment

We obtained final, corrected data either from a publicly accessible data repository (EBAS, http://ebas.nilu.no/) or through personal communication with station PIs (see Table 1). Data temporal resolution varied from hourly for aerosol optical properties to several days for chemical components. Figure 1 shows data from ten observatories where various in situ measurements were conducted. The

chemical variables we consider include equivalent black carbon (EBC), particulate sulfate (SO_4^{2-}), nitrate

270 (NO_3^-), ammonium (NH_4^+), MSA, sodium (Na^+), non-sea salt calcium ($nssCa^{2+}$), non-sea salt potassium ($nssK^+$) and iron (Fe). We calculated the nss components Ca^{2+} and K^+ based on their abundance in sea salt relative to the Na^+ concentration (Pilson, 2012). The former can be regarded as tracer for soil dust and the latter for biomass burning (e.g., Sharma et al., 2019). Table 1 gives an overview of the measurement methodology for each species at the different sampling sites.

275 We also report four different optical properties, the scattering (σ_{sp}) and absorption (σ_{ap}) coefficients at 550 nm, single scattering albedo (*SSA*) at 550 nm, and scattering Ångström exponent at 550/450 (*SAE*). While the first two variables depend on the total aerosol concentration, i.e. extensive properties, the *SSA* and *SAE* are intensive properties which do not depend on loading. The *SSA* is a measure of how much the aerosol population is able to scatter light relative to the extinction (absorption+scattering), i.e. a value of

280 *SSA* equal to 1 indicates a purely scattering aerosol. *SSA* was calculated by dividing the scattering coefficient by the sum of the scattering and absorption coefficients. The *SAE* describes the wavelength dependence of the aerosol scattering coefficient and can be interpreted as a measure of particle size, where larger particles have smaller *SAE* values (Delene and Ogren, 2002). We calculated *SAE* based on the following Eq. 1:

285 $$SAE = -\frac{\log(\sigma sp_{450}) - \log(\sigma sp_{550})}{\log(\lambda_{450}) - \log(\lambda_{550})}$$ Equation 1

where σ_{sp} is the scattering coefficient at the indicated wavelength (λ). Values below 0.045 Mm^{-1} for σ_{sp} were not used in the calculation of the *SAE* since they were found to be close to detection limit.

Not all species are available for every station. Table 2 gives an overview of the variables and the periods for which they were obtained at each station.

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Table 1: Overview over methods applied to derive aerosol properties and the respective data sources

Station	Variables	Method	Remark	Data Source
Alert	SO_4^{2-}, NO_3^-, NH_4^+, MSA, Na^+, K^+	HI-volume sampler, ion chromatography,	Total suspended particulates (TSP)	EBAS* (Sharma et al., 2019)
	EBC	Aethalometer AE06	880 nm, TSP	EBAS (Sharma et al., 2019)
	Ca^{2+}, Fe	Instrumental neutron activation analysis (INAA), inductively coupled plasma mass spectrometry (ICP-MS)	TSP INAA: 1988 – 2013 for Ca ICP: 1980 – 1997 for Ca, 1980 – 2007 for Fe, data were averaged when both methods are available	(Sharma et al., 2019)
	Scattering coefficient	Nephelometer (TSI 3563)	550 nm, particulate matter < 1 µm (PM_1)	S. Sharma, personal communication
	Absorption coefficient	Particle soot absorption photometer, continuous light absorption photometer (PSAP/CLAP) 1λ and 3λ	PM_1, converted to 550 nm (via inverse wavelength relation)	S. Sharma, personal communication

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Barrow	SO_4^{2-}, NO_3^-, NH_4^+, MSA, Na^+, K^+	Ion chromatography, PM_1 (supermicron data are also available but were not used here)		(Quinn et al., 2007), submicron : https://data.pmel.noaa.gov/pmel/erddap/tabledap/submicron.html
	EBC	Aethalometer and PSAP	PM_{10} measured or converted to 880 nm, AE8 until 2001, PSAP 1λ 2001 – 2006, PSAP 3λ 2006 – 2010, AE31 from 2010 – 2016, AE33 after 2016	(Sharma et al., 2013), and for newer values personal communication E. Andrews
	Scattering coefficient	Nephelometer (TSI 3563)	550 nm, PM_{10}	EBAS
	Absorption coefficient	PSAP, CLAP	PM_{10}, 1λ PSAP 1997-2006 3λ PSAP 2006 – 2014 3λ CLAP 2014 - present, converted to 550 nm	EBAS
Zeppelin	SO_4^{2-}, MSA, Na^+, Ca^{2+}, K^+	Ion chromatography	PM_{10}	(Sharma et al., 2012)
	EBC	Aethalometer (AE31)	880 nm, TSP	personal communication: K. Eleftheriadis, and (Eleftheriadis et al., 2009)
	Scattering coefficient	Nephelometer (TSI 3563)	TSP	(Heslin-Rees et al., 2020)

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Villum	SO_4^{2-}, NO_3^-, NH_4^+, Na^+, K^+, Ca^{2+}	Ion chromatography, proton induced X-ray emission (PIXE) , ICP-MS	SO_4^{2-}, NO_3^-, NH_4^+, Na^+: Ion chromatography PIXE: 1990 – 2009 for K^+, Ca^{2+} ICP-MS: 2011 – 2017 for K^+, Ca^{2+} TSP	A. Massling, personal communication
Gruvebadet	SO_4^{2-}, NO_3^-, MSA	Ion chromatography,	PM_{10}	R. Traversi, personal communication (Becagli et al., 2019, 2016)
	EBC	PSAP 3λ	TSP	(Gilardoni, 2019)
Kevo	EBC	Thermal optical methods	TSP	(Dutkiewicz et al., 2014)
Tiksi	EBC	Aethalometer (AE31)	880 nm, PM_{10}	EBAS
	Scattering coefficient	Nephelometer (TSI 3563)	550 nm, ambient humidity, PM_{10}	EBAS
	Absorption coefficient	Aethalometer (AE31)	converted to 550 nm, PM_{10}	EBAS
Thule	SO_4^{2-}, NO_3^-, MSA	Ion chromatography	PM_{10}	(Becagli et al., 2019)
Summit	Scattering coefficient	Nephelometer (TSI 3563)	550 nm, $PM_{2.5}$	EBAS
	Absorption coefficient	CLAP-3λ	550 nm, $PM_{2.5}$	EBAS
Pallas	Scattering coefficient	Nephelometer (TSI 3563)	550 nm, TSP until 2008, from 2008 PM_{10}	EBAS

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	Absorption coefficient	MAAP (Thermo_5012)	637 nm**, PM_{10}	EBAS

* ebas.nilu.no/

** Converted to 550 nm based on equation 5 in Schmeisser et al. (2018) and assuming the Ångström exponent to be equal to one.

2.2 Annual cycles and trend calculations

For the annual cycles, we calculated the median and interquartile range from all data points per month for the available periods. To derive the anomaly, we subtracted the overall median value and normalized by the overall interquartile range. The overall values are based on all data entries available for the given time period. They are shown in the box plots in Figure 2. Note that data for Gruvebadet is not available for all winter months.

We derived long-term trends for all variables, where data from at least five consecutive years were available. Trends are calculated for the haze season (January, February, March, April, JFMA) and the summer season (June, July, August, September, JJAS) following the example of Quinn et al. (2009) for data obtained at Barrow. This allows for comparison with previously derived trends for aerosol properties at Arctic stations. The haze season is dominated by anthropogenic emissions, while the summer season reflects a mix of anthropogenic and natural emissions. The seasonal separation hence allows tracking the evolution of anthropogenic and natural influences more specifically. It also avoids the need for prior de-seasonalization of the data. Note, for MSA, we divided the seasons into April-May, and June-August, because there were not enough data above the detection limit before and after. A similar approach to separate data to derive seasonal trends has been introduced by Hirsch et al. (1982). Trend significance was tested with the non-parametric Mann-Kendall test on each season. Non-parametric means that no prior assumption of a specific distribution is necessary. The test has been widely applied to environmental data sets including in the Arctic (Tunved and Ström, 2019; Skov et al., 2020; Heslin-Rees et al., 2020; Collaud Coen et al., 2020b, a), not least because of its non-parametric nature and its tolerance of missing

values. The null hypothesis of the test is that no trend exists. We defined a trend to be significant if the p-value is smaller or equal to 0.05 (green dots and upward pointing arrows in Table 2). Trends with a p-value between 0.05 and 0.1 are marked orange and larger values (p-value > 0.1) in red (Table 2). The results of the Mann-Kendall test are strongly dependent on autocorrelation, whereby a positive autocorrelation increases the likelihood that a trend is determined when there is none (Kulkarni and von Storch, 1995). To avoid an autocorrelation signal, and to accommodate the highly diverse time resolution of the data set, we calculate seasonal median values. The slopes of the trends were calculated using the Theil-Sen slope estimator, another non-parametric method. Significance and slope were derived for entire time series and on a decadal basis. Values for specific decades allow for intercomparison between variables and stations. However, decadal values are also given if there are at least five consecutive years of data within a decade. Hence the time periods within decades are not equal in all cases and for all stations. Table 2 shows with the grey bars for which years which variables are available per station. At Villum, a large time gap exists between 2003 and 2007. We derived trends separately for before and after the gap.

It is important to note that the Mann-Kendall test requires a monotonic trend for the time period over which it is calculated. This is often the case for anthropogenic tracers during the haze season. However, some variables do not show a monotonic behavior. For example, at Alert MSA exhibited a significant negative trend between 1981 and 1999 and a non-significant positive trend thereafter (Sharma et al., 2019). To reflect this, we split the data set and calculated trends separately.

## 3 Results

3.1 Annual cycles

3.1.1 Chemical composition

Figure 2 shows the statistical distribution of concentrations per chemical species, as box plots for each station on the right side and as monthly anomalies of the normalized concentrations as annual cycles on the left side.

SO_4^{2-} makes up the highest concentrations at each station with medians between 0.1 and 0.3 µg m^{-3}, followed by Na$^+$ (0.05 to 0.15 µg m^{-3}). NH$_4^+$, NO$_3^-$, EBC, Fe and nssCa^{2+} all range between 0.01 and 0.06 µg m^{-3}. NssK$^+$ and MSA exhibit smaller concentrations between 0.003 and 0.015 µg m^{-3}. EBC, SO$_4^2$, NO$_3^-$ and NH$_4^+$ are typical anthropogenic tracers during the Arctic haze season where the highest levels are observed. Their dominant sources in the mid-latitudes are fossil fuel combustion (EBC, SO$_4^{2-}$, NO$_3^-$), residential heating (EBC) and agriculture (NH$_4^+$) (AMAP, 2015). With regards to EBC, Tiksi (71°N) and Kevo (69.4°N) experience the highest concentrations and the largest variability, likely a result of their proximity to both anthropogenic source regions and potential local influences as well. The annual cycles are similar for all stations showing the typically elevated concentrations during the haze period, however, Zeppelin experiences relatively lower concentrations in January and February. This is likely related to stronger influence of precipitation on the particles concentrations, because the island of Svalbard experiences the precipitation maximum during winter, in contrast to all other regions in the Arctic, where the maximum precipitation occurs in (late) summer (Freud et al., 2017).

Alert, Zeppelin, Barrow and Villum are comparable in their SO$_4^{2-}$ concentrations and variability. The four stations show almost identical annual cycles with the characteristic haze peak in late winter/early spring and the minimum in summer. Thule and Gruvebadet show a more pronounced relative increase in April. NH$_4^+$ and SO$_4^{2-}$ are very similar in their annual cycles, which is consistent with the formation of ammonium sulfate or ammonium bisulfate. At Barrow NH$_4^+$ concentrations are roughly twice as high as at Alert and Villum, indicating higher availability of NH$_4^+$ and less acidic particles since the SO$_4^{2-}$ concentrations are similar at all three stations. At Villum and Alert, ammonium's statistical distribution is almost identical. NO$_3^-$ concentrations are higher than those of NH$_4^+$ at these two stations and variable. This could be related to atmospheric chemical processes of particulate NO$_3^-$ formation, which depends on the aerosol acidity, water content and ambient temperature (see the discussion of the long-term trends for more details).

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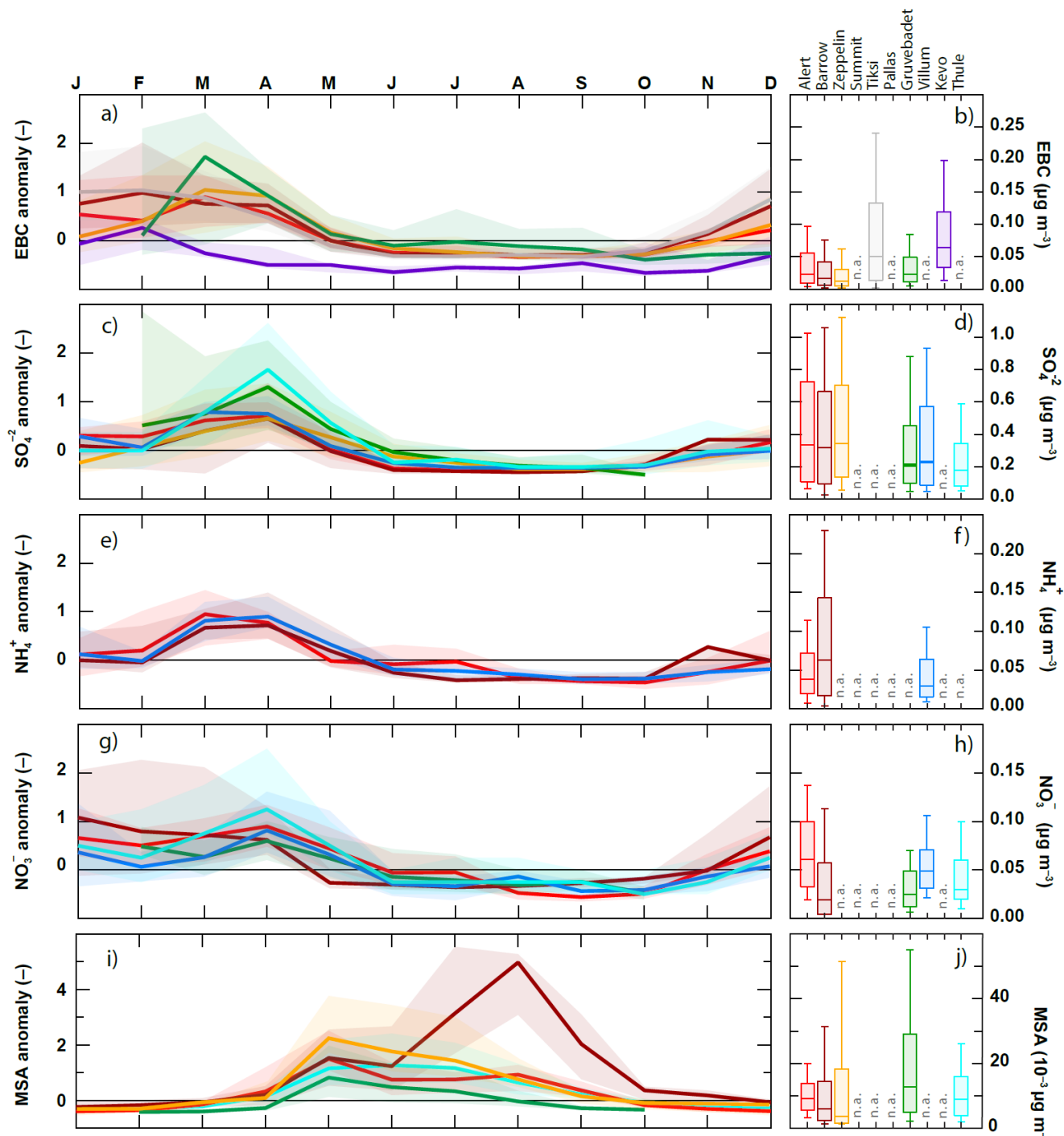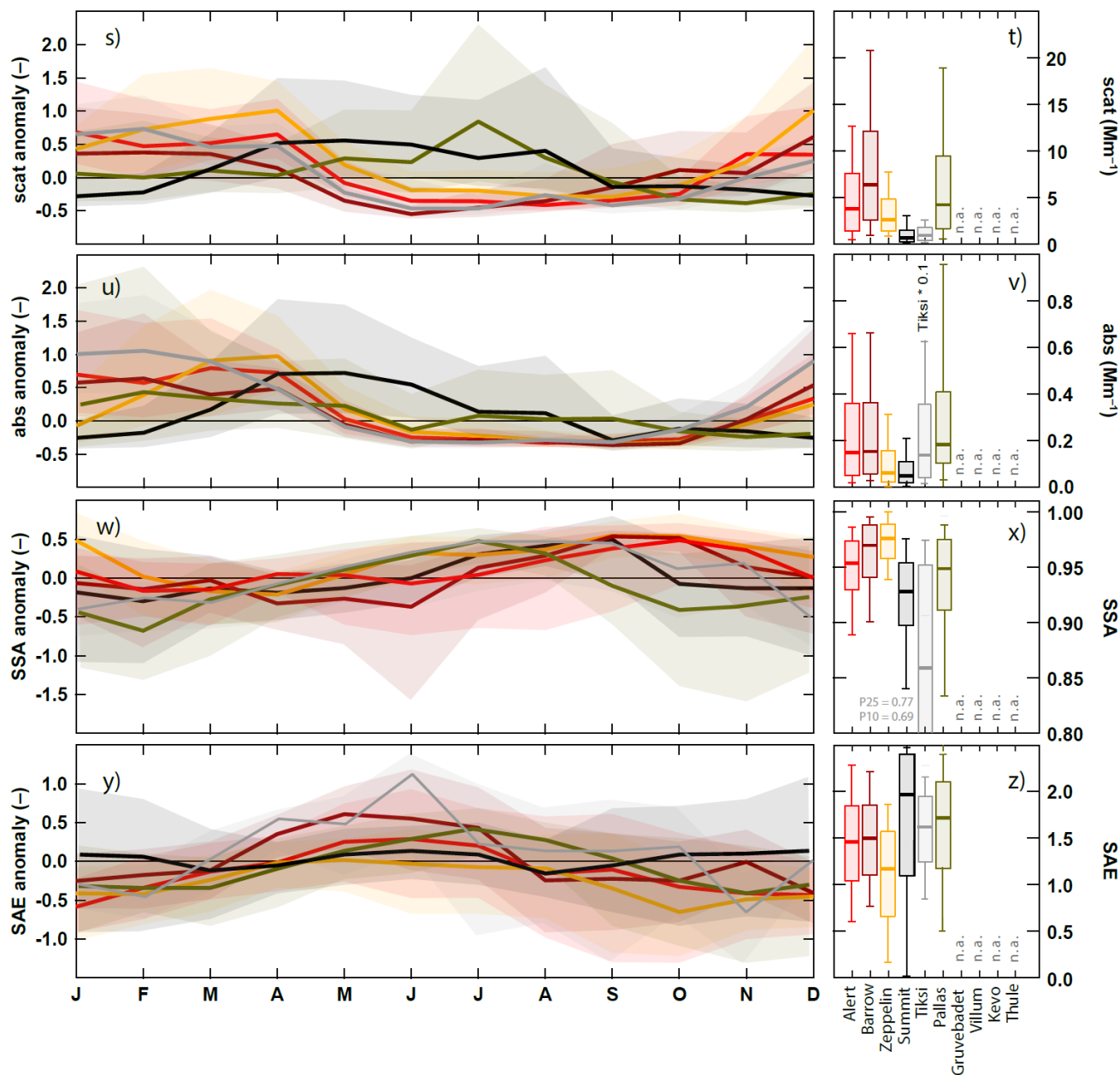
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Figure 2: The left column shows annual cycles of aerosol chemical compounds and physical properties for each available data set as anomaly. The data are normalized by the median and interquartile range (see Methods). Colors represent stations given in the right column of the figure (see panel b). The right column provides statistical values (median, interquartile range and 10th and 90th percentiles) per variables and station. Chemical compounds were derived from the following size ranges (see also Table 1): TSP for Alert and Villum, PM10 for Zeppelin, and PM1 for Barrow.

The other chemical tracers are less uniformly distributed throughout the year and there are significant differences among the stations. MSA is an oxidation product formed from DMS, which is derived from marine algae. It is hence associated with the presence of open water (or marginal sea ice zones), nutrients, sunlight and DMS producing algae as is reflected in the annual cycle. At all stations, a characteristic spring bloom peak in April-May is visible (Becagli et al., 2019; Moffett et al., 2020). In August, Barrow features a second and even higher peak, likely influenced by regional phytoplankton blooms (Moffett et al., 2020; Quinn et al., 2002). The other three stations do not show such strong signals and their highest MSA peaks are confined to spring, when southerly air masses transport MSA from productive waters in the mid-latitudes (Sharma et al., 2019).

Another natural contribution to aerosol is Na^+, which originates from sea spray and salt-laden blowing snow. The annual cycles at Alert and Barrow are similar with highest concentrations in winter, which is evidence of mixed contributions from blowing snow (Huang and Jaeglé, 2017) and southerly advection of sea spray (Quinn et al., 2002). Zeppelin shows lower enhancement during winter likely because of its elevation (476 m a.s.l). Villum features an annual cycle very similar to Alert, albeit with less enhancement in between November and December.

$NssCa^{2+}$ and Fe have been used as mineral and soil dust tracer by Sharma et al. (2019) for Alert, where they show a spring and autumn peak. The spring peak has been interpreted as long-range transported dust from Asian deserts based on a Lagrangian model study (Groot Zwaaftink et al., 2016), whereas the autumn peak corresponds to minimum snow coverage and hence potential local soil dust sources. The latter could also be the case for Villum for $nssCa^{2+}$, where the main peak occurs in autumn, but the actual source is difficult to pinpoint (Nguyen et al., 2013; Heidam et al., 2004). Barrow exhibits a strong $nssCa^{2+}$ enhancement between January and February, which has been interpreted as a signal of long-range transported dust from Eurasia (Quinn et al., 2002). At Zeppelin the annual cycle does not show any pronounced features.

$NssK^+$ can be interpreted as biomass burning tracer originating from domestic wood burning, agricultural or forest fires contributing during different times of the year. At Alert and Barrow the annual cycle of $nssK^+$ is similar to that of typical anthropogenic tracers, e.g., EBC, and has been identified as a haze tracer at Barrow by Quinn et al. (2002). Based on carbon isotope measurements and Lagrangian dispersion

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modeling it has been shown that domestic wood burning is an important source of black carbon in winter (Stohl et al., 2013; Winiger et al., 2019). Hence, it is conceivable that nssK$^+$ could also be an indication of domestic wood burning. However, at Villum nssK$^+$ has been found to originate potentially also from fossil fuel burning (Nguyen et al., 2013; Heidam et al., 2004). The seasonal cycle of nssK$^+$ at Villum

410   shows a late summer enhancement from soil dust (Heidam et al., 2004). At Zeppelin nssK$^+$ remains enhanced throughout May to September. The presence of nssK$^+$ and its interpretation as biomass burning marker is in agreement with carbon isotope source apportionment by Winiger et al. (2019), who show an elevated level of 60 % biomass burning contribution to black carbon at the station. It has been shown for Zeppelin that long-range transport of agricultural fires can have a strong impact on the aerosol population

415   in spring (Stohl et al., 2007).

3.1.2 Optical properties

Figure 2 also shows aerosol optical properties, which are tightly linked to the chemical composition and absolute aerosol mass for the scattering and absorption coefficients. Similar figures showing the absolute

420   values as annual cycles have been discussed in Schmeisser et al. (2018) for data between 2012 and 2014, and by Quinn et al. (2002) and Delene and Ogren (2002) for Barrow for 1997 to 2000. Here, we show all data available to this study (see Table 2). The annual cycle of σ_{sp} reflects the higher aerosol burden during the haze season and the low concentrations during summer for the observatories at Alert, Barrow, Zeppelin and Tiksi. Pallas and Summit exhibit an opposite behavior with peak values in spring and

425   summer, respectively. Pallas, being located relatively far south is much more influenced by Eurasian anthropogenic emissions being downwind (easterlies prevail) of significant emission sources (Aalto et al., 2002; Asmi et al., 2011) and also by biogenic emissions from the surrounding boreal forest (Hyvärinen et al., 2011; Tunved et al., 2006). Summit is located 3238 m a.s.l. and is therefore not affected by winter haze, which manifests itself in the lower layers of the atmosphere. It is however subject to long-range

430   transport of biomass burning and anthropogenic emissions during spring and summer (Schmale et al., 2011; Thomas et al., 2017; Stohl et al., 2006). The σ_{ap} is closely related to EBC (and correlated 1:1 for all stations where EBC was derived with a uniform mass absorption cross section, except at Kevo where a

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thermal-optical method was used) and both variables follow a similar annual cycle (Figure 2, panels a, u) with the maximum values during the haze period at Alert, Barrow, Tiksi and Zeppelin. Summit shows

435 much lower σ_{ap} values and follows a different annual cycle with a maximum in spring and summer, comparable to the observation of σ_{sp}. Here again, the influence of long-range transported biomass burning emissions as well as emissions from other anthropogenic sources that can reach Summit throughout spring and summer is evident as well as the absence of haze pollution in winter. *SSA*, an inherent aerosol property independent of the absolute abundance of aerosol observed at Alert, Barrow, Zeppelin, Tiksi, Pallas and

440 Summit, peaks in autumn and reflects the minimum in the σ_{ap} during that time of the year. At Pallas *SSA* peaks earlier in summer than at the other stations and also drops more strongly during winter, likely due to the anthropogenic influence in winter and the stronger boreal forest contribution in summer. The lower *SSA* at Summit in autumn compared to the other stations has been interpreted by Schmeisser et al. (2018) as a darker background aerosol due to increased local air traffic from the station (despite a pollution sector

445 controlled data product). *SAE*, another intensive property, behaves similarly at all six stations (Alert, Pallas, Summit, Barrow, Tiksi, and Zeppelin) with a peak in spring lasting through early summer. Given that larger *SAE* values are indicative of smaller particle diameters, this behavior is in agreement with the annual cycle of the particle number size distributions as discussed e.g., in Croft et al. (2016) and Freud et al. (2017) due to the absence of larger submicron modes. During the *SAE* peak, more frequent new particle

450 formation enhances the Aitken mode population, while regular wet deposition depletes the coarser particles acting as cloud condensation nuclei (Freud et al., 2017).
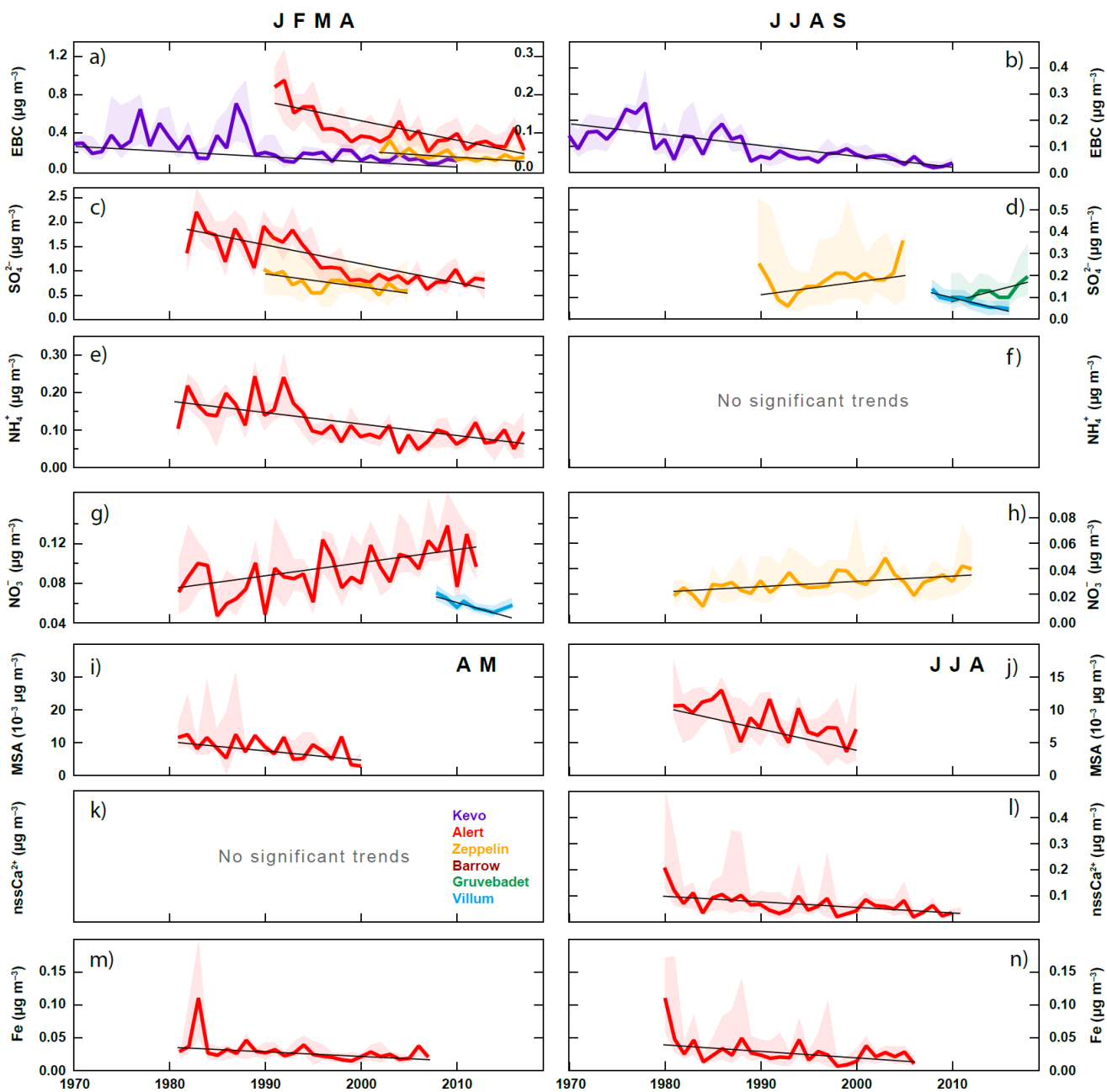
3.2 Trend calculations based on Mann-Kendall and Theil-Sen's Slope

Table 2 gives an overview of the calculated trends and their significance for all stations and parameters.

455 The left part of the table provides the significance of the overall trend as a numeric value and optically by a colored marker (green means significant, see Methods) split by haze (JFMA) and summer (JJAS) seasons. An upward or downward pointing arrow, only for significant trends, represents the direction of the slope and the value indicates the change per year in the unit of the variable as indicated in the leftmost column. To the right the duration of the measurements are shown as grey bars, and within each decade

460   the marker and arrow indicate the decadal trends and their significance. Numerical values are given in SI

Table S1. Generally, the optical property records represent the last two decades and are shorter than

chemical records at several stations, which date back about three decades. For MSA, the record across

stations is fragmented, with only Alert and Barrow providing decadal information.

465   **Table 2. Data coverage and trends. The grey bars on the right side indicate data availability per station and variable. The p-values and slopes on the left side indicate the overall trends characteristics per station and variables separated by season (JFMA and JJAS). Round markers indicate the significance of the trend, where green corresponds to a p-value < 0.05, orange to 0.05 < p-value < 0.10, and red p-value > 0.10. The arrows indicate whether the trend is positive or**
470   **negative. Decadal trend significance and direction are given by markers only in the respective time period. The two left (right) symbols correspond to JFMA (JJAS). Chemical compounds were derived from the following size ranges (see also Table 1): TSP for Alert and Villum, PM_{10} for Zeppelin, and PM_1 for Barrow*.**
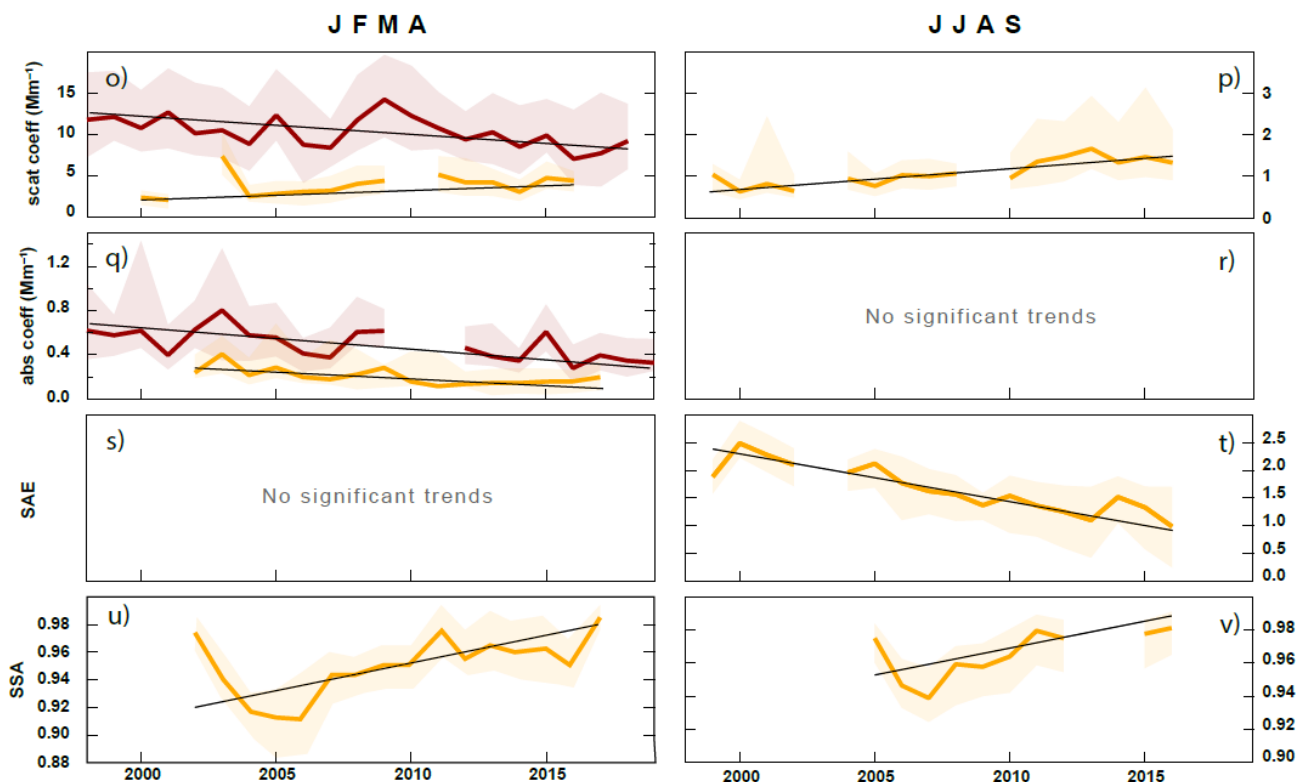
species	station	all years JFMA p-value	slope	all years JJAS p-value	slope
EBC µg m⁻³	Alert	0.00 ↓	-2.61E-03	0.19	
	Barrow	0.09 ↓	-1.22E-03	0.17	
	Zeppelin	0.03 ↓	-1.05E-03	0.44	
	Gruve	0.80		0.26	
	Kevo	0.00 ↓	-5.71E-03	0.00 ↓	-3.75E-03
	Tiksi	0.33		0.85	
SO₄²⁻ µg m⁻³	Alert	0.00 ↓	-3.67E-02	0.09 ↓	-7.50E-04
	Barrow	0.57		0.79	
	Zeppelin	0.01 ↓	-2.33E-02	0.02 ↑	7.50E-03
	Gruve	0.83		0.05 ↑	9.29E-03
	Villum (1992 - 2002)	1.00		0.45	
	Villum (2008 - 2017)	0.13		0.02 ↓	-6.86E-03
	Thule	0.28		0.65 ↑	5.00E-03
NO₃⁻ µg m⁻³	Alert	0.00 ↑	1.38E-03	0.00 ↑	4.66E-04
	Barrow	0.59		0.07 ↑	1.67E-04
	Gruve	1.00		1.00	
	Villum	0.10 ↑	2.89E-03	0.14	
	Thule	0.30		0.70	
NH₄⁺ µg m⁻³	Alert	0.00 ↓	-3.02E-03	0.70	
	Utqiagvik	0.97		0.16	
	Villum	0.09 ↓	-1.42E-03	0.55	
MSA µg m⁻³	Alert (till 1999)	0.02 ↓	-3.29E-04	0.00 ↓	-2.80E-04
	Alert (from 2000)	0.79		0.14	
	Barrow	n/a		0.79	
	Zeppelin	0.17		0.55	
	Gruve	0.14		0.14	
	Thule	NA	NA	0.60	
Na* µg m⁻³	Alert	0.36		0.97	
	Barrow	0.57		0.97	
	Zeppelin	0.98		0.33	
	Villum	0.37		0.75	
nssCa²⁺ µg m⁻³	Alert	0.42		0.00 ↓	-2.15E-03
	Barrow	0.38		0.38	
	Zeppelin	0.82		0.58	
	Villum	0.76		0.81	
nssK⁺ µg m⁻³	Alert	0.10		0.97	
	Barrow	0.82		0.15	
	Villum	0.33		0.22	
Fe, µg m⁻	Alert	0.00 ↓	-4.73E-04	0.02 ↓	-5.54E-04
Scat Mm⁻¹	Alert	0.12		0.11	
	Barrow	0.01 ↓	-1.75E-01	0.67	
	Zeppelin	0.01 ↑	1.79E-01	0.00 ↑	5.48E-02
	Summit	0.46		0.62	
	Tiksi	NA	NA	NA	NA
	Pallas	0.14		0.24	
Abs Mm⁻¹	Alert	0.09 ↓	-1.50E-02	0.38	
	Barrow	0.00 ↓	-1.37E-02	0.56	
	Zeppelin	0.02 ↓	-7.84E-03	0.80	
	Summit	0.14		0.32	
	Pallas	0.39		0.71	
	Tiksi	0.28		0.77	
SSA	Alert	0.81		0.81	
	Barrow	0.05 ↑	9.18E-04	0.24	
	Zeppelin	0.11		0.01 ↑	2.82E-03
	Pallas	0.39		0.39	
	Tiksi	NA		NA	
	Summit	0.88		0.14	
SAE	Alert	0.10 ↓	-3.65E-02	0.11	
	Barrow	0.90		0.07 ↓	-1.32E-02
	Pallas	0.75		0.26	
	Tiksi	NA		NA	
	Zeppelin	0.00 ↓	-6.63E-02	0.00 ↓	-8.43E-02

(The right portion of the table contains a year grid spanning 1980–2019 divided into JFMA and JJAS seasonal columns with colored markers and trend arrows.)

475

n/a means there are not enough values above detection limit to calculate a p-value.

* For Barrow, optical property data go further back in time than shown here (see e.g., Collaud Coen et al., 2020b). Here, we use the same time series as shown in the AMAP report (2021), sind this work is based on it.

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Figure 3: Significant trends (p < 0.05) per variable and station for the entire time period over which data are available. For Villum we considered the time period 2007-2016 here. MSA is given for the months of April-May and June-July-August.

485

3.2.1 Long-term trends of SO_4^{2+}, NH_4^+, NO_3^- and EBC

For the full period of each measurement, during the haze season 29 % of the trends are significant ($p <$ 0.05) with 13 negative, and three positive trends. For the summer, 21 % of overall trends are significant

490 with six negative and six positive trends. We show all 28 significant trends in Figure 3. In winter, typical haze tracers such as EBC, SO_4^{2-} and NH_4^+ make up seven of the 13 significant and full period negative trends at Alert, Zeppelin, Villum and Kevo. This is consistent with the reduction of air pollution emissions in North America, Europe and Russia, which we show in Figure 4 based on the ECLIPSEv6b (Höglund-Isaksson et al., 2020) and CMIP6 emission data between 1970 and 2014 (Hoesly et al., 2018). For BC,

495 the emissions data show a gradual decrease from 1970 to 2000 for the Western Arctic (Western Arctic

27

refers to the USA and Canada), while emissions in the Eastern Arctic (Eastern Arctic refers to the Kingdom of Denmark, Finland, Iceland, Norway, the Russian Federation, and Sweden) and the rest of Europe show a strong decrease between 1990 and 2000. Asia (Japan, People's Republic of China, Republic of India, Republic of Korea, Republic of Singapore) shows a decrease in the 1990s and a

500   subsequent increase since the early 2000s. The EBC trend at Kevo reflects the sharp decline in 1990, and also at Alert most of the EBC concentration decline happened before 2000, reflecting primarily the economic change in the former Soviet Union (Dutkiewicz et al., 2014; Sharma et al., 2019). However, a significant decadal winter trend is only visible for Alert and Barrow in the 1990s. Otherwise, decadal trends are not significant. Emissions in the 2000s were slightly on the rise for northern Eurasia and North

505   America, and strongly elsewhere. Arctic EBC concentrations do not show a clear decline from 2000 onwards during winter, likely a result of the short period analyzed and the interannual variability.

To demonstrate the influence of short-term variability in the last decade on the significance of trends, we tested the required yearly rate of increase or decrease in EBC concentrations to detect a significant trend (p-value < 0.05) within the last ten or close to ten years of measurements at the following stations: Alert

510   (2007 - 2017), Zeppelin (2007 - 2017), Barrow (2009 – 2019), Gruvebadet (2011 – 2018), and Tiksi (2010 – 2018). We calculated the trend line for the years given in parenthesis, de-trended the EBC concentration data with it and then added x_i % of EBC increase per year in steps of 1 (i.e. $x_1 = 1$ %, $x_2 = 2$ % and so on) until the trend emerged as significant. The results for Alert and Zeppelin are 5 % yr^{-1}, for Barrow 6 % yr^{-1}, for Tiksi 11 % yr^{-1}, and for Gruvebadet 16 % yr^{-1}. Hence, the present-day short-term variability

515   requires relatively large changes to occur to derive significant trends with the Mann-Kendall Theil-Sen method for time periods less than 10 years. This explains why trends on the order of 1% or less per year cannot be significant over single decades but are significant over a record of 20 or more years (as for the case of EBC at Zeppelin in winter, see Table 2). Therefore, even if none of the EBC trends in the haze season turned out to be significant for the 2010-2019 period (in contrast with previous decades), such

520   "stagnation" must be considered carefully. In addition, detection of a 1 % change would not reflect large changes in air quality. The absence of trend detection over shorter periods also highlights the importance of maintaining monitoring activities at the Arctic observatories in the long run, as decadal changes

(occurring over periods of > 10 years) in atmospheric composition can be detected with more statistical certainty.

525 SO_2 emissions, a precursor to particulate SO_4^{2-}, decreased strongly in Europe and the Western Arctic countries since 1980, and showed a clear dip between 1990 and 2000 in the Eastern Arctic, probably due to the economic change in the former Soviet Union. Elsewhere emissions increased strongly, except for the late 1990s in Asia. Winter concentration trends at Alert and Zeppelin follow this pattern, showing the strongest decline in the 1990s and then leveling off. There is only one significant decadal wintertime trend

530 for SO_4^{2-}, which occurred at Alert in the decade of the strongest SO_2 emission reductions, 1990-2000. There are only few observations of NH_4^+, and only Alert shows a negative overall trend during the haze season. NH_4^+ forms ammonium bisulfate and, to a lesser extent, ammonium sulfate in the particle phase and hence follows the trend of SO_4^{2-}. This is also observed at the other two stations that measured NH_4^+, Barrow and Villum, but the p-values do not indicate significance for the overall period ($0.05 <$ p-value

535 <0.1).

NO_3^- shows opposing significant trends, i.e. positive at Alert and negative at Villum. It is important to keep in mind that Alert is the only site where a measurement record longer than 30 years has been acquired. For Villum the time span of data available (2007 to 2017) is short. The positive trend at Alert is opposite to the emission reduction efforts of NO_x (see Figure 4), the precursor to NO_3^-. However, with

540 the reduction in particle acidity, mainly caused by lower emissions of SO_2 and subsequent formation of particulate sulfate, more particulate nitrate can be formed (Sharma et al., 2019). In addition, cold temperatures and high particle water content will favor the partitioning of nitrate into the aerosol phase (Guo et al., 2016; Shah et al., 2018). The analysis of NO_3^- clearly shows that not only emission changes are relevant for chemical trends of Arctic aerosols, but also the type of dominating chemical reactions

545 that change due to an altered chemical composition of the atmosphere. Moreover, there seem to be different processes at play at different stations because Villum shows a different record compared to Alert. Some of the above discussed tracers also show significant trends during summer for the overall period. EBC decreased at Kevo, here again with almost a step change in the 1990s, due to the site's proximity to the former Soviet Union which collapsed in the 1990s. Sulfate decreased significantly at Villum over the

550 periods from 2008 to 2017, however from 2010 to 2017 there is no significant trend. Thus, our primary

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conclusion is that the duration of observations is not long enough to show significant trends with the methods applied here. Enhanced microbial activity during the summer has been hypothesized to be a responsible factor for particle mass formation at Villum (Dall´Osto et al., 2018), even though the summer trend is negative. However, the aforementioned study focused on the smallest particles, while the filter

555 based determination of SO_4^{2-} targets the accumulation mode and even larger particles. Similar new particle formation studies have been conducted for Zeppelin and Gruvebadet (Becagli et al., 2019; Dall´Osto et al., 2017; Beck et al., 2020), where SO_4^{2-} trends are positive and significant during summer. A plausibility check, on whether marine microbial activity is the driving factor, could be done using MSA concentrations. While those are not available at Villum, no significant trend in MSA has been found at

560 Zeppelin station. The absence of an MSA trend might be an indication that reaction pathways are too complex to expect a directly measurable concentration effect in both particulate SO_4^{2-} and MSA based on microbial activity. Also note that Villum is sea ice dominated for ten months of the year. Mahmood et al. (2019) found that enhanced microbial emissions can lead to higher CCN number concentrations which implies a subsequent removal of the corresponding emission products by wet deposition and hence might

565 not be observed at permanent stations. The increase in SO_4^{2-} around Svalbard is likely not due to increased shipping activities, e.g. from tourism. Eckhardt et al. (2013) show a strong influence in summer on sulfur from ships directly in Kongsfjorden, but on an annual basis this is a small effect. Moreover, recently new emission regulations require low sulfur-content fuel.

Particulate nitrate, consistent with the wintertime trends, shows a positive trend at Alert, where the

570 favored partitioning into the particle phase is likely the main reason also for the similar trend observed during summer at Alert. Generally, observatories that started measurements only after the year 2000, detect neither significant haze nor summer trends for anthropogenic tracers, because most of the air pollution reduction happened prior to that.

575 **3.2.2 Long-term trends of MSA, Na^+, $nssK^+$, $nssCa^{2+}$, and Fe**

The other chemical constituents, MSA, $nssCa^{2+}$, and Fe are responsible for five significant trends, three in winter and two in summer. All are negative. Focusing on MSA first, interestingly, at Alert, which is the only station providing long-term continuous measurements of MSA, the concentrations of this ion

show negative trends for the haze and summer seasons between 1980 and 1999. Here, the haze

580 concentrations were related to long-range transport from more southerly latitudes, while the summer

concentrations were related to more regional Arctic ocean sources (Sharma et al., 2019). Hence reasons

for the two seasonal trends might be different and could reflect both variability in marine biological

activity as well as long-range transport patterns and removal processes on route. Within the decade 2000

to 2010, the summer trend turns significant and positive as discussed in Sharma et al. (2012).The overall

585 trend of MSA at Barrow in the haze season is negative for the period between 1998 and 2012 (not shown

for reasons discussed below). The summer trend is not significant over this period at this station.

Conversely, concentrations increased significantly at Barrow between 1999 and 2007 as discussed in

Quinn et al. (2009). Note that they used July through September for the summer months, while here we

use June through August. Thereafter, MSA concentrations drop until 2012. Calculating trends over the

590 decade 2000 to 2010 hence results in no significant trend at Barrow (p-value of 0.09 and negative slope).

This seeming contradiction is an important point, because the newly available longer time series shows

that Quinn et al. (2009) described a development over a period which happened to show an increase, but

which is not necessarily indicative of the longer-term trend. Recently the Barrow MSA time series was

extended by Moffett et al. (2020) who found an overall positive trend for July through September between

595 1998 and 2017, further underlining the argument that trend calculations over different periods can lead to

different results. This is particularly true for MSA concentrations which do not seem to follow a

monotonic long-term trend, but instead show substantial variability over time scales of five to 15 years,

which renders the Mann-Kendall approach inappropriate for further investigation. Note that these latest

data from Moffett et al. (2020) were not available when this manuscript was being prepared.

600 Na^+ can also be regarded as a natural tracer, i.e. for sea spray its dominant source. No significant trend is

observed, despite the fact that the potential for more sea spray aerosol formation due to retreating sea ice

is often referred to in the literature (Browse et al., 2014; Struthers et al., 2011). However, not observing

increased Na^+ concentrations does not mean that there is no enhanced sea spray production. Sea salt

particles are efficient CCN and hence may preferentially be removed from the atmosphere by wet

605 deposition as has been shown for other hygroscopic substances (Mahmood et al., 2019), or air mass

transport patterns can change.

NssCa^{2+}, a tracer for mineral and soil dust, shows a negative trend at Alert in summer, which is consistent with the results by Sharma et al. (2019). There is no evident explanation for the decrease, particularly given the decreasing snow cover and potential local sources of dust in late summer (see discussion of annual cycles). Sharma et al. (2019) hypothesize that long-range dust transport could play a role in this context. Iron is another dust tracer and exhibits a similar annual cycle compared to nssCa^{2+} at Alert, and the relative summer trend is also negative supporting the hypothesis of similar sources and or processes. However, the haze trend of Fe is also negative, while the nssCa^{2+} trend is not significant, indicating that, at least during winter, sources might be different for the two constituents. The shape of the long-term trend curve of Fe is similar to that of EBC, SO$_4$$^{2+}$ and NH$_4$$^+$, with the strongest decline in the mid-1990s. Given that Fe is emitted in fossil fuel combustion in addition to originating from soil dust (Matsui et al., 2018), this observation points towards an anthropogenic contribution to Fe observed at Alert.

No significant trend for nssK$^+$ was found. Given the recent increase in fire activity in the Arctic, one might have expected an emerging trend. Reasons for the absence of a trend may be that biomass burning emissions are carried further aloft in the troposphere and are not well captured at the surface, and that station records do not cover the last years, where the strongest emissions in consecutive years occurred (McCarty et al., 2021).

3.2.3 Long-term trends of optical properties

Aerosol optical properties, measured in situ at the surface, have also changed significantly at several stations during the last decades. Significant trends are observed in the scattering coefficient at Barrow and Zeppelin during winter, and at Zeppelin during summer with regard to the overall time period. At Barrow, the trend is negative during the haze period for all observations (1998-2019), and for both haze and summer seasons in the recent decade (2010-2019). Note that these overall and recent decreasing trends in scattering coefficient during winter are consistent with Collaud Coen et al. (2020b). However They did not find a significant trend in scattering coefficient during the summer months at Barrow for the most recent decade, though they used a different seasonal definitions. The scattering coefficient can be interpreted as a measure of the total measured aerosol concentrations and thus provides evidence of a decline in overall particulate matter, if hygroscopic growth or swelling is prevented through drying

635 particles before measurements, which is the case at the discussed sites. At most stations, Na^+, SO_4^{2-} and organics make up most of the mass (Petäjä et al., 2020) and are also known to scatter light rather than absorb it. A direct comparison with the aerosol mass is difficult for Barrow, because most of the chemical and optical data only overlap between 1997-2013. In particular those species which contribute significantly to the mass, such as Na^+ and SO_4^{2-}, are relevant as has been shown by Quinn et al. (2002)

640 for data between 1997 and 2000, where on the basis of linear regression Na^+ has a dominant role on aerosol scattering in winter and SO_4^{2-} in spring. Aerosol filter samples have been taken throughout the full period (1997-present), but data were not available beyond 2013 for this work. Future analysis will help answer the question whether an anthropogenic or natural source, or both, contribute to the observed decline. Conversely, at Zeppelin the overall summer and winter trends are positive for σ_{sp}, and strongly

645 driven by changes in the decade between 2000 and 2010. The *SAE* and σ_{sp} trends have been discussed in detail by Heslin-Rees et al. (2020) who found that the increased scattering signal is likely due to an increased presence of coarse mode particles. This is reflected by the significant negative trend in the *SAE* (also for both seasons). While it could be hypothesized that the retreat of sea ice is responsible, Heslin-Rees et al. (2020) found that a change in the atmospheric circulation, in which increased advection of air

650 masses from the southwest, i.e. the Atlantic, can explain the observations. The conclusion is that more sea spray aerosol is transported towards Zeppelin contributing to the increased coarse mode particles. This is not confirmed by the derived trends of sea-salt particles with the same method (Na^+, see previous section). Here trends in Na^+ were not found to be significant for the Zeppelin station. However, the physico-chemical characterization of coarse-mode aerosols at Gruvebadet (Song et al., 2021; Udisti et al.,

655 2016) showed the occurrence of several populations of anthropogenic, sea-salt and crustal particles exhibiting distinct coarse mode size-distributions, which complicates the attribution of trends in *SAE* to individual changes in emissions

The absorption coefficient, which is mostly related to the presence of combustion related aerosol and, to a much lesser extent, to mineral dust, exhibits negative trends for the overall time periods of observations

660 at Barrow and Zeppelin in winter, consistent with the wintertime trends for absorption coefficient reported in Collaud Coen et al. (2020b). These trends are in line with the decreasing EBC concentrations during the haze season, where the EBC trend is significant at Zeppelin, while at Barrow the p-value is 0.09.

Pallas shows a negative decadal trend for *SAE* between 2000 and 2009 in summer. Thereafter variability has become larger between the years and the amplitude of the signal also increased. No particular reason

665 has been found. In contrast, Collaud Coen et al. (2020b) observed both increasing and decreasing statistically significant *SAE* trends at various Arctic sites depending on the time period and season considered.
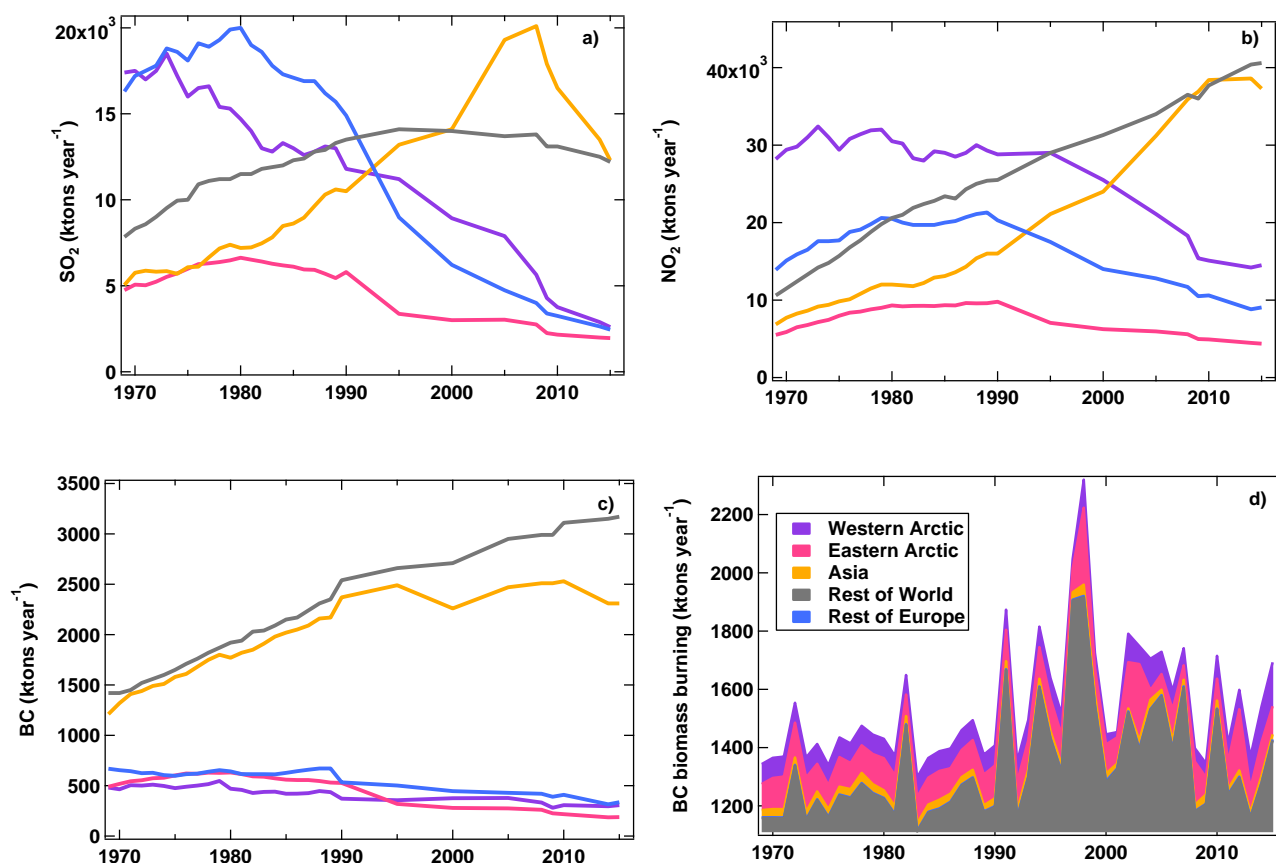
Figure 4: Anthropogenic emissions of SO₂ (a), NO₂ (b), BC (c), and BC from biomass burning (d).
Anthropogenic emissions are based on ECLIPSE v6B (Höglund-Isaksson et al., 2020) . Data from
before 1990 are based on CMIP6 (Hoesly et al., 2018; van Marle et al., 2017) scaled linearly to
match ECLIPSE v6B. Biomass burning data are from CMIP6. The Western Arctic region refers to
the following countries: USA and Canada; Eastern Arctic region: Kingdom of Denmark, Finland,
675 **Iceland, Norway, the Russian Federation, and Sweden. The grouping follows the report of the**
Arctic Monitoring and Assessment Programme on short-lived climate forcers (AMAP, 2021).

4 Discussion

The results presented above reflect the effort, which has been made to characterize the aerosol chemical
and optical properties across the Arctic over several decades, and they highlight the importance of this
long-term effort. Importantly, to obtain a pan-Arctic view, records must be taken over the same period of
time. Based on the data set behind this study, in the 1980s, on average, measurements of nine variables
(chemical and optical) were carried out each year at any station considered here. In the 1990s the number
increased to 23, and further to 36 variables in the 2000s. In the latest decade, annual measurements of 39
variables were available on average. Note that not all data might have been released for the most recent
years and hence the number is a lower estimate. We also limited the selection of aerosol variables
measured to meet the objectives of the presented study (long-term trends).

The data allow us to draw conclusions on how changes in emissions, processes and transport in the
northern hemisphere have changed the aerosol properties across the Arctic. Note that the applied
methodology here, i.e. the Mann-Kendall significance test and Theil-Sen slope estimator, relies on
monotonic change. Hence, time series which do not change monotonically are less likely to exhibit a
significant trend (see e.g. the MSA concentration at Alert). We find in total 26 significant trends over full
records, i.e. spanning more than a decade, compared to 17 significant decadal trends. The majority of
significantly declining trends in anthropogenic tracers occurred during the haze period, and was driven
by changes in the 1990. Even though the decade from 1990 to 1999 stands out, only four of the trends
calculated for this decade are significant. This result means that long-term observations that cover
multiple decades are extremely valuable and necessary to document the anthropogenic influence on the
Arctic aerosol concentration. In addition, it can be concluded that observations which started after the
1990s cannot reflect the most important decline of the anthropogenic aerosol concentration in the Arctic
in the recent past. Nevertheless, the more recently established monitoring activities will provide valuable
insight into recent trends in the years to come with respect to anthropogenic emission changes within
(Kolesar et al., 2017; Aliabadi et al., 2015; Schmale et al., 2018) and outside of the Arctic, as well as to
potential changes in natural emissions based on climate feedback mechanisms. For example, we find from

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the recent records that EBC concentrations appear to be stagnating, i.e., no significant change is observed.

705 Because trends in the latest decade are not significant, continuing observations in the future is critical to understand the present-day development. The newly established time series at additional geographical locations will help evaluate this trend in the future; particularly in light of the current black carbon reduction policies, such as those initiated by the Arctic Council countries through the Fairbanks Declaration 2017 (Arctic Council, 2017), and whether those policies will have an effect across the Arctic.

710 Anthropogenic influence has not been restricted to the haze season. Two significant negative trends in summer for EBC and Fe seem to demonstrate this, because there are corresponding negative trends during the haze period as well. With the decline of anthropogenic air pollution, natural aerosol components emerge more strongly now in summer than previously and are hence responsible for the larger part of the summertime aerosol climate effects, including both the direct and indirect effects. The vanishing

715 anthropogenic summer influence coincides with rapid and unprecedented environmental change in the high northern latitudes. September Arctic sea ice is retreating by 13.1 % per decade since 1979 compared to the 1981-2010 average (Arctic Sea Ice Minimum, 2020). Sea ice retreat creates more open ocean, which creates larger fetch for wave development (Casas-Prat and Wang, 2020) and hence more sea spray emission. DMS emissions have increased by 33 % per decade in ice-free water since 1998 (Galí et al.,

720 2019). Boreal forest fires are also becoming more frequent and severe (Rogers et al., 2020). For example, BC emissions have roughly doubled from 2010 to 2020 from wildfires north of 60°N (McCarty et al., 2021). Warmer temperatures, which are the main cause of enhanced fire activity, also cause enhanced biogenic emissions, such as isoprene (a secondary organic aerosol precursor originating from vegetation (Arneth et al., 2016)). These and other ongoing and anticipated environmental changes (e.g., changed

725 wind and air mass transport patterns) will have an effect on the natural contribution to aerosol properties and concentrations (Schmale et al., 2021). However, the trend analyses of natural aerosol tracers used in this study do not show strong or unambiguous evidence for such changes yet. There can be several reasons. With respect to SO_4^{2-}, there could be compensating effects of reduced SO_2 emissions and increased DMS emissions. For example, a high interannual variability in natural sources hinders the

730 detection of long-term trends. Additionally the complex relationships between natural emissions, atmospheric transport, chemical reactions, particle removal processes and observed airborne aerosol

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concentrations, means that an increase in emissions does not automatically translate to enhanced concentrations of corresponding end products or the species itself. Furthermore, our analyses are conducted with the Mann-Kendall Theil-Sen Slope test only. Other methods might yield different results.

735 For example, increased wave breaking on a more ice-free ocean should lead to higher Na^+ concentrations. However, none of the four stations that report Na^+ show a significant trend. This could be due to the inherently large variability in the Na^+ signal, which requires hence longer data sets to extract a monotonic signal with the Mann-Kendall Theil-Sen methodology. For Alert, if the Na^+ concentration for the first five years of the record and the last five years are averaged, there is a 20 % increase in Na^+. It could also

740 be due to the preferential wet scavenging of Na^+ due to its high hygroscopicity. In addition to the chemical signature, which can document changes in sea spray aerosol, size distribution data can provide indications and improve such analysis. Using the *SAE*, Heslin-Rees et al. (2020) showed an increase in coarse mode particles at Zeppelin, which is likely due to an increase of observations in sea spray emissions and atmospheric transport changes. At Zeppelin, the source is the mid-latitude Atlantic rather than the

745 increasingly ice-free Arctic Ocean. While at Alert, the *SAE* trend was not found to be significant, it nevertheless has a low p-value of 0.09 and a negative slope, which is consistent with the decrease in Na^+. Concerning marine biogenic emissions, observations of MSA concentrations provide mixed results. While at Barrow the MSA time series from 1997 to 2017 shows an increase (Moffett et al., 2020), measurements at Alert do not indicate a significant upward trend between 1981 and 2018, but rather a

750 negative trend between 1981 and 1999, and a positive trend in the decade 2000 to 2009 in MSA. Becagli et al. (2019) find a positive trend in the most recent decade at Gruvebadet on Svalbard and a negative one at Thule, Greenland. Our analyses in this study show similar trends for the two latter stations, but no significance. In a modeling study (Browse et al., 2014) found that enhanced marine biogenic emissions do contribute to enhanced aerosol concentrations, but they also cause increased wet deposition, such that

755 higher concentrations of marine biogenic tracers might not be measured at the observatories.

Regarding the potential increase in biomass burning aerosol, there is no observatory indicating a significant positive trend in relevant components (e.g., EBC, absorption coefficient, $nssK^+$). The absence of trends might be due to atmospheric transport patterns, meaning observatories might not be located in the regions that are consistently influenced by enhanced biomass burning. Importantly, biomass burning

760 emissions are also ejected higher aloft into the atmosphere, meaning surface stations might not capture these layers due to down-limited mixing in the stratified polar atmosphere. It could also partly be owing to the lack of observations of more specific tracers for biomass burning.

Less snow cover in summer might lead to the hypothesis that enhanced local soil dust emissions can contribute to the aerosol burden at Arctic sites. While soil dust tracer data were available for four stations,

765 none of them showed a significant increase for the overall period, neither during winter nor summer. In contrast, summer nssCa^{2+} declined at Alert, indicating that potentially long-range transport is the more important factor at this station.

The current absence of significant trends in natural aerosol components might be a result of the choice of aerosol tracers measured at the observatories. While MSA is a unique tracer for marine biogenic

770 emissions, and the Na$^+$ signal is governed by sea salt (Udisti et al., 2016), there is a lack of unique tracers for dust and terrestrial biogenic emissions. Those need to be identified using statistical tools or models. For the latter, observations and speciation of organic aerosol with spatial coverage in the Arctic, and in particular secondary organic aerosol, is desperately needed for better source apportionment (Petäjä et al., 2020; Frossard et al., 2011). Analyses based on short-term efforts clearly show the potential of various

775 analyses techniques (mass spectrometry or Fourier transform infrared spectral analysis) to isolate signatures of anthropogenic versus natural organic aerosol contribution (Fu et al., 2009; Kawamura et al., 2010; Leaitch et al., 2018; Willis et al., 2017). Isotopic analyses of carbon, nitrogen and sulfur can also provide more details on sources. This has been demonstrated by carbon isotope analyses on atmospheric and deposited aerosols (Rodríguez et al., 2020; Winiger et al., 2019). Nitrogen isotopic analysis has been

780 used to elucidate the NO$_x$ budget in the Arctic, however the study was more focused (Dall´Osto et al., 2018; Schmale and Baccarini, 2021) on trace gas chemistry rather than aerosol source apportionment (Morin et al., 2008). Sulfur isotopic analysis has been applied a limited number of times for shorter term data sets in the Arctic to distinguish anthropogenic, biogenic and sea salt contributions (Ghahremaninezhad et al., 2016; Seguin et al., 2014). This can also be achieved by online mass

785 spectrometry as has been done at Villum (Nielsen et al., 2019). Also, microphysical properties such as particle size distributions can reveal changes that bulk aerosol composition cannot. For example, chemical analyses are mostly mass based, and hence changes in particles with smaller diameters are not well

captured by such analysis. Those however have already been shown to reflect enhanced contribution of marine biogenic emissions to new particle formation and growth (Dall´Osto et al., 2017; Beck et al.,
790 2020). In addition, optical properties can reflect changes in particle size distributions, for example the SAE at Zeppelin, where the aerosol population seems to evolve in a way that larger particles dominate. While this is attributed to more frequent observations of sea spray from the Atlantic (Heslin-Rees et al., 2020), the chemical measurements of Na^+ at Zeppelin did not capture this change at the 95th % significance level. However, a combination of chemical and optical measurements can shed more light
795 on particle origin and driving factors of climate-relevant optical aerosol properties (Quinn et al., 2002). Despite the large data set discussed here, the chemical and optical observations only coincide marginally. Hence, long-term trends of their co-evolution are not investigated, but such a study would provide very useful insights, particularly adding detailed size distribution information.

5 Conclusions and outlook

800 We presented and analyzed more than a cumulative 1000 years of measurements of aerosol chemical and optical properties recorded at ten surface observatories across the Arctic between 1967 and 2019. Variables include EBC, SO_4^{2+}, NO_3^-, $NH4^+$, MSA, Na^+, $nssCa^{2+}$, $nssK^+$, Fe, scattering and absorption coefficients, *SSA* and *SAE* (Table 2).

Annual cycles of all variables clearly show that the wintertime is still dominated by anthropogenic haze
805 across the whole Arctic, while the summer is more strongly influenced by natural aerosol components. Significantly decreasing long-term trends of anthropogenic tracers provide evidence of the decline of human emissions further south which is observed during the haze season at Arctic sites. The strongest decline in SO_2 and BC emissions took place in the 1990s and is visible in large decreases of particulate sulfate and EBC in the North American and European Arctic. Available data from the Eastern Arctic
810 covers only the most recent decades and hence could not capture the change. In the recent past, EBC concentrations seemed to stagnate or slightly increase (not significantly). We calculated that concentration increases of at least 5 % per year are currently needed to detect a significant trend per decade, due to the interannual variability of EBC concentrations. Lower rates of change are detectable

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only over longer periods of time. Future data will show, whether recent efforts by the Arctic Council

815 members in reducing emissions, particularly of black carbon, are effective (Arctic Council, 2017).

Among all long-term trends, 29 % are significant in the haze season where 13 out of 16 show a decline. The two positive trends include NO_3^- at Alert and scattering coefficient at Zeppelin. Despite NO_x emissions decrease at mid-latitudes, particulate nitrate might increase at Alert due to atmospheric chemical processes favoring nitrate partitioning into particles due to the less acidic aerosol (less sulfate).

820 At Zeppelin the enhanced scattering coefficient comes along with a decrease in the *SAE*, indicating a stronger contribution of larger particles, i.e. sea spray from the mid-latitude Atlantic (Heslin-Rees et al., 2020).

For the summer period, no uniform picture of trends has emerged. Twenty-one percent of trends, i.e. eleven out of 57, are significant, whereby five are positive and six are negative. Negative trends include

825 not only anthropogenic tracers such as EBC at Kevo, but also natural indicators such as MSA and $nssCa^{2+}$ at Alert. Positive trends are observed for SO_4^{2+} at Zeppelin and Gruvebadet. Those could be related to enhanced marine biogenic emissions, but analogous significant trends in MSA were not found. Hence, no clear evidence of a significant change in the natural aerosol contribution can be seen yet, despite the evident sea ice decline, increased DMS, and increased fire and terrestrial vegetation emissions (Arneth et

830 al., 2016; Galí et al., 2019; Arctic Sea Ice Minimum, 2020; Rogers et al., 2020). This points towards four main action items for monitoring Arctic aerosol.

- First, long-term observations targeted at natural aerosol tracers are needed. Increased natural particle and precursor emissions might produce signal enhancements, which are small compared to the inherent signal variability. Hence, new trends might not yet be visible but might emerge in

835 the years to come. They are important indicators of feedback mechanisms in Arctic amplification.

- Second, the location of an observatory matters for which trends might emerge as significant or not. Not every observatory will exhibit all types of natural aerosol changes clearly. For example, Becagli et al. (2019) find a difference in MSA trends between Gruvebadet and Thule, likely due to different evolution of marine ecosystems. A natural change hot spot map of the Arctic combined

840 with simulations of atmospheric transport and aerosol processes can suggest areas that require

new measurement efforts. Moreover, most stations are located near sea level, and only Summit provides valuable insights into processes further aloft including in the free troposphere.

- Third, an assessment of whether the currently monitored aerosol properties are appropriate to capture changes in natural aerosol compounds is needed. Source apportionment based on

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  speciated organic aerosol components is currently lacking except for a few short-term case studies (Petäjä et al., 2020; Frossard et al., 2011). Those together with isotopic analyses of carbon, nitrogen and sulfur can shed light into natural aerosol changes. Microphysical properties such as the size distribution and inherent optical characteristics such as the single scattering albedo and scattering Ångström exponent can further reveal changes in the number, size and composition and

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  thus origin of particles, which chemical analyses alone, especially of bulk aerosols, cannot.

- Fourth, standard operation procedures for the measurement of particle properties are needed such that they can be compared (e.g. same cut-off size on the inlets).

In summary, aerosol properties are an important tool to analyze changes in anthropogenic and natural atmospheric chemical influences on the Arctic. Tremendous monitoring efforts have already been

855 conducted, which have helped to understand the implications for past, present and future Arctic climate change. The decision to start regular monitoring of key anthropogenic components in the early 1990's - or even before – turned out to be important and timely, because it enabled us to capture a remarkable change in the concentrations of anthropogenic climate-forcing agents in the Arctic lower atmosphere that occurred in the late twentieth century. Establishing specifically targeted observations for the ongoing and

860 future change of natural aerosols are essential to develop improved scenarios of their climatic and biogeochemical effects on the Arctic. Continuation of the monitoring programs targeting changes in variables during the haze season linked to anthropogenic emissions at mid-latitudes is a prerequisite to give further evidence to the declining trends determined today and for their quantification feeding global climate and earth system models.

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the aegis of NSF and NOAA, and aethalometer measurements at Tiksi occurred through NOAA

895 cooperation with Roshydromet that ended in 2018.

Data availability

Most data are publicly available on ebas.nilu.no or else on request via the personal communication

contacts listed in Table 1. All these contacts are co-authors and in charge of observatory data, which needs

900 to undergo specific control before it can be posted publicly. This publication partly precedes open access

data publication. All data will be available via ebas.nilu.no in the future.

Author contributions

LU, PKQ, EA, AM, JBP, SS, SD, KE processed and analyzed data from the observatories. JS, SS, and

905 SD calculated annual cycles and seasonal trends. KVS provided emissions data. JS wrote the manuscript.

All authors contributed to interpretation and commented on the manuscript.

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