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Characterization of distinct Arctic aerosol accumulation modes and their sources

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ABSTRACT

In this work we use cluster analysis of long term particle size distribution data to expand an array of different shorter term atmospheric measurements, thereby gaining insights into longer term patterns and properties of Arctic aerosol. Measurements of aerosol number size distributions (9–915 nm) were conducted at Villum Research Station (VRS), Station Nord in North Greenland during a 5 year record (2012–2016). Alongside this, measurements of aerosol composition, meteorological parameters, gaseous compounds and cloud condensation nuclei (CCN) activity were performed during different shorter occasions. K-means clustering analysis of particle number size distributions on daily basis identified several clusters. Clusters of accumulation mode aerosols (main size modes > 100 nm) accounted for 56% of the total aerosol during the sampling period (89–91% during February–April, 1–3% during June–August). By association to chemical composition, cloud condensation nuclei properties, and meteorological variables, three typical accumulation mode aerosol clusters were identified: *Haze* (32% of the time), *Bimodal* (14%) and *Aged* (6%). In brief: (1) *Haze* accumulation mode aerosol shows a single mode at 150 nm, peaking in February–April, with highest loadings of sulfate and black carbon concentrations. (2) Accumulation mode *Bimodal* aerosol shows two modes, at 38 nm and 150 nm, peaking in June–August, with the highest ratio of organics to sulfate concentrations. (3) *Aged* accumulation mode aerosol shows a single mode at 213 nm, peaking in September–October and is associated with cloudy and humid weather conditions during autumn. The three aerosol clusters were considered alongside CCN concentrations. We suggest that organic compounds, that are likely marine biogenic in nature, greatly influence the *Bimodal* cluster and contribute significantly to its CCN activity. This stresses the importance of better characterizing the marine ecosystem and the aerosol-mediated climate effects in the Arctic.

1. Introduction

The Arctic, one of the most sensitive regions to climate change, is warming at a rate twice as rapid as the global average (AMAP, 2011). Global climate models attribute large effects on temperature to Arctic aerosols (Chylek et al., 2016; Yang et al., 2014; Bellouin et al., 2011). Aerosol particles may perturb the radiation balance of the Arctic environment in numerous ways (Carslaw et al., 2013; Ramanathan et al., 2001). Overall, different aerosol chemical species, as well as particle size and abundance, may determine the magnitude of the aerosol induced direct climate forcing. Furthermore, aerosols also constitute the seeds upon which cloud droplets form (Ramanathan et al., 2001), resulting in an indirect climate effect. Improved understanding of the spatial and temporal variability of the microphysical properties of Arctic aerosols is required, in order to determine the magnitude and direction of future climate change in this important region.

Arctic aerosols have been shown to be highly variable. Broadly, over the Arctic region aerosol mass and number size distribution properties are a strong function of season. This seasonality is repeated on a yearly basis (Freud et al., 2017; Nguyen et al., 2016; Tunved et al., 2013). It is well established that the Arctic atmosphere in winter and spring is more heavily impacted by transport of air pollution from lower latitudes compared to in summer (Heidam et al., 2004; Law and Stohl, 2007). The continent-derived winter and spring aerosols, known as Arctic haze, reach their maximum number concentration during late spring, approximately in April. The transition from Arctic haze conditions, to

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the lower aerosol mass loadings in the summer period, is driven by increased wet scavenging due to increasing temperatures over a period of about two weeks (Browse et al., 2012; Croft et al., 2016; Engvall et al., 2008). It is becoming increasingly evident that biogenic ultrafine (< 100 nm) (including freshly nucleated) particles dominate ambient aerosol numbers in Arctic areas during summer (Dall'Osto et al., 2017a; Dall'Osto et al., 2018). Occasionally long-range pollution transport events also occur during summer (Iziomon et al., 2006; O'Neill et al., 2008). Towards the end of summer, sunlight intensity decreases. Despite low concentrations of large particles and thus small condensation and coagulation sinks that would facilitate new particle formation (Dal Maso et al., 2002), the production of nucleating vapors is too slow to create new particle formation events.

Aerosol number size distributions from multi-year measurements have been reported from different Arctic research stations: Zeppelin (Tunved et al., 2013), Tiksi (Asmi et al., 2016), Alert (Croft et al., 2016), Barrow (Lathem et al., 2013) and Villum Research Station (VRS), Station Nord (Nguyen et al., 2016). All studies broadly converge in a similar scenario: The haze period characterized by a dominating accumulation mode (> 100 nm) aerosol (March–May), is followed by the sunlit summer with high concentration of ultrafine particles (June–August). The remaining year is characterized by low concentration of accumulation mode particles as well as a negligible abundance of ultrafine particles (September–February).

Despite this, information on different types of accumulation mode aerosols is scarce. At Zeppelin, Svalbard, Tunved et al. (2013) reported the occurrence of an accumulation mode with a typical mode mean particle diameter (Dp) of 161–185 nm during winter months (November to March) and 130–163 nm during April to October. Nguyen et al. (2016) reported that the larger Dp accumulation mode persists further into the summer at VRS, Greenland, than at Zeppelin, with a typical mode mean diameter Dp of 167–179 nm for months November to May and 107–119 nm for months June to September. A recent intercomparison of particle number size distributions from several Arctic stations by Freud et al. (2017) suggests variations between the different stations throughout the year. The most prominent differences are observed between the stations at Barrow and Zeppelin. Barrow features a wider accumulation mode, with higher concentrations and smaller Dp than Zeppelin in months September to May.

Considerable attention has been given to the role of anthropogenic and biomass burning particles as warming agents in the Arctic (UNEP, 2011). Black carbon (BC) contributes to Arctic warming, yet sources of Arctic BC and their geographic origins remain uncertain (Xu et al., 2017). BC particles and especially aged BC particles affect the radiation budget directly by scattering and absorbing incoming solar radiation (Massling et al., 2015). Sensitivity simulations (Xu et al., 2017) suggest that anthropogenic emissions in eastern and southern Asia have the largest effect on the Arctic BC column burden in both spring (56%) and annually (37%). Clean Arctic marine air masses are expected to be associated with BC concentrations smaller than about 20 ng m^{-3} (Gogoi et al., 2016; Cavalli et al., 2004). By investigating the relationship between aerosol clusters and black carbon concentration, we have previously demonstrated that such pristine clean conditions (eBC < 18 ng m^{-3}) co-occur with ultrafine dominated aerosols (Dall'Osto et al., 2018). In this work we elucidate the eBC loading patterns of different Arctic accumulation mode aerosols, that are largely association with non-pristine conditions.

To the best of our knowledge, no long-term studies on Arctic aerosol have identified several distinctively different accumulation mode aerosols. In this study we provide evidence that multiple accumulation mode aerosol clusters exist in the Greenlandic high Arctic, and that these are present at different proportions throughout the year. These accumulation mode aerosol clusters are characterized both physically and chemically, and statistically significant differences are highlighted. We show that especially during summer, accumulation mode aerosol with strong biogenic influence has high cloud condensation nuclei (CCN) activity. In conclusion, the aim of the present paper is to improve the understanding of Arctic accumulation mode aerosols, and to describe them in tandem with meteorological parameters, gaseous concentrations, aerosol chemical species and cloud condensation nuclei properties.

2. Methodology

2.1. Location

All the data presented in this work was recorded at the Villum Research Station, Station Nord, Greenland, located at 81° 36′ N, 16° 40' W. The station is situated in the most north-eastern part of Greenland, on the coast of the Fram Strait. The sampling took place about 2 km south-west of the main facilities of the Station Nord military camp in two different sampling stations. Measurements were relocated in summer 2015 from the original hut called "Flygers hut" to the new air observatory, 300 m west of "Flygers hut". The sampling locations are upwind from the station the vast majority of the time. Detailed descriptions of the site and analysis of predominant wind directions are available elsewhere (Nguyen et al., 2013, 2016).

2.2. Scanning Mobility Particle Sizer (SMPS)

We analyzed continuous Scanning Mobility Particle Sizer (SMPS) data collected in the period 2012–2016 in the particle size range of 9–915 nm in diameter with a 5 min time resolution. The sampling setup has been described in detail by Nguyen et al. (2016). Our measurements differ in the way that since summer 2015 the SMPS has been situated in the newly constructed air observatory measurement hut, described above. The instrument is custom-built with a Vienna-type medium column, similar to the SMPS instruments described in Wiedensohler et al. (2012). Our SMPS used either a condensation particle counter (CPC) TSI model 3010 or TSI model 3772. To ensure correct functioning, volumetric flow rates, temperatures and relative humidity (RH) of the aerosol and sheath flows were monitored, as well as inlet ambient pressure. No additional aerosol drying was performed, as the transition from the low ambient temperatures outside of the huts (−45 to +15 °C, yearly average −15 °C) to the heated inside (20 °C) generally provides sufficient decrease of RH. The SMPS sample flow RH only exceeded 35% during 0.35% of all measurements. An algorithm according to Pfeifer et al. (2014) was used to invert the SMPS measurements. The resulting particle number size distribution series were quality controlled to ensure correct functioning of the instrument and absence of influence from local pollution from near-by vehicles or by the military camp. During this quality control 12.4% of the SMPS data was excluded from further analysis.

2.3. Concentrations of gaseous pollutants

O_3 and NO_x were measured using gas analyzers (API photometric O_3 analyzer (M400), API chemiluminescence NOx analyzer (M200AU)). NO_x data was available for most of 2012 with half hour time resolution, whereas O_3 data was available with half hour time resolution throughout the period of 2012–2016. O_3 data from this measurement setup has earlier been published in Skov et al. (2004) and is largely quality assured and controlled in accordance with standard EN14625:2012. The standard assurance procedure is not complied in the way, that calibration is not performed as often as required, but when practically possible, about once every half year.

2.4. Particulate matter properties

In the period from May 2011 to August 2013, observations of the aerosol light absorption coefficient were conducted using the Multiangle Absorption Photometer (MAAP, Model 5012 Thermo

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