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Abiotic and biotic sources influencing spring new particle formation in North East Greenland

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ABSTRACT

In order to improve our ability to predict cloud properties, radiative balance and climate, it is crucial to understand the mechanisms that trigger the formation of new particles and their growth to activation sizes. Using an array of real time aerosol measurements, we report a categorization of the aerosol population taken at Villum Research Station, Station Nord (VRS) in North Greenland during a period of 88 days (February–May 2015). A number of New Particle Formation (NPF) events were detected and are herein discussed. Air mass back trajectories analysis plotted over snow-sea ice satellite maps allowed us to correlate early spring (April) NPF events with air masses travelling mainly over snow on land and sea ice, whereas late spring (May) NPF events were associated with air masses that have passed mainly over sea ice regions. Concomitant aerosol mass spectrometry analysis suggests methanesulfonic acid (MSA) and molecular iodine (I_2) may be involved in the NPF mechanisms. The source of MSA was attributed to open leads within the sea ice. By contrast, iodine was associated with air masses over snow on land and over sea ice, suggesting both abiotic and biotic sources. Measurements of nucleating particle composition as well as gas-phase species are needed to improve our understanding of the links between emissions, aerosols, cloud and climate in the Arctic; therefore our ability to model such processes.

1. Introduction

In the Arctic, clouds are the dominant factor in the control of the incoming and outgoing energy balance at the Earth's surface (Intrieri et al., 2002). Aerosols act as cloud condensation nuclei (CCN), upon which cloud droplets are formed (Ramanathan et al., 2001). It is long established that aerosols and their cloud seeding role, the so-called indirect aerosol effect, have a substantial impact on the Earth's albedo and climate (Charlson et al., 1987). However, what regulates the number of aerosol particles and their capability to act as CCN still is poorly known and constitutes one of the largest sources of uncertainty in climate understanding and modelling (Carslaw et al., 2013). In the Arctic climate system the picture is even more complicated because there are many dynamic inter-connections between processes occurring on partly snow-covered land and in sea ice and the ocean. All of these environmental components, and their changes over time (Serreze and Stroeve, 2015) influence not only the surface albedo but also the composition and radiative properties of the atmosphere.

New particle formation (NPF), if accompanied by stabilization and growth, increases aerosol number concentrations in the atmosphere (Kulmala et al., 2004; Spracklen et al., 2006), providing a pool of secondary aerosols with potential to increase CCN population. Background particle concentrations in the summertime Arctic are typically very low ($\sim 100\,cm^{-3}$). Cloud properties in this region are therefore highly sensitive to the mechanisms by which new particles form and grow (Merikanto et al., 2009). A better understanding of the physical and chemical processes leading to a higher nucleation potential requires detailed knowledge about aerosol precursor emissions from the different types of surfaces in the Arctic environment. In the Arctic lower troposphere, nucleation and Aitken mode particles dominate the size distribution during summer, in contrast to the accumulation mode-dominated winter and spring (Ström et al., 2011; Heintzenberg and

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Leck, 2012; Tunved et al., 2013; Leaitch et al., 2013). At very high latitudes in sea ice-covered regions or near the marginal ice zone, it has also been suggested that small primary particles may arise from bubble bursting (Leck and Bigg, 1999). Particle nucleation has been observed in this region as well (Heintzenberg et al., 2015; Wiedensohler et al., 1996). Recently, Dall'Osto et al. (2018) categorized the aerosol population via cluster analysis of aerosol number size distributions taken at Villum Research Station, Station Nord (VRS) in North Greenland during a 7 year record (2010–2016). Most NPF events (annually 9% of the time, up to 39% during summer months) were associated with air mass back trajectories passing over open water and melting sea ice regions, suggesting these regions as the main source. The origin of these ultrafine particles seems to be marine biological activities within the open leads in the pack ice and/or along the melting marginal sea ice zone (MIZ). The results from the period 2010–2016 at VRS are in line with recent results from the period 2000–2010 at the Zeppelin Mountain Station (Dall'Osto et al., 2017a). However, it is important to stress that the NPF source regions and corresponding precursor components are still a topic of intense research, and not only include emissions of precursor gases associated with biological communities on or near sea ice margins (Dall'Osto et al., 2017a; b; Levasseur, 2013), but also seabird colonies (Croft et al., 2016; Weber et al., 1998) and intertidal zones (O'Dowd et al., 2002; Allan et al., 2015; Sipilä et al., 2016). Overall, there is an increasing number of studies reporting observations of secondary organic components of the Arctic aerosol (Leaitch et al., 2018) from emissions of biogenic volatile organic compounds (BVOCs; e.g., isoprene and terpenes), including oxygenated VOCs (OVOCs; Mungall et al., 2017) and trimethylamines (Köllner et al., 2017). Additionally, it is important to stress that nucleation events in Greenland have been also associated with air masses that travelled over snow regions, and mechanisms based on gas precursors released from the snowpack were suggested (Ziemba et al., 2010).

Traditionally, experiments conducted in the Arctic have mainly focused on the Arctic Haze phenomenon during late winter and spring and on NPF events occurring mainly only during summer (Tunved et al., 2013; Freud et al., 2017). Here, for the first time we focus our attention on springtime NPF events in coastal North Greenland: we identify these events by aerosol size distributions, characterize key chemical components, and relate them to air mass trajectories over categorized surfaces.

2. Experimental measurements

Aerosol measurements were undertaken at VRS, 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 shifted 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 (97.3%). Detailed descriptions of the site and analysis of predominant wind directions are available in Nguyen et al. (2016) and Nguyen et al. (2013). VRS at Station Nord is a unique Arctic station located close to sea level at the ice stream from the Arctic Ocean. VRS is furthermore always located north of the polar Vortex representing the conditions of the high Arctic throughout the whole year.

Detailed information about scanning mobility particle sizer (SMPS) measurements can be found elsewhere (Nguyen et al., 2016). Measurement of particle number size distributions at Station Nord was initiated in July 2010 using a TROPOS-type Mobility Particle Size Spectrometer as described in Wiedensohler et al. (1996). Soot particle aerosol mass spectrometer (Aerodyne, SP-AMS) was deployed for four months over the period February–June 2015. The instrument is described in details elsewhere. The SP2 provides rBC mass loadings, particle number concentrations, and size distribution measurements, though these measurements are constrained by single particle detection limits of ~0.7 fg/particle (Schwarz et al., 2010). The SP-AMS is described in detail elsewhere (Onasch et al., 2012). In this study, high-resolution (HR) mass concentrations of SO_4^{2-}, NO_3^-, NH_4^+, organics, Cl, rBC (reflactive black carbon), and signals for I_2 and MSA are obtained from the SP-AMS. The SP-AMS was operated in 2 min laser off and 2 min laser on in V-mode and alternated between the mass spectrum mode and the particle time-of-flight (pToF) to obtain PM_1 (particulate matter with diameter below 1 μm) concentration and particle size distribution, respectively. The data was analyzed with the standard AMS Igor Pro-based (version 6.35 Wavemetrics, Inc) software SQUIRREL (version 1.57G) and PIKA (version 1.16H). The analysis was treated with general accepted principles described in DeCarlo et al. (2006), Jimenez et al. (2003) and Onasch et al. (2012).

Using the BADC Trajectory Service (British Atmospheric Data Centre Trajectory Service), 5 day back trajectories arriving at VRS station were calculated arriving at 100m altitude (hourly resolution). The length of the back-trajectory calculation is chosen as a balance between the typical lifetime of the aerosols in the Arctic troposphere, which is up to two weeks (shorter in summer and longer in winter/spring) for the accumulation-mode particles, and the increasing uncertainty in the calculation the further back in time it goes. Simple calculations were also made for each of the 5-day back trajectories using daily Arctic maps of gridded sea ice information. For each of the positions alone each of the trajectories the sea ice information was logged into a file from which the exposure to sea ice of all the air masses arriving at SN could be calculated. The Polar Stereographic map of the Northern Hemisphere classified each of 1024x1024 24 km grid cells as land, sea, ice or snow ice, and from this, the percentage of time each clustered back trajectory spent over each type could be calculated. The snow and ice coverage values were produced by the NOAA/NESDIS Interactive Multisensor Snow and Ice Mapping System (IMS) developed under the direction of the Interactive Processing Branch (IPB) of the Satellite Services Division (SSD). A similar calculation was repeated but using daily maps of sea ice percentage concentration measured on a 12.5 km grid. These Arctic polar sterographic maps of 12.5 km resolution contained sea ice concentration, available since 1992. The percentages assigned from these maps to each trajectory step allow a 'spectrum' of sea ice concentration of 5% width from 0 to 100% to be calculated for each of the trajectory clusters.

3. Results and discussion

3.1. NPF overview

K-means cluster analysis (Dall'Osto et al., 2018a; Lange et al., 2018) of particle number size distributions using 33,678 hourly distributions collected over 7 years (2010–2016, 55% data coverage during the period, see Nguyen et al., 2016) was carried out. Briefly, the K-means method aims to minimize the sum of squared distances between all points and the cluster centre. In order to choose the optimum number of clusters the Dunn-Index (DI) was used. The DI aims to identify dense and well-separated clusters. DI is defined as the ratio between the minimal intercluster distance to maximal intracluster distance. Consequently, we wanted to find the clustering which maximizes this index. The use of cluster analysis was justified in this work using a Cluster Tendency test (Beddows et al., 2009; Dall'Osto et al., 2011). Based on such cluster analysis, we identified eight categories of aerosol number size distributions (Dall'Osto et al., 2018a), and subsequently overlapped them with the time period where the SP-AMS was operating between 21st February 2015 and 23rd May 2015 (88 days in total). During this winter-spring period, 69 days (79%) were classified as Arctic Haze, carrying the highest total number concentration in the accumulation mode (> 100 nm, Fig. 1), peaking at about 170 nm and unimodal in appearance (Lange et al., 2018). Only 12 days (14%) were classified as "pristine" Arctic conditions (Fig. 1), characterized by very low particle