Apportioning aerosol natural and anthropogenic sources thorough simultaneous aerosol size distributions and chemical composition in the European high Arctic

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1   **ABSTRACT**

2   Understanding aerosol size distributions is crucial to our ability to predict aerosol number

3   concentrations. When of favourable size and composition, both long range transported

4   particles as well as locally formed ones may serve as Cloud Condensation Nuclei (CCN);

5   small changes may have a large impact on the low CCN concentrations currently

6   characteristic of the Arctic environment. Here, we present a cluster analysis of particle size

7   distributions (PSD, size range 8-500nm) simultaneously collected from three high Arctic

8   sites across Europe during a three year period (2013-2015). Two sites are located in the

9   Svalbard archipelago: Zeppelin research station (474m above ground), and the nearby

10 Gruvebadet Observatory (about 2Km distance from Zepplelin, 67m above ground). The

11 third site (Villum Research Station – Station Nord, 30m above ground) is 600 km to the

12 west-northwest of Zeppelin, at the tip of north-eastern Greenland. An inter-site comparison

13 exercise is carried out for the first time including the Gruvebadet site. K-means analysis

14 provided eight specific aerosol categories, further combined into broad PSD with similar

15 characteristics, namely: pristine low concentrations (12-14%), new particle formation (16-

16 32%), Aitken (21-35%) and accumulation (20-50%). Confined for longer time periods by

17 consolidated pack sea ice regions, the Greenland site shows PSD with lower ultrafine

18 mode aerosol concentrations during summer, but higher accumulation mode aerosol

19 concentrations during winter relative to the Svalbard sites. By association with chemical

20 composition and Cloud Condensation Nuclei properties, further conclusions can be

21 derived. Three distinct types of accumulation mode aerosol are observed during winter

22 months, associated with sea spray (largest detectable sizes), Arctic haze (main mode at

23 150nm) and aged accumulation mode (main mode at 220nm) aerosols. In contrast, locally

24 produced and most likely of marine biogenic origin particles exhibit size distributions

25 dominated by the nucleation and Atiken mode aerosol during summer months. The

1 obtained data and analysis set now the stage for future studies; including apportioning the

2 relative contribution of primary and secondary aerosol formation processes to the aerosol

3 size distribution in high Arctic, and elucidating anthropogenic aerosol dynamics, transport

4 and removal processes across the Greenland sea. In a region of enormous importance for

5 future climate on Earth, it is imperative to continue strengthening international scientific

6 cooperation, in order to address important research questions on scales beyond singular

7 station or measurement events.

9 **1. INTRODUCTION**

10 The Arctic is a sensitive region affected by perturbations of the radiation budget, with

11 complex feedback mechanisms resulting in a temperature increasing more than twice the

12 global average since the 1980s (so-called "Arctic amplification", Cohen et al., 2014, Pithan

13 and Mauritsen, 2014). Aerosols are able to perturb the radiation balance of the Arctic

14 environment in numerous ways (Carslaw et al., 2013). The contribution by aerosols to

15 radiative forcing is considered a very important parameter, although still highly uncertain in

16 a recent climate assessment (IPCC, 2014). In order to improve our knowledge in

17 estimating direct and indirect climate effects, a better knowledge of the aerosols is an

18 essential requisite, including their properties and seasonal variability, their sources, and

19 the associated atmospheric reactions and transport processes. One of the main basic

20 properties to characterize an aerosol is the size distribution. Atmospheric aerosol particles

21 span over several orders of magnitude in diameter (Dp), from a few nanometer to

22 hundreds of micrometers. Small particles, in particular nucleation mode (typically with Dp

23 <10 nm) and Aitken mode (10 nm<Dp<100 nm) particles contribute little to total particulate

24 mass in background air; however, they contribute significantly to surface area and

1   dominate particle number concentration (Dall´Osto et al., 2011). Measurements of Arctic

2   aerosols have shown a strong annual cycle. The first full year of aerosol size distribution

3   and chemical composition Arctic measurements was conducted at the Zeppelin station on

4   Svalbard (Strom et al. 2003), showing a very strong seasonal dependence of the number

5   mode particle size. Tunved et al. (2013) subsequently reported a qualitative and

6   quantitative assessment of more than 10 yr of aerosol number size distribution data

7   observed in the Arctic environment (Mt. Zeppelin, Svalbard); reporting that seasonal

8   variation seems to be controlled by both dominant sources as well as meteorological

9   conditions. This can be broadly summarised in three distinctly different periods:

10   accumulation mode aerosol during the haze period (March–May), followed by a high

11   concentration of small particles locally formed (June–August), leaving the rest of the year

12   with a low concentration of accumulation mode particles and negligible abundance of

13   ultrafine particles (September–February). Additional results from multi-year measurements

14   reported similar conclusions using aerosol number size distributions collected at Tiksi

15   (Asmi et al., 2016), Alert (Croft et al., 2016), Barrow (Lathem et al., 2013) and Villum

16   Research Station - Station Nord (Nguyen et al., 2016).

17   Currently, the Arctic haze is not well represented within atmospheric models, mainly due

18   to inadequate representation of scavenging processes different transport mechanisms and

19   underestimation and an unknown number of aerosol sources. As regards natural aerosol

20   sources, they have been emphasized to be much more important than transport from

21   continental sources. Recently, the aerosol population was categorised via cluster analysis

22   of aerosol size distributions taken at Mt Zeppelin (Svalbard, Dall´Osto et al., 2017a) during

23   an 11 year record (2000-2010) and at Station Nord (Greenland, Dall´Osto et al., 2018b)

24   during a 7 year period (2010-2016). Air mass trajectory analysis linked these frequent

25   nucleation events to biogenic precursors released by open water and melting sea ice

1 regions. Both studies reported a striking negative correlation (r = -0.89 and -0.75,

2 respectively) between sea ice extent and nucleation events. Given the likely decrease in

3 future sea ice extend in the Arctic, it is likely that the impact of natural ultrafine Arctic

4 aerosols will increase in future. However, it was stressed that further studies are needed

5 given other new particle formation source regions and mechanisms exist, including an

6 influence of emissions from seabird colonies (Croft et al., 2016; Weber et al., 1998) and

7 intertidal zones (O´Dowd et al., 2002; Sipila et al., 2016).

8 It is becoming evident that coordinated field measurement studies of the atmospheric size

9 distribution of aerosols are essential to elucidate the complex interactions between the

10 cryosphere, atmosphere, ocean, and biosphere in different regions (Dall´Osto et al., 2018

11 a, b). In this regard, an emerging multi-year set of observed aerosol number size

12 distributions in the diameter range of 10 to 500 nm from five sites around the Arctic Ocean

13 (Alert, Villum Research Station – Station Nord, Zeppelin, Tiksi and Barrow) was recently

14 assembled and analysed (Freud et al., 2017). Major accumulation mode aerosol sources

15 were found over central Siberia and western Russia, and wet removal by snow or rain was

16 found to be the main sink for accumulation-mode particles. It was argued that there is no

17 single site that can be considered as fully representative for the entire Arctic region with

18 respect to aerosol number concentrations and distributions. Following the pioneering study

19 of Freud et al. (2017), the aim of this paper is to present a detail analysis of the main

20 differences and similarities of the aerosol general features of the number size distributions

21 between three different sites across the Arctic in the North Atlantic sector. Here, we use

22 the stations named Gruvebadet (GRU), Zeppelin (ZEP) and Villum Research Station –

23 Station Nord (VRS). The European Arctic is understood here as the part of the circumpolar

24 Arctic located between Greenland and northwest Russia. Geographically, Greenland is

25 part of the continent of North America. However, the island is politically and culturally

1   associated with Europe (specifically with Norway, Denmark, as well as the nearby island of

2   Iceland). In between Greenland and Svalbard there is the Fram Stait - located roughly

3   between 77°N and 81°N latitudes and centered on the prime meridian - the only deep-

4   water connection between the World Oceans and the Arctic. It is important to stress that

5   the Svalbard archipelago is among the Arctic regions that has experienced the greatest

6   temperature increase during the last three decades (Nordli et al., 2014); therefore

7   comparing aerosol measurements simultaneously collected in Greenland and Svalbard is

8   essential to better understand aerosol sources and processes that may affect our

9   changing climate. Previous studies have focused on the characterization via air mass

10 origin frequency and occurrence of different aerosol modes over time scales of the order of

11 periods of weeks to years (Strom et al., 2003; Tunved et al., 2010; Nguyen et al., 2016;

12 Lupi et al., 2016), but only using a single station as monitoring site. A brief comparison

13 between ZEP and GRU was made in Lupi et al (2016), showing good agreement over a

14 period of three months. However, to capture all scales of the variability of Arctic aerosols,

15 it is important to merge intensive field campaigns and long-term measurements across

16 different stations. Provision of the extensive resource-demanding equipment required is

17 only possible by means of international collaborations such those created in the present

18 work. A growing effort in understanding recent drastic changes in the Arctic climate has

19 stimulated more measurements, and a growing number of monitoring sites and

20 atmospheric measurements are taking place. Freud et al. (2017) for the first time

21 assembled and analysed a multi-year set of observed aerosol number size distributions in

22 the diameter range of 10 to 500 nm from five sites around the Arctic Ocean (Alert, Villum

23 Research Station – Station Nord, Zeppelin, Tiksi and Barrow). Here, aerosol size

24 distributions are analyzed by using k-means cluster analysis (Beddows et al., 2009)

25 applied to a long term dataset composed of three years (2013-2015) simultaneous data

1   from three stations (GRU, ZEP, VRS). This is the first time that the GRU site is used in a

2   multi-year set of observed aerosol number size distributions. All size distributions are

3   quality assured, and not filtered according to any other criteria. The cluster analysis herein

4   applied uses the degree of similarity and difference between individual observations to

5   define the groups and to assign group membership. By doing so, our clustering method

6   provides a specific number of size distributions which can be compared across different

7   time periods and across different monitoring sites (Beddows et al., 2009; Dall´Osto et al.,

8   2018b). Whilst a number of intensive field studies have been focusing on single site

9   datasets (Tunved et al., 2004; Dall´Osto et al., 2017a, Dall´Osto et al., 2018b), cluster

10   analyses of multi-site year-long particle size distributions measurements are scarce (Freud

11   et al., 2017; Dall´Osto et al., 2018b). It is important to stress in this study the only aim was

12   to compare the three stations by apportioning different aerosol categories and possible

13   source associations. Future studies will look at transport, both vertical (i.e. between VRS

14   and GRU/VRS) and horizontal (i.e between GRU and ZEP) of both anthropogenic and

15   natural aerosols.

17   **2. METHODS**

18   **2.1 Site Description**

19   Ultrafine aerosol size distributions were measured at three different sites. Fig. 1 (a,b)

20   shows the location and the sea ice coverage across the whole of 2015 taken as an

21   example.

22   The first measurement site is situated at 78º 580N and 11º 530E on the Zeppelin Mountain

23   in the Ny-Alesund community on Svalbard. The Zeppelin (ZEP) station is located 474m

24   above sea level, it is easily accessible yet practically unaffected by local sources.

1   Compared to stations closer to sea level the Zeppelin station is less affected by local

2   particle production occurring in the surf zone, and to local air flow phenomena such as

3   katabatic winds (Strom et al., 2003). The ZEP station is part of ACTRIS Data Centre

4   (ACTRIS DC, developed through the EU project Aerosols, Clouds, and Trace gases

5   Research InfraStructure Network - URI: http://www.actris.eu - within the EC 7th

6   Framework Programme under "Research Infrastructures for Atmospheric Research"), part

7   of the Global Atmosphere Watch (GAW) programme; and it is likely the longest Arctic

8   aerosol size distribution dataset existing (Strom et al., 2003, Tunved et al., 2010; Freud et

9   al., 2017).

10 The Gruvebadet (GRU) observatory is located in the proximity of the village of Ny-Alesund

11 (78º 55 N, 11º 56 E) in the island archipelago of Svalbard. The observatory is 67 m above

12 sea level, located south-east of the main buildings of the village. The instrument collected

13 aerosol size distribution usually from the end of March to the beginning of September. It is

14 located at about 2Km distant from the ZEP station, at about 350m lower altitude.

15 About 800Km away from Svalbard we have the Station Nord Villum research station

16 (VRS). Located at 81° 36' N, 16° 40' W the station is situated in the most north-eastern

17 part of Greenland, on the coast of the Fram Strait. The sampling took place about 2 km

18 south-west of the main facilities of the Station Nord military camp in two different sampling

19 stations as measurements were shifted in summer 2015 from the original hut called

20 "Flygers hut" to the new air observatory, 300 m west of "Flygers hut". The sampling

21 locations were located upwind from the station for most of the time. Detailed descriptions

22 of the site and analysis of predominant wind directions are available elsewhere (Nguyen et

23 al., 2016; Nguyen et al., 2013)

1 ## 2.2 Dataset

2 ### 2.2.1 ZEP DMPS

3 The Differential Mobility Particle Sizer (DMPS) system comprises a custom-built twin DMA

4 (differential mobility analyser) setup including one Vienna-type medium DMA coupled to a

5 TSI CPC 3010 covering sizes between 25 and 800 nm and a Vienna-type short DMA

6 coupled with a TSI CPC 3772, effectively covering sizes between 5 and 60 nm. The

7 number size distributions from the two systems are transferred to a common size grid and

8 then merged. Both systems use a closed-loop setup. The instrument has been inter-

9 calibrated during an ACTRIS (www.actris.eu) workshop. Sizing and number concentrations

10 are within 1 and 5% from the standard DMPS, respectively (Freud et al., 2017).

12 ### 2.2.2 GRU SMPS

13 Aerosol size distribution in the diameter range from 10.4 to 469.8 nm using 54 channels

14 was evaluated with a commercial Scanning Mobility Particle Sizer SMPS TSI 3034,

15 (Hogrefe et al. 2006), with a time resolution of 10 min and particle size with a resolution of

16 dlogDj equivalent to 0.0312, where Dj indicates the instrumental class size. Further

17 information can be found elsewhere (Lupi et al., 2016).

18 ### 2.2.3 VRS SMPS

19 Scanning Mobility Particle Sizer (SMPS) data were collected in the period 2013-2015 in

20 the size range of 9-915 nm in diameter. The SMPS used either a condensation particle

21 counter (CPC) model TSI 3010 or model TSI 7220. To ensure correct functioning,

22 volumetric flow rates, temperatures and relative humidity (RH) of the aerosol- and sheath

23 flow were monitored, as well as inlet ambient pressure. No additional drying was

1 performed, as the transition from the low ambient temperatures outside of the huts (-45 to

2 +15 ℃, yearly average -15 ℃) to the heated inside (>20 ℃) generally provides sufficient

3 decrease in RH.

5 **2.3 Data analysis and additional chemical and physical supporting data**

6 SMPS data from the three different stations were merged and only days where

7 measurements were taken simultaneously at the three stations were considered in this

8 analysis, resulting in 584 total days. Additional chemical and physical data were included

9 in this study, in order to better describe the aerosol particle types sampled. PM_{10} sampling

10 was performed at GRU station by a TECORA Skypost sequential sampler equipped with a

11 PM_{10} sampling head operating following the EN 12341 European protocol. Aerosol

12 samples were collected daily on Teflon (PALL Gelman) filters from March to September

13 2013-2015, in total 385 daily samples were analysed. MSA was determined by ion

14 chromatography on the aqueous extract obtained from one half of each filter (Becagli et

15 al., 2016). Gaseous NH_3 and SO_2 data, and inorganic aerosol species (Na, Mg, Cl, K,

16 sulphate, nitrate, ammonium) at the ZEP monitoring site were obtained at daily resolution

17 at the NILU website data for the period 2013–2015 (total days 650). Concentrations of

18 Cloud Condensation Nuclei (CCN) were measured continuously using a commercial

19 available Droplet Measurement Technology (DMT) CCN counter at the ZEP station. In this

20 study we selected CCN values when supersaturation is 0.4%. In total, 723 days of

21 sampling were obtained at hourly resolution for the years 2013-2015. The size distribution

22 data were averaged over 24 hours using the start and end time of the chemical

23 measurements.

3 RESULTS AND DISCUSSION

3.1 Average monthly size distributions

The monthly averaged aerosol size distributions at the three sites are presented in Fig. 2.

Simultaneously collected data are presented for the whole years (2013-2015), but only for

8 months (March-October) considering all three sites, due to GRU not having data

coverage during winter months (November through February). The average size

distributions at ZEP and VRS are broadly similar during the months of January and

February (2a-b), with low particle number concentrations and a broad accumulation mode,

although larger at the ZEP site (about 250nm) than at the VRS one (about 180nm). The

months of March and April (Fig. 2c-d) present similar size distributions among the three

stations, showing a main large accumulation mode peak at about 190nm, likely associated

with the Arctic haze occurring mainly during these months. It is worth noting that higher

ultrafine particle number concentrations are seen in these two months relative to Jan-Feb

(Fig. 2a-b). During the month of May (Fig. 2e) a clear increase of ultrafine particles can be

seen at the Svalbard sites (GRU, ZEP) due to local new particle formation. The increased

occurrence of new particle formation events (NPF) in May was found to correspond to the

increasing concentration of biogenic aerosol in the Svalbard sites (Becagli et al., 2016;

Dall´Osto et al., 2017a). Interestingly, the VRS site does not show this enrichment, likely

due to the fact that sea ice is still covering most of the areas near the north-east corner of

Greenland. In contrast, during the summer months of June-August, higher concentrations

of ultrafine particles can be seen progressively at all sites. The changes in sources, sinks

and processes associated with colder autumn months (Tunved et al., 2013; Freud et al.,

2017) progressively shifts the aerosol modes seen at about 20-40nm (September, Fig. 2i)

to a bimodal-like aerosol distribution seen in October (Fig. 2j) with two main aerosol modes

1   at about 50nm and 150nm, respectively. The remaining winter months show low particle

2   number concentrations, and data are available for ZEP and VRS only. The data herein

3   presented so far help us to compare the three monitoring sites. As expected, whilst the

4   sites at GRU and ZEP are broadly similar, the VRS site located in Greenland seems to

5   have fewer new particle events with a shorter summer frequency. In order to fully elucidate

6   the chemical and physical processes affecting the aerosol size distributions, we use

7   statistical tools to reduce the complexity of these SMPS datasets.

10   **3.2 K-means clustering analysis**

13   Approximately 25,000 aerosol size distributions obtained at one hour resolution at the

14   three monitoring sites were averaged to daily resolution and then normalised by their

15   vector-length and cluster analysed (Beddows et al., 2009). The standard procedure used

16   (Beddows at al., 2014) including the Cluster Tendency test provided a calculated a

17   Hopkins Index of 0.20 (Beddows et al., 2009). The method used minimize the sum of

18   squared distances between all points and the cluster centre, allowing identification of

19   homogeneous groups by minimizing the clustering error defined as the sum of the squared

20   Euclidean distances between each dataset point and the corresponding cluster center.

21   The complexity of the dataset is reduced allowing characterization of the data according to

22   the temporal and spatial trends of the clusters. In order to choose the optimum number of

23   clusters the Dunn-Index (DI) identified dense and well separated clusters, it provided a

24   clear maximum for 8 clusters, some of which belonged only to specific times of the day,

25   specific mechanisms as well as specific seasons. The eight K-means clusters obtained

26   exhibited frequencies which varied between 1% and 42% (Table 1), without main clusters

1   dominating the overall population. The individual clusters could be distributed into three

2   main groups named nucleation, Aitken and accumulation categories. This additional

3   categorisation was based not only upon their similar size distributions among each other

4   (see Fig. 3a–d) but also by considering strong similarities between other chemical and

5   physical parameters presented in the next sections. The reduction to the three more-

6   generic classifications was based on our data interpretation, and the average aerosol size

7   distributions of each aerosol category are presented in Fig. 3: (a) pristine and nucleation

8   mode particle types; (b) Aitken mode dominating particle type and (c) accumulation mode

9   dominating particle type, and presented together in Fig. 3 (d).

11   **3.2.1 Aerosol categories and occurrence**

13   An aerosol K-means cluster can be interpreted as a particle size spectrum which is

14   determined by a superposition of individual sources and processes. Therefore, the name

15   associated with each cluster aims only to reflect a main feature associated with the particle

16   size spectrum. It is not possible to associate a single source or process, given that each

17   cluster results mainly from a combination of multiple sources. Fig. 3a (blue line) shows that

18   the *pristine* category is associated with very low particle number concentrations (<100

19   particles cm^{-3}). Figure 3a shows average aerosol number concentrations across different

20   sizes, with two minor modes at 35nm and 135nm. The *nucleation* category (Fig. 3a, red

21   line) shows average daily aerosol number size distributions peaking in the smallest

22   detectable size at 10nm. The name of this category - which will be used below to represent

23   new particle formation events - stands for continuous gas-to-particle growth occurring after

24   the particle nucleation event. By contrast, Figure 3a (green line) shows the average

25   number size distribution with an ultrafine mode peaking at about 20-30nm. We refer this

1 *bursting* category to a population that bursts and begin to exist or develop. Contrary to the

2 *nucleation* category, this one fails to grow to larger sizes. The origins of this particle type

3 can be multiple, including new particle formation with limited growth (so called "apple" new

4 particle formation events), or open ocean nucleation, an Arctic ultrafine primary origin can

5 also not be ruled out.

6 Fig. 3b shows two main aerosol categories with a dominating aerosol mode peaking in the

7 Aitken size range at about 40-60nm. Whilst aerosol category *nascent* possess a main

8 mode at about 40nm, the category *nascent broad* shows a much broader Aitken mode

9 peaking at about 60nm. By contrast, Fig. 3c shows three aerosol categories whose aerosol

10 size distributions are all mainly distributed in size ranges larger than 100nm (accumulation

11 mode dominating. Main modes can be seen at 150nm (category *accumulation_150*), at

12 220nm (category *accumulation_220*) and in the largest detected modes at about 400-

13 500nm (category *coarse*).

14 The temporal frequency during the years 2013-2015 of the eight aerosol categories is

15 presented in Table 1. The category *pristine* presents a remarkably similar occurrence

16 among the three monitoring sites (12-14%). The *nucleation* category seems more frequent

17 at the Svalbard sites (11-15%) relative to the VRS site (8%). Similar patterns can be seen

18 for the *bursting* category, also more frequent at GRU-ZEP (14-21%) relative to VRS (8%).

19 Interestingly, the *bursting* shows high occurrence at GRU (21%), perhaps reflecting some

20 processes occurring near sea level across the fiord. The two Aitken categories (*nascent*

21 and *nascent broad*) do not show much variability (7-21%). By contrast, strong differences

22 are seen in the accumulation dominating mode aerosol categories. For example,

23 *accumulation_150* is frequent at the ZEP site (19%), whereas at the VRS site the category

24 dominating is *accumulation_220* (42%), confirming a recent study specific on

25 characterization of distinct Arctic aerosol accumulation modes and their sources (Lange et

al., 2018). Finally, aerosol category *coarse* shows minor occurrence at all three sites (1-4%).

3.2.2 Annual behaviour

The *pristine* category did not present a clear annual seasonality at the ZEP and VRS sites, although at the GRU site it occurred mainly during spring months (Fig. 4a). The *nucleation* category clearly showed high strong occurrence during summer months at the VRS site. By contrast, at the Svalbard sites (GRU, ZEP) two main periods dominate in May and in August (Fig. 4b). Similar trends can be seen for the *bursting* category (Fig. 4c). Whilst at the VRS site this category shows occurrence similar to the *nucleation* category (Fig. 4b), the Svalbard sites (GRU, ZEP) mainly occur during spring months. As previously discussed (Dall´Osto et al., 2017a, Dall´Osto et al., 2018) the lack of gaseous precursors during spring may be the limiting factors. The two Aitken dominant mode aerosol categories (*Nascent* and *Nascent broad*) show very similar temporal trends peaking mainly during summer months at all three stations (Fig. 4d, e). Previous studies already discussed freshly and locally produced aerosol particles dominating the Arctic summer, driven by an increase in both biological activity and photochemistry as well as limited long range transport from mid latitudes (Ström et al., 2009). Therefore, particles are not growing more than into a pronounced Aitken mode in summer month, particularly in July and August (Tunved et al., 2013, Dall'Osto et al., 2017a). The two accumulation dominant aerosol mode categories mainly occur during wintertime. The *accumulation_150* peaks mainly during the months of February-April (maximum in March) confirming its association with the Arctic haze phenomenon (Fig. 4f) in all three stations. By contrast, the larger *accumulation_220* mode category occurs during all autumn and winter months, including

1 October-December (Fig. 4g). Finally, the last minor cluster (*coarse*) does not show any

2 clear trend due to its low frequency (Fig. 4h). The overall annual temporal frequency can

3 be summarised in Fig, 5, where broader categories can be seen. It is well known that the

4 Arctic atmosphere is heavily impacted by transport of air pollution from lower latitudes in

5 spring compared to in summer (Heidam et al., 2004; Law and Stohl, 2007). The continent-

6 derived winter and spring aerosols, known as Arctic haze, reach their maximum number

7 concentration during late spring, approximately in April (Tunved et al., 2013; Nguyen et al.

8 2016) A recent intercomparison of particle number size distributions from several Arctic

9 stations by Freud et al. (2017) suggests differences between the studied stations

10 regarding cluster frequency of occurrence throughout the year. The most prominent

11 differences were observed between the stations at Barrow and Zeppelin, but the GRU site

12 was not consider in their analysis.

14
15 **3.2.3 Association of aerosol categories with chemical and physical parameters**
16
17
18 Different chemical species of natural and anthropogenic origin may contribute to the Arctic

19 aerosol (Tunved et al., 2013; Hirdman et al., 2010). SO_2 in the Arctic has both

20 anthropogenic and natural sources (Barriel et al., 1986), but in our study it is shown mainly

21 occurring with large accumulation mode during wintertime (Fig. 6a). Combustion-derived

22 particles can be transported to the Arctic and can be accompanied by aging of the aerosol

23 through condensational processes. Our study confirm previous ones where SO_2 was

24 shown to parallel with black carbon both at VRS and ZEP (Nguyen et al., 2013; Massling

25 et al., 2015, Dall´Osto et al., 2017a). By contrast, we find the highest concentrations of

26 ammonia associated with the *nucleation* category. Interestingly, also the two Aitken

27 dominating mode categories (*nascent* and *nascent broad*) show high concentrations of

1 ammonia (Fig. 6b). Ammonia can impact increasing rates of new particle formation and

2 growth via stabilization of sulphuric acid clusters (Kirkby et al., 2011). There is growing

3 interest to better constrain the location, population, and ammonia emissions of the Arctic.

4 Zooplankton excretion and bacterial remineralization of phytoplankton-derived organic

5 matter is believed to be a dominant source in the marine environment (Carpenter et al.,

6 2012) although there remains considerable uncertainty (Lin et al., 2016). The melting of

7 sea ice is also a significant source of ammonium (Tovar-Sanchez et al., 2010), with

8 protein-like compounds accumulating in the sea-ice interface (Galgani et al., 2016), similar

9 processes are also seen in Antarctic sea ice (Dall´Osto et al., 2017b). There is evidence

10 that coastal seabird colonies are sources of NH_3 in the summertime Arctic (Wentworth et

11 al., 2016), although there is still uncertainty (Riddich et al., 2012). Recently, ammonia from

12 seabirds was found to be a key factor contributing to bursts of newly formed coastal

13 particles at Alert, Canada (Croft et al., 2016). However, regions of open water and melting

14 sea ice were found to drive new particle formation in North East Greenland (Dall´Osto et

15 al., 2018b), such events seem not to be related to bird colonies from coastal zones.

16 The association with selected chemical components measured in aerosols collected at

17 GRU and ZEP are shown in Fig. 7. It is important to stress that the aerosol chemical

18 composition shown are derived from PM_{10} measurements, and thus does not necessarily

19 reflect the chemical composition of the aerosol covered by the size distribution analysis

20 herein presented and discussed. Nevertheless, the comparison may help in apportioning

21 aerosol sources and processes. Fig 7 (a-c) shows similar trend for three chemical

22 elements (Cl, Na, Mg). Mechanically generated sea salt particles are normally found in the

23 coarser size fraction which points to a marine source for Na, Mg and Cl. Indeed, the

24 highest concentrations are seen for the *coarse* category (about 350 ng m^{-3}, 300 ng m^{-3} and

25 40 ng m^{-3} for Cl, Na and Mg, respectively), followed for categories *accumulation_150*,

1 *accumulation_220* and *pristine*. Sea spray aerosol (SSA) is generated by bubble bursting

2 due to surface winds. SSA contributes to the global aerosol burden multiple times more

3 than the anthropogenic aerosol (Raes et al., 2000; Grythe et al., 2014). Potassium can be

4 associated with sea salt, although K-rich particles are often also attributed to biomass

5 burning (Hudson et al., 2004; Moroni et al., 2017), correlating with gas-phase

6 measurements of acetonitrile, a good biomass-burning tracer. Indeed, accumulation

7 aerosol categories show high concentrations of potassium (about 25-30 ng m^{-3}), but the

8 trend is not observed for the *pristine* class. Aerosols not only originate from primary

9 sources like sea spray and biomass burning, but can also be formed via secondary

10 processes, through chemical transformation of gas-phase species in the atmosphere, most

11 notably sulphur dioxide, oxides of nitrogen, and volatile organic compounds (Harrison et

12 al., 1999). Non-sea-salt sulphate (nss-SO$_4$) is a mixed source tracer with a large

13 anthropogenic fossil and biomass fuel component and a smaller biogenic marine

14 component. Aerosol nitrate is predominantly anthropogenic and arises from the oxidation

15 of NOx from combustion processes associated with vehicles and industrial activity. A

16 considerable proportion of the these acidic nitric and sulphuric aerosols are neutralized in

17 the atmosphere by NH$_3$ (Asman et al., 1998).The two categories with the highest

18 concentrations of sulphate, nitrate and ammonium are found to be *accumulation_150* and

19 *accumulation _220* (about 500 ng m^{-3}, 120 ng m^{-3} and 65 ng m^{-3}, respectively) suggesting

20 as expected these two are composed of a number of primary and secondary components

21 of anthropogenic origin. It is interesting to note that ammonium is only partly neutralising

22 the Arctic aerosols (in average with one-third) and in the Arctic, the aerosols are highly

23 acidic.

24 Overall, the lowest aerosol mass concentrations seen in Fig. 7 (a-e) are seen for the

25 *nucleation*, *nascent* and *nascent broad* categories. This is not surprising, because the

1   occurrence of new particle formation events and growth to the Aitken mode is mainly

2   controlled not only by the precursor gaseous presence but also by pre-existing particle

3   concentrations (Kulmala et al., 2001). Indeed, these events are often found under low

4   aerosol concentration conditions in remote areas (Tunved et al., 2013). The low aerosol

5   mass concentrations associated with these recently formed categories still allow us to

6   draw some important conclusions about the possible sources forming these new particles

7   in the Arctic. An opposite trend relative to the previously discussed chemical aerosol

8   markers can be seen in Fig. 7h showing methane sulphonate (MSA) concentrations

9   sampled at the GRU monitoring site. The highest concentrations can be seen for the

10   categories *bursting*, *nucleation*, *nascent* and *nascent broad*. MSA is formed via oxidation

11   of dimethyl sulfide (DMS), a gas produced by marine phytoplankton (Gali et al., 2015). It is

12   the most abundant form of biogenic sulphur released from the ocean (Lovelock et al.,

13   1972; Stefels et al., 2007). Previous studies show that the emission of oceanic DMS may

14   impact aerosol formation in the Arctic atmosphere (Levasseur et al., 2013; Becagli et al.,

15   2016, Dall´Osto et al., 2017a). Recent study at the ZEP size shows that during summer,

16   the impact of the anthropogenic sources upon sulphate is lower (42%), with a contribution

17   comparable to that coming from biogenic emissions (35%) (Udisti et al., 2016).The

18   association of MSA not only with the *nucleation* but also with the *bursting* category

19   suggests that secondary processes may drive both categories, somehow pointing to a

20   lower ultrafine primary origin association with this particle type. However, it is important to

21   stress that high uncertainty regarding the mechanism of aerosol production in the Arctic -

22   especially from leads and open pack ice - still remains (Leck et al., 2002). The interactions

23   between the surface layer of the ocean and the atmosphere are highly variable and

24   ecosystem interactions are more important than any single biological variable. For

25   example, Park et al. (2018) discussed the atmospheric DMS in the Arctic Ocean and its

1   relation to phytoplankton biomass. The DMS production capacity of the Greenland Sea

2   was estimated to be a factor of three greater than that of the Barents Sea, whereas the

3   phytoplankton biomass in the Barents Sea was more than two fold greater than that in the

4   Greenland Sea, stressing the occurrence of a greater abundance of DMS-producing

5   phytoplankton in the Greenland Sea than in the Barents Sea during the phytoplankton

6   bloom periods.

7   The chemical nature and origin of the fine particulate matter over Arctic regions, and

8   especially of its organic fraction, are still largely unknown (Kawamura et al., 1996a, b;

9   Leaitch et al., 2018). Water-soluble dicarboxylic acids and related polar compounds,

10 including oxocarboxylic acids and α-dicarbonyls are ubiquitously found from the ground

11 surface to the free troposphere (Decesari et al., 2006; Kawamura and Bikkina, 2016).

12 Primary sources include fossil fuel combustion and burning of biomass and biofuels.

13 Secondary sources include production via photooxidation of volatile organic compounds

14 (VOCs) and unsaturated fatty acids (UFAs) derived from anthropogenic and biogenic

15 sources. VOC sources include wildfire, emissions from snow, ocean, sea ice, boreal forest

16 and tundra (Tunved et al., 2006; Carpenter et al., 2012, Kos et al., 2014; Haque et al.,

17 2016, Mungall et al., 2017). For this study, we were able to compare our SMPS aerosol

18 categorization with two organic chemical species measured at daily time resolution at the

19 GRU monitoring sites. Results are shown in Figure 8. A clear association can be seen for

20 oxalic and pyruvic acids, anti-correlating between them. Broadly, in the remote marine

21 atmosphere, pyruvic acid may be produced by photochemical oxidation of isoprene and

22 other biogenic volatile organic compounds (BVOCs) emitted from marine biota, which are

23 finally oxidized to produce oxalic acid (Carlton et al., 2007; Carpenter et al., 2012; Bikkina

24 et al., 2014). Oxalic acid is often found as the most abundant water-soluble organic

25 compound, and in-cloud processing is recognized as its major production pathway (Yu et

1   al., 2005). Figure 8 further supports our studies suggesting that the aerosol categories

2   defined by low mass concentrations and high ultrafine sub-50nm particles are associated

3   with rather local secondary processes from marine VOC sources. Recent studies have

4   found that lower organic mass (OM) concentrations but higher ratios of OM to non-sea-salt

5   sulfate mass concentrations accompany smaller particles during the summer (Leitch et al.,

6   2018), stressing the importance of marine Arctic organic components responsible for the

7   ultrafine aerosol population.

8   We conclude this section by reporting the calculated average CCN concentrations

9   corresponding to each aerosol category. CCN number concentrations influence cloud

10   microphysical and radiative properties, and consequently the aerosol indirect radiative

11   forcing (IPCC, 2014). The variability of even low concentrations of CCN is important in the

12   Arctic, an environment where cloud formation – and hence cloud forcing – is limited by the

13   CCN availability (Mauritsen et al., 2011). Figure 9 shows that the two accumulation

14   categories (*accumulation_150* and *accumulation_220*) are associated with the highest

15   CCN concentrations (about 125 particles cm^{-3}) with also the highest ratio of CCN over N.

16   Usually, ultrafine particles smaller than 100nm in diameter are considered too small to

17   activate to cloud droplets. However, Leaitch et al. (2016) concluded that 20–100 nm

18   particles from Arctic natural sources can have a broad impact on the range of cloud droplet

19   number concentrations (CDNC) in clean environments, affirming a large uncertainty in

20   estimating a baseline for the cloud albedo effect. Changes in pressure and temperature

21   may not be efficient to generate the required supersaturations needed to activate smaller

22   particles, with the Kelvin effect acting as limiting factor (Browse et al., 2014; Leaitch et al.,

23   2013). However, the low concentrations of accumulation mode aerosols often found in the

24   Arctic may lower water vapour uptake rates during droplet formation, and the resulting

25   increase supersaturation may enable smaller particles to become cloud droplets. The

1 *nascent* and *nascent broad* categories also show associations with high CCN

2 concentrations despite the much lower average size distributions (Fig. 3d) associated with

3 these particle types. Despite anthropogenic influence on the accumulation mode is

4 maintained through winter months, natural sources indeed have a significant impact on

5 particle number over summer. Hereby these natural sources facilitate aerosol activation to

6 cloud droplets and thus cloud formation. *Pristine*, bursting and *nucleation* categories show

7 very low associated CCN concentrations (about 50-75 particles cm^{-3}), only about 30% of

8 the total N being activated. In the previously mentioned study by Dall'Osto et al. (2017a) it

9 is also shown that the new particle formation (NPF) events and the growth of these aerosol

10 particles to a larger size can affect the CCN number concentration, reporting an increase

11 of the CCN number concentration (measured at a supersaturation of 0.4%) of 21%, which

12 is linked to NPF events. Low level clouds are one of the major players controlling the

13 radiative balance in the Arctic. Further multidisciplinary studies are needed in order to

14 understand the processes that determine cloud properties on which particles actually form

15 cloud droplets under various conditions.

18 **4 IMPLICATIONS AND CONCLUSIONS**

20 Aerosol size distributions sampled simultaneously in three background location in the

21 Arctic during 2013-2015 (584 days total data coverage simultaneous at 3 stations) were

22 analyzed by K-means clustering techniques. The K-means analysis identified 8 distinct

23 aerosol size distributions, representing specific aerosol categories: low particle number

24 Arctic concentrations (*pristine*, 12-14%), new particle formation and bursts of ultrafine

25 particles (*nucleation*, 8-21%; *bursting*, 11-21%), ultrafine aerosols dominating the Aitken

26 mode (*nascent*, 7-21%; *nascent broad*, 10-14%), accumulation dominating aerosol mode

1   (*accumulation_150*, 13-42%; *accumulation_220*, 8-19%) and coarse sea spray aerosols

2   (*coarse*, 1-4%). During winter months, mass concentrations of atmospheric aerosols in the

3   Arctic are higher compared to summer. Broadly, this is due to differences in the transport

4   of anthropogenic particles and wet scavenging (Stohl, 2006); local boundary layer height,

5   stability and stratification also play a role (Brooks et al., 2017). By contrast, total aerosol

6   number concentrations in the Arctic are often found to be similar throughout the period of

7   March–September (Tunved et al., 2013). However, the number concentrations in spring

8   (March–April) are almost exclusively governed by accumulation mode aerosols peaking at

9   150nm, while the summer concentrations are associated with elevated numbers of Aitken

10   mode particles and frequent new particle formation events. The main findings of this work

11   follow:

12   • The three monitoring sites experience very pristine low particle number

13   concentrations only 12-14% of the time.

14   • New particle formation and growth and bursts of sub-30nm particles are detected 8-

15   21% of the time. The lower frequencies detected at the Greenland site (VRS 8%)

16   relative to the Svalbard sites (ZEP-GRU, 11-21%) are likely due to the former site

17   being surrounded by the ice stream from the Arctic Ocean and isolated from open

18   ocean and melting sea ice regions emitting biogenic gas precursors. The Aitken

19   mode aerosol categories dominate the summer time periods at all sites (19-35%),

20   but at VRS one has a shorter summer season due to longer sea ice coverage and

21   14º degrees lower yearly average temperature compared to the stations at

22   Svalbard.

23   • Two types of accumulation mode aerosols are found, one associated with the Arctic

24   haze peaking in March-April (monomodal at about 150nm) and one seen during the

25   winter months (monomodal at about 220nm). The site in Greenland (VRS) is

1   exposed to accumulation mode aerosols longer than the one at Svalbard. This is

2   likely due to different transport pathways into the polar dome, a boundary which

3   separates cold air in the Arctic from the relatively warm air in midlatitude regions

4   (Stohl, 2006).

6   The aerosol size distributions data herein compared from three different stations were

7   intercompared for the first time. The study builds additional knowledge to the findings

8   presented by Freud et al. (2017), with a focal point on the new particle formation

9   phenomena as observed in the Arctic environment. This important exercise had to be

10 carried out, and the expected results - although not striking - set the ground for important

11 future studies. In the future, a decrease in sea ice coverage across the Arctic Ocean may

12 increase the annual primary production (Arrigo et al., 2008), and may alter species

13 composition of phytoplankton (Fujiwara et al., 2014). Hence, the emissions of biogenic

14 sulphur gases that are aerosol precursors and hence affect aerosol growth and formation

15 would increase change in summer. In this regard, the location of the monitoring sites at

16 Svalbard and Greenland are ideal to study aerosol formation and transport across the two

17 different regions. The two stations are separated by the Greenland Sea, a highly

18 productive region with a great abundance of DMS-producing phytoplankton (Park et al.,

19 2018). As the DMS production capacity of the ocean depends critically on the

20 phytoplankton species composition and the complex food web mechanisms (Stefels et al.,

21 2007), multidisciplinary studies across these regions are warranted. The recent

22 transformations in the Arctic and their global causes and consequences have put

23 international cooperation in the Arctic Council at the forefront of research in governance

24 (Knecht et al., 2016). Larger atmospheric chemistry and physics datasets are being

25 collected by a number of countries, and this work highlights the benefit that can be gained

from international cooperation. Given that the present work has validated the quality of the presented aerosol size distributions, these data will be used again to address specific questions, including vertical transport (i.e. the two sites at the Svalbard) and horizontal transport (i.e. Arctic aerosol transport from Greenland to Svalbard regions). The significant costs associated with these types of coordinated international collaborations can provide far more information than individual sites operating on their own. This may help to understand better the complex interactions and feedbacks between the aerosol, the clouds, the longwave and shortwave radiation, the ocean dynamics, the biota and the environment (Browse et al., 2014). Special concern is arising also from increasing navigability in the rapidly melting Arctic Ocean with expanding community re-supply, fishing, tourism, fossil fuel exploitation and cargo trading, which is projected to cause a large increase in emissions by 2050 (Melia et al., 2016). Future studies looking simultaneously at different Arctic monitoring sites will reduce the uncertainties in future projections of Arctic climate changes and its implications for our planet (Koivurova et al., 2012; Byers, 2013; Conde Perez et al., 2016). Our study supports international environmental cooperation concerning the Arctic region.

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2
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4   **LIST OF TABLES**
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6

Aerosol category	GRU	ZEP	VRS
(1) *Pristine*	13	12	14
(2) *Nucleation*	11	15	8
(3) *Bursting*	21	14	8
(4) *Nascent*	21	11	7
(5) *Nascent Broad*	14	10	11
(6) Accumulation 150	13	14	42
(7) Accumulation 220	6	19	8
(8) *Coarse*	2	4	1
Total	100	100	100

Summary of main aerosol modes			
Pristine (1)	13	12	14
Nucleation (2,3)	32	29	16
Aitken (4,5)	35	21	19
Accumulation (6,7,8)	20	38	52
Total	100	100	100

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11 **Table 1. Occurrence of the K-means cluster analysis featuring the eight aerosol**
12 **categories detected at the three monitoring sites. At the bottom of the table reported**
13 **are general aerosol size distribution modes representing as sum of selected aerosol**
14 **categories.**
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2   **LIST OF FIGURES**

5   (a) January   (b) February   (c) March

       (d) April   (e) May   (f) June
7

10   (g) July   (h) August   (i) September

12   (j) October   (k) November   (l) December
13
14   **Figure 1a. Sea ice (light blue), open water (dark blue), snow on land (grey) and land**
15   **(light green) maps for the period March-October (a-h). Land borders are marked in**
16   **dark green.**
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    (a) January   (b) February   (c) March

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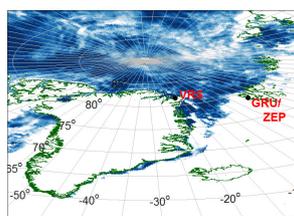
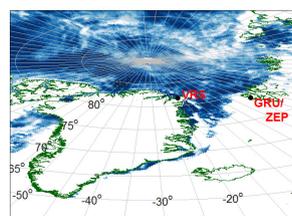
    (d) April   (e) May   (f) June

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    (g) July   (h) August   (i) September

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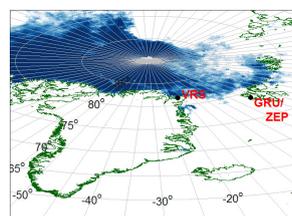
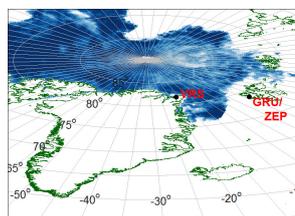
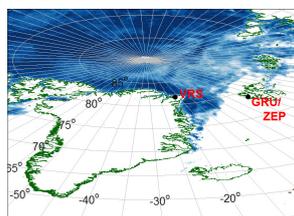
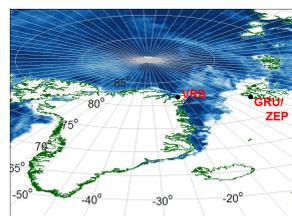
    (j) October   (k) November   (l) December

14 **Figure 1b. Sea ice maps (sea ice in dark blue) for the period March-October (a-h).**
15 **Land borders are marked in dark green. Snow, land and open water in white.**
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(a) January   (b) February

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(c) March   (d) April

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(e) May   (f) June

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(g) July   (h) August

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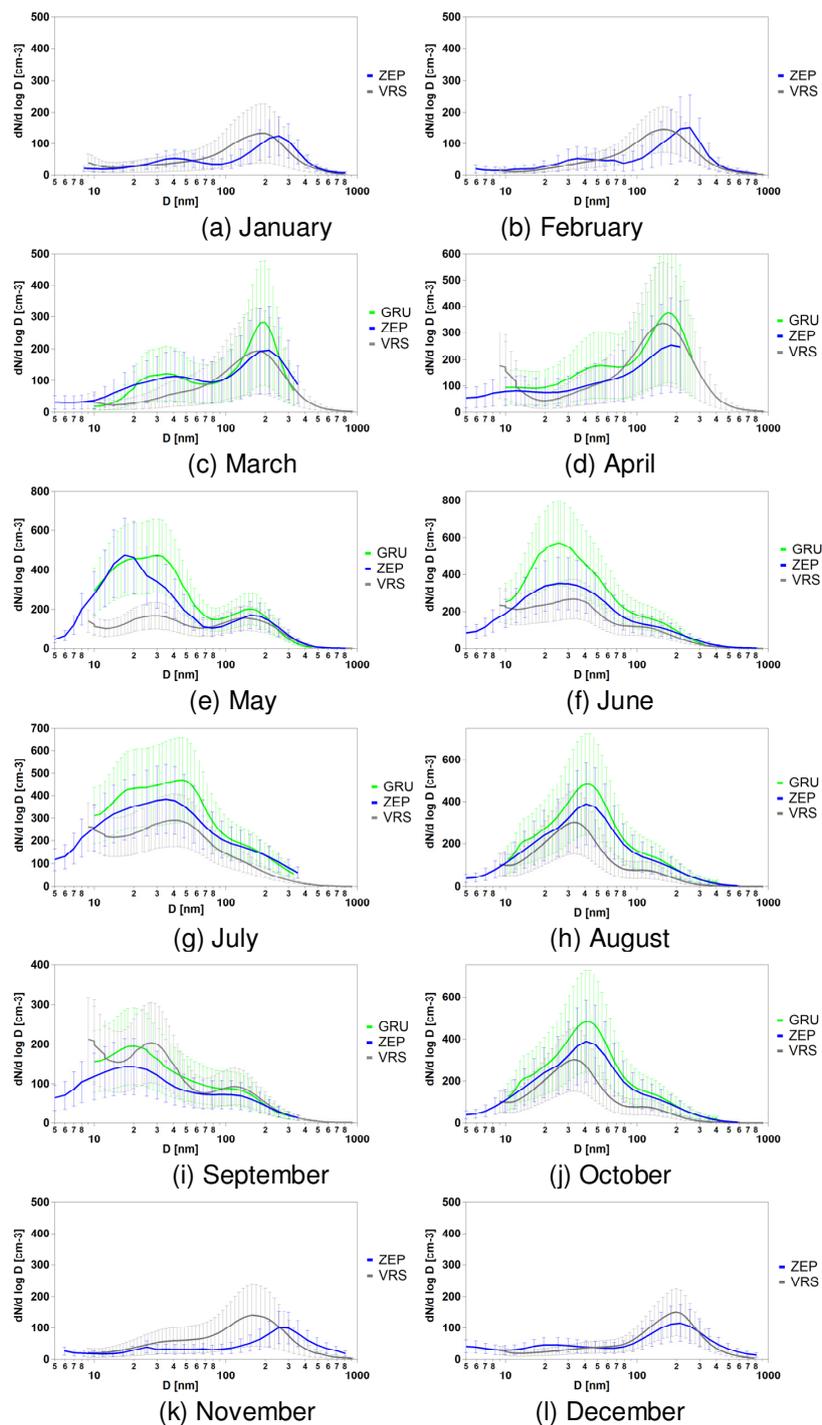
(i) September   (j) October

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(k) November   (l) December
13 **Figure 2. Monthly average size distributions taken at the three sampling sites for the**
14 **period January-December (a-l).**

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(d)
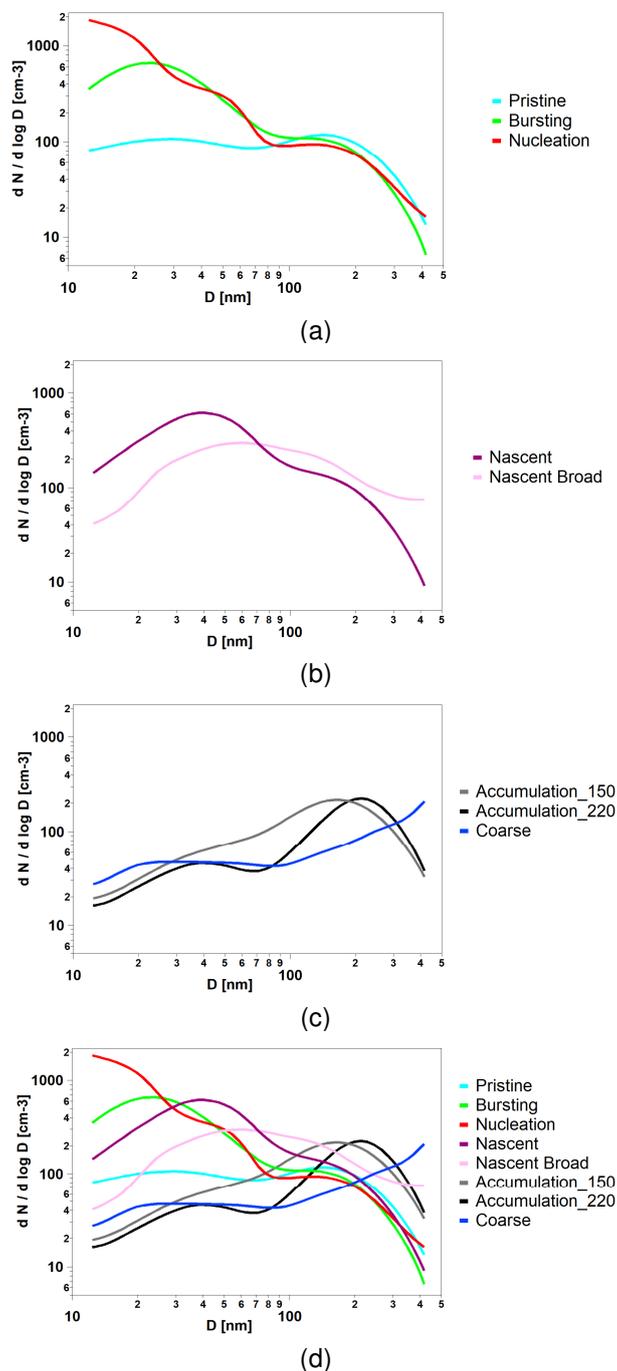
Figure 3. K-means aerosol categories for (a) *Pristine, Bursting, Nucleation,* **(b)** *Nascent, Nascent Broad,* **(c)** *Accumulation_150, Accumulation_220, Coarse,* **and (d)** **all average number aerosol size distributions for each of the three sub-groups.**

(a) *Pristine*
(b) *Nucleation*

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(c) *Bursting*
(d) *Nascent*

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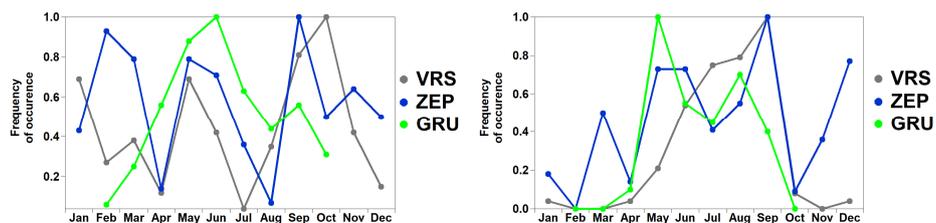
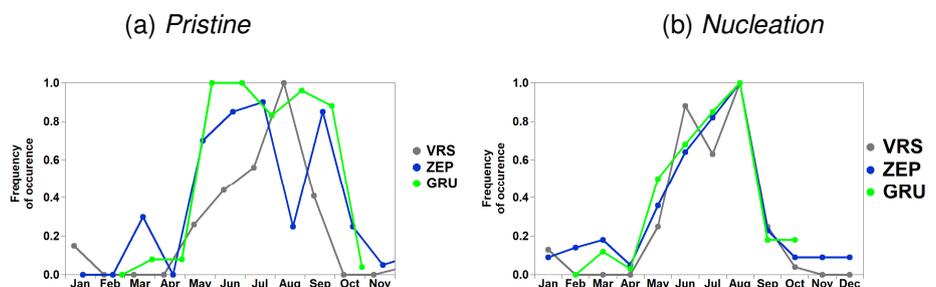
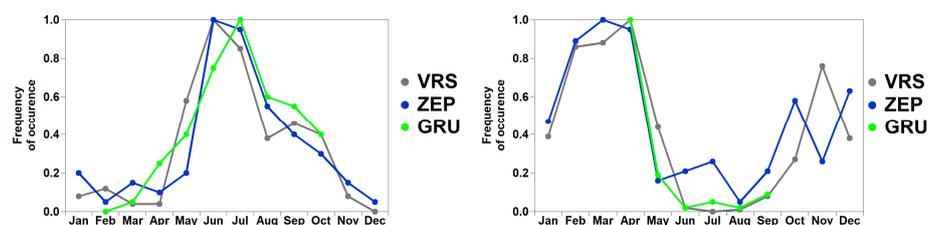
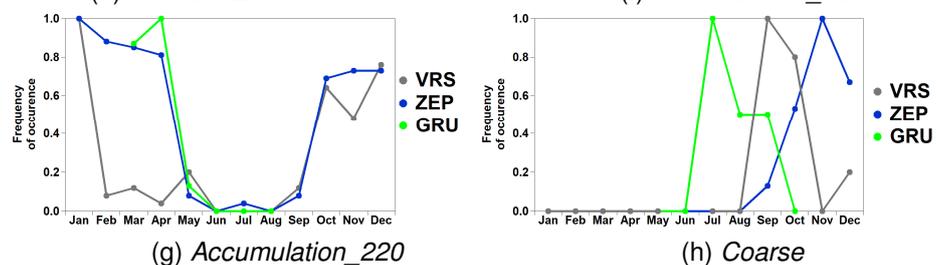
(e) *Nascent Broad*
(f) *Accumulation_150*

(g) *Accumulation_220*
(h) *Coarse*

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17 **Figure 4. Annual variation of the frequency of the monthly cluster count for the**
18 **three stations (VSR, ZEP, GRU).**
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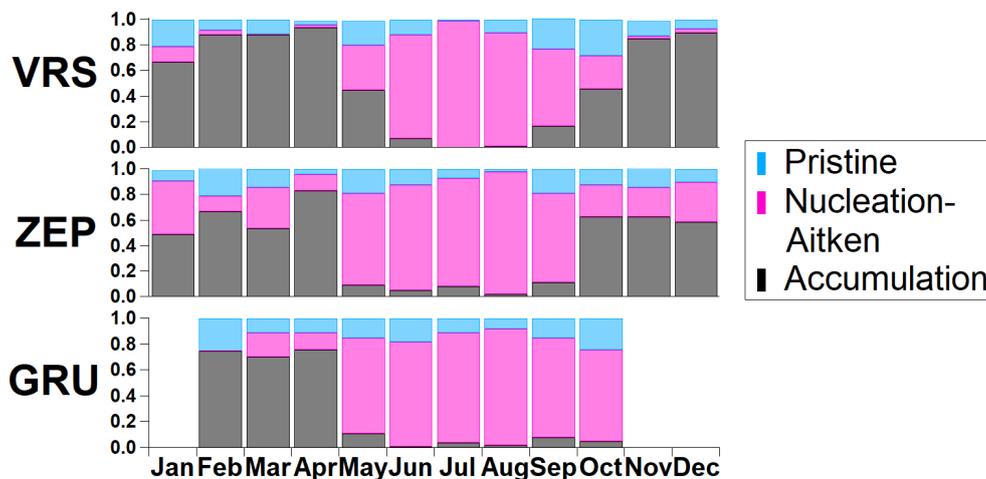
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11 **Figure 5. Annual variation of the frequency of the monthly cluster count for the**
12 **three stations (VRS, ZEP, GRU) summarized in sub-aerosol main categories.**
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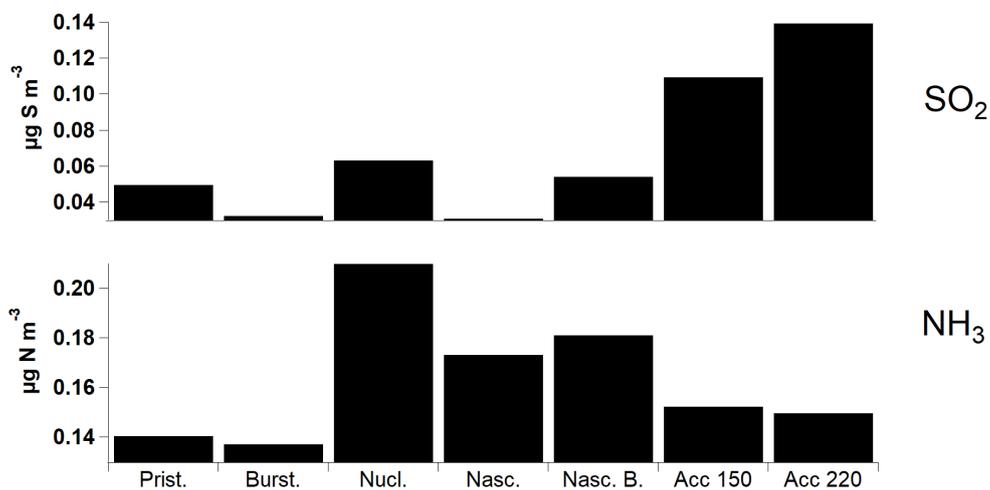
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12 **Figure 6. Average daily concentrations of selected chemical tracers for each aerosol**
13 **category (ZEP only).**
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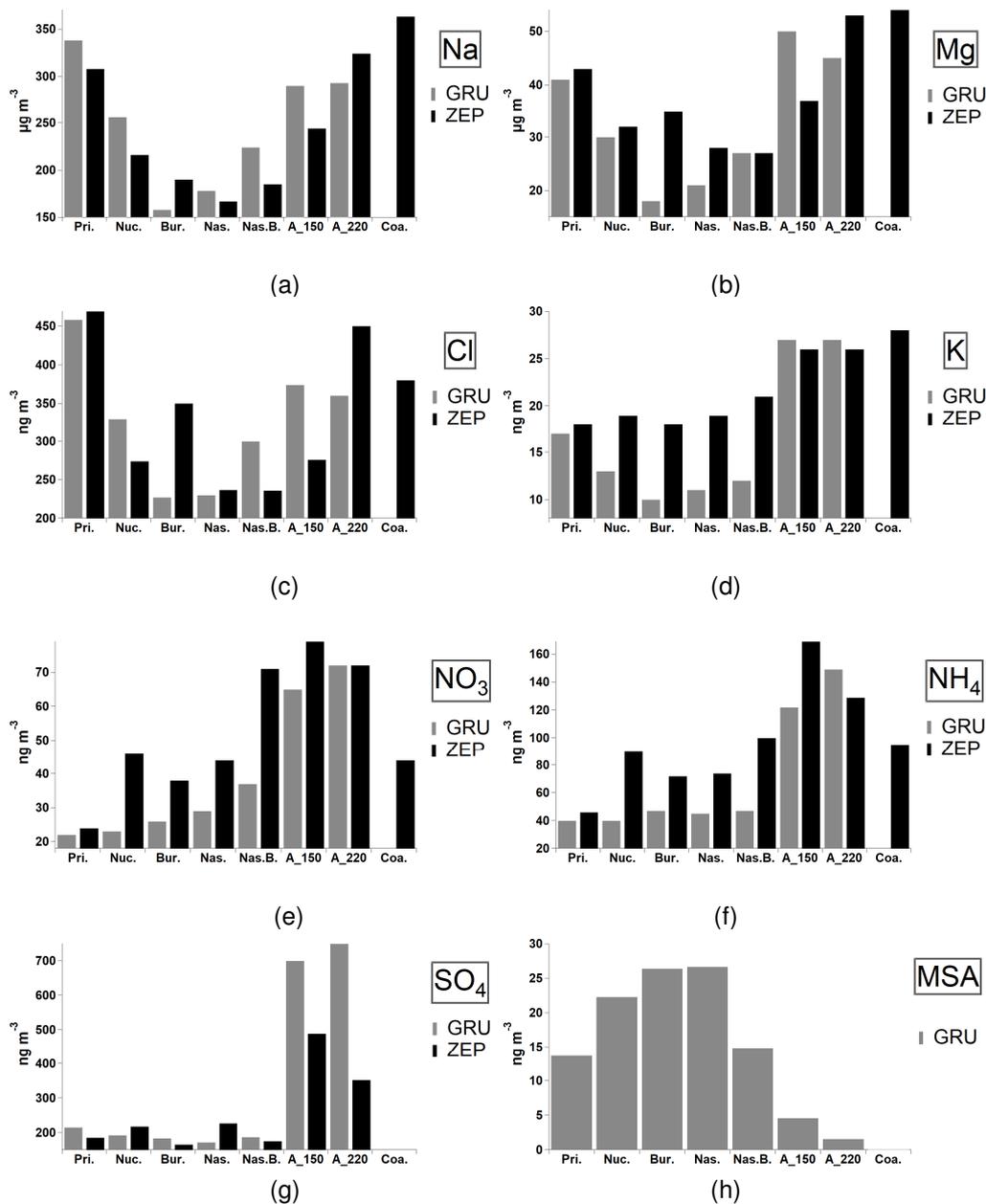
Figure 7. Average daily concentrations of selected chemical tracers for each aerosol category (ZEP and GRU only).

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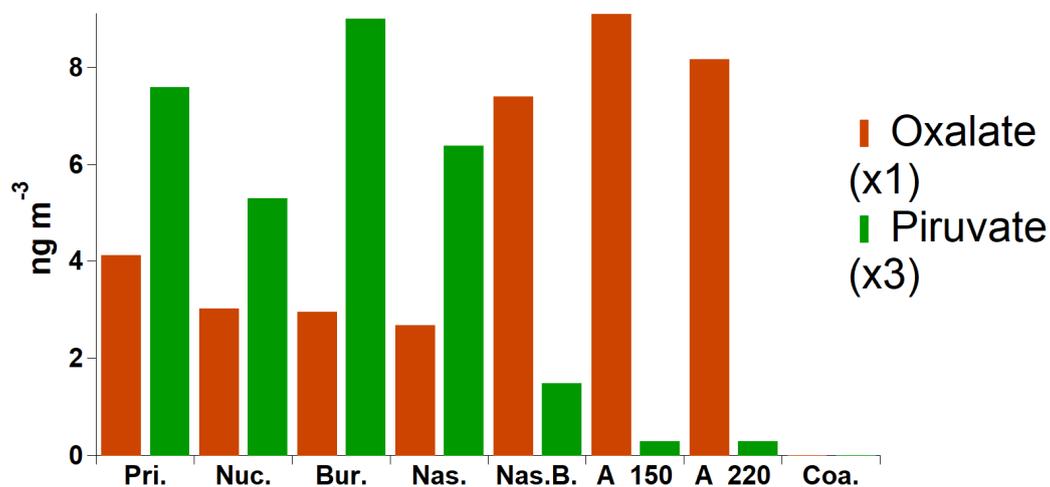
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9   **Figure 8. Average daily concentrations of selected chemical tracers for each aerosol**
10   **category (GRU only).**
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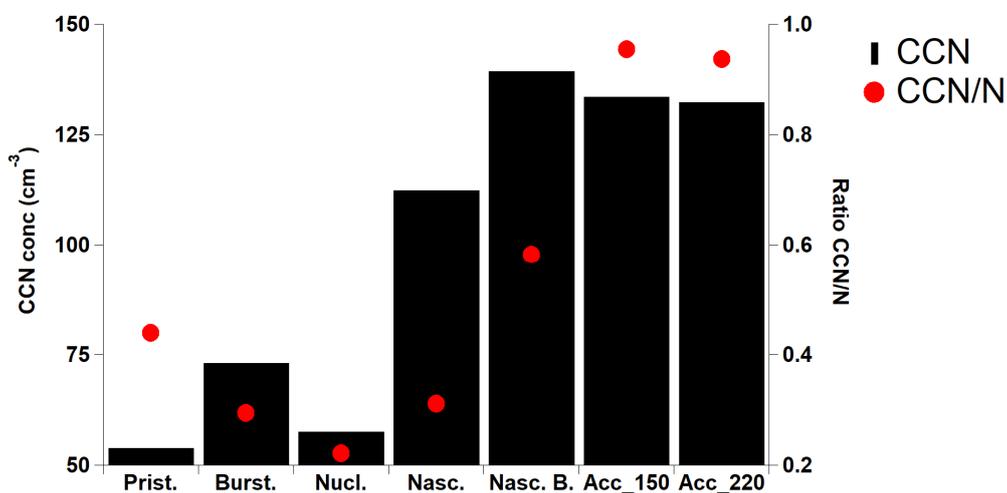
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16 **Figure 9. Average daily concentrations of CCN concentrations for each aerosol**
17 **category (ZEP only).**