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Atmospheric black carbon and sulfate concentrations in Northeast Greenland

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Abstract

Measurements of Black Carbon (BC) in aerosols at the high Arctic field site Villum Research Station (VRS) at Station Nord in North Greenland showed a seasonal variation in BC concentrations with a maximum in winter and spring at ground level. The data was obtained using a Multi Angle Absorption Photometer (MAAP). A similar seasonal pattern was found for sulfate concentrations with a maximum level during winter and spring analyzed by ion chromatography. A correlation between BC and sulfate concentrations was observed over the years 2011 to 2013. This finding gives the hint that most likely transport of primary emitted BC particles to the Arctic was accompanied by aging of the aerosols through condensational processes. This process may have led to the formation of secondary inorganic matter and further transport of BC particles as cloud processing and further washout of particles is less likely based on the typically observed transport patterns of air masses arriving at VRS. Additionally, concentrations of EC (elemental carbon) based on a thermo-optical method were determined and compared to BC measurements.

Model estimates of the climate forcing due to BC in the Arctic are based on contributions of long-range transported BC during spring and summer. The measured concentrations were here compared with model results obtained by the Danish Hemispheric Model, DEHM. Good agreement between measured and modeled concentrations of both BC and sulfate was observed. The dominant source is found to be combustion of fossil fuel with biomass burning as a minor though significant source. During winter and spring the Arctic atmosphere is known to be impacted by long-range transport of BC and associated with the Arctic haze phenomenon.

1 Introduction

Black carbon (BC) is a component of the atmospheric aerosol, which originates from incomplete combustion of fossil fuels, waste, or natural and anthropogenic biomass

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burning (Roberts and Jones, 2004; Yttri et al., 2014). Long-range transported BC to the Arctic is an important atmospheric component as it contributes to Arctic haze, which has a significant influence on the Arctic radiation budget (Quinn et al., 2008; Hirdmann et al., 2010; Flanner et al., 2011; Wang et al., 2011).

BC particles and especially aged BC particles affect the radiation budget directly by scattering and absorbing incoming solar radiation. In case of haze situations the energy can be re-emitted via long-wave radiation (Lubin and Simpson, 1994). In summary, this results in a heating effect below and within the polluted aerosol layer (e.g. AMAP report, 2011). As primary BC particles are hydrophobic, their cloud condensation nucleus activity is very limited unless they do not appear in very large sizes. Particles of inorganic matter have much better potential to act as CCN. Nevertheless, after a certain degree of chemical aging BC particles may have taken up inorganic matter and such coating can lead to modified particles with sufficient CCN and IN activity. In this way, aged BC particles may also affect the radiation budget indirectly by acting as cloud condensation nuclei (CCN) or ice nuclei (IN) and thereby they may change cloud albedo (Koehler et al., 2009). In addition, deposited BC particles on snow or ice-covered surfaces have a heating effect in the lower atmosphere due to a lowering of the surface albedo (Quinn et al., 2008; Wang et al., 2011). Such an effect is estimated to cause more rapid melting in the Arctic (Quinn et al., 2008).

Sulfate is another important component of the atmospheric aerosol. Sulfate particles are very efficient in scattering incoming solar radiation resulting in a cooling effect (IPCC, 2013; Crutzen, 2006). These particles are also highly associated with aging processes as they appear as secondary inorganic aerosols, e.g. as $(NH_4)_2SO_4$ or NH_4HSO_4 (Seinfeld and Pandis, 2006). Furthermore, inorganic sulfate can react with organic aerosol species to form organosulfates (Liggio and Li, 2006; Surratt et al., 2007), which have also been observed in the Arctic (Hansen et al., 2014). The presence of sulfate also enhances the CCN activity in atmospheric aerosol samples (Quinn and Bates, 2011). Sulfate aerosol plays a major role in the Arctic atmosphere because of the climate impact through the scattering effect and cloud forming potential. It has previ-

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ously been shown that the sulfate concentrations observed at Villum Research Station (VRS) at Station Nord, North Greenland, are dominated by anthropogenic emissions and to a lesser extent are associated with the emission of sea spray particles (Heidam et al., 2004; Nguyen et al., 2013).

Several studies have been conducted to characterize the physico-chemical properties of Arctic haze over the past 30 years (e.g. Heidam et al., 1984, 1999, 2004; Iversen, 1984; Barrie et al., 1989; Skov et al., 2006). Lately, studies have been motivated by the potentially significant climate effects of BC in the Arctic (Sharma et al., 2004, 2006; Quinn et al., 2008; Law and Stohl, 2007; Hirdman et al., 2010; Wang et al., 2011). It is well established that the Arctic winter and spring atmosphere is more heavily impacted by transport of air pollution from lower latitudes compared to summer (Heidam et al., 2004; Law and Stohl, 2007).

Recent intercomparisons between model results and measurements of northern hemispheric BC concentrations generally show large discrepancies when simulating the seasonality and magnitude of BC in the Arctic (e.g. Koch et al., 2009; Vignati et al., 2010). However, Huang et al. (2010) demonstrated that a reasonable agreement can be obtained between modeled and measured BC concentrations in the Arctic.

The main focus of this paper is to present the dynamics and the seasonality of black carbon (BC), elemental carbon (EC) and sulfate concentrations over a period of two years and three months at the high Arctic site VRS at Station Nord, North Greenland. In this study the term EC is applied for measurements based on a thermal-optical method whereas BC is used for measurements based on a light absorption/optical method (Petzold and Schönlinner, 2004). Modeled BC and sulfate, and measured BC/EC and sulfate concentrations are compared and the relation between BC and sulfate is put into context of possible aging and transport mechanisms, which explain the resulting observations.

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2 Sampling site and experimental

2.1 VRS at Station Nord

At the Danish military station – Station Nord in North Greenland (81°36′ N, 16°40′ W, and 24 m above mean sea level (m.s.l.)) a monitoring station, Villum Research Station (VRS), for atmospheric measurements is located. The main sampling site during this project is a hut (Flygers hut) situated approximately 2.5 km southeast of the military camp. The hut is equipped with inlets for measuring gases and particles. All measurements presented in this article were carried out at this site except the sampling with a High Volume Sampler used further for elemental carbon (EC) analysis, which is described in more detail in a later section.

2.2 Instrumentation

MAAP (Multi Angle Absorption Photometer)

In the period from May 2011 to August 2013, observations of the aerosol light absorption coefficient have been conducted at VRS at Station Nord (Flygers hut) using the Multi Angle Absorption Photometer (MAAP, Model 5012 Thermo Scientific) (described in details in Petzold and Schönlinner, 2004). The instrument was operated with a sample flow of about 1 $m^3 h^{-1}$ and the aerosol light absorption coefficient was measured at a wavelength of λ = 670 nm. The inlet was used without a size cut off. Aerosol light absorption coefficients were converted to BC concentrations using a specific absorption coefficient of 6.6 $m^2 g^{-1}$, which is a default setting for the MAAP. The specific absorption coefficient is reported to be site-dependent (Petzold et al., 1997; Sharma et al., 2002). The specific absorption coefficient varies strongly with the distance from the source and thus with the aging of the aerosols. According to Petzold et al. (1997) the observed variability ranges from 5 $m^2 g^{-1}$ in extremely remote areas to 14 $m^2 g^{-1}$ at urban locations and up to 20 $m^2 g^{-1}$ at near-street measuring sites in urban areas using an aethalome-

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ter operated at a wavelength of 670 nm. A specific absorption coefficient of 10 $m^2 g^{-1}$ for the Zeppelin measurement station in Norway, Svalbard (78°54′ N, 11°53′ E, and 478 m above m.s.l.) has been reported by Stohl et al. (2007). Also, Sharma et al. (2002) investigated the specific absorption coefficient of BC using a Particle Soot Absorption Photometer (PSAP) and an aethalometer at the Canadian site, Alert, and used a value of about 10 m^2 g^{-1} for the PSAP. It was stated that this coefficient tended to decrease at more remote sites among six different sites.

In the light of these findings, a value of 6.6 $m^2 g^{-1}$ for the specific absorption coefficient was used in this study for a high Arctic remote site. This value is the standard value for the MAAP instrument and it was shown that it matches best in different atmospheric environments using a Multi Angle Absorption Photometer (Petzold et al., 2002).

The specific absorption coefficient is known to vary where Bond and Bergstrom (2006) reported a value of 7.5 ± 1.2 $m^2 g^{-1}$ for freshly emitted (uncoated) soot particles at 550 nm based on a literature review. However, after emission, condensation processes in the atmosphere lead to coating of the BC particles, which can also enhance the absorption of the particles. Based on Andreae and Gelencsér (2006) the specific absorption coefficient for aged BC particles can exceed the value of 7.5 $m^2 g^{-1}$ by a factor of 2. Based on these findings applying a value of 6.6 $m^2 g^{-1}$ may overestimate the estimated black carbon concentration.

Because of relatively low BC concentrations, data for the MAAP were averaged for weekly samples. Combining the noise level of the instrument and the flow uncertainty in the sample flow, we estimated an overall relative uncertainty of 20 % for BC concentrations according to these two factors. But this estimate does not include any uncertainty of the specific absorption coefficient as discussed above. Data of black carbon concentrations originating from absorption measurements have rather more to be interpreted with respect to the uncertainty of the specific absorption coefficient.

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HVS (High Volume Sampler)

EC concentrations at VRS at Station Nord were determined during the same time period, using a Digitel DHA 80 high volume sampler (HVS, Digitel/Riemer Messtechnik, Germany) for PM_{10}. The high volume sampler was operated at a flow rate of 500 L min^{-1} using 150 mm prebaked quarts fiber filters (Advantec, Japan). The sampling was done in weekly intervals implying 5000 m^3 of air were sampled on each filter. The samples were collected at the military station at the edge of the camp at Station Nord located 2.5 km from the main sampling site (Flygers hut). The location of the high volume sampler has practical reasons. It was first established to measure the long range transport of long lived pesticides where there is not any local sources and because there was not enough power also to run a HVS at Flygers Hut. Pollution from the camp with respect to EC cannot be excluded here, but the data have been included in the analysis as a comparison of different techniques still creates new knowledge on the relationships between EC and BC. As the data are on weekly average, the data could not be screened for potential local pollution, which might happen on very short time scales.

The carbon analyzer was a thermal-optical OC/EC instrument from Sunset Laboratory Inc. (Tigard, OR, USA). Punches of 2.5 cm^2 were cut from the filters and analyzed according to the EUSAAR-2 protocol (Cavalli et al., 2010). Only front filters were available, so it was not possible to correct for possible positive artifacts, where volatile species adsorb to the filter and add to the particulate organic carbon mass. In this regard, OC but not EC may be overestimated and should only be used with great caution. The detection limit of this set up is 0.37 µg C for a 2.5 cm^2 filter punch, i.e. 0.0045 µg m^{-3} (Birch and Cary, 1996).

(FPS) Filter Pack Sampler

In addition, weekly aerosol samples were collected at the monitoring station VRS (Flygers hut) using a filter pack sampler (FPS, custom built), which consists of a setup

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with 8 filterpacks in series which are operated for a week at a time. An additional filter pack is always taken as a blank. The filter pack consists of three individual filters. The first filter collects particles. The subsequent filters are specially impregnated to collect various gases, notably SO_2. The filter pack is described in detail in Skov et al. (2006) and Heidam et al. (2004). A flow of 40 L min^{-1} was sucked through the filter pack sampler and a total of about 400 m^3 was sampled on a millipore membrane filter made of mixed cellulose esters. The resulting filters were extracted and analyzed for sulfate using ion chromatography. The technique is described in more detail by Heidam et al. (2004). The uncertainty of the measurements far from the detection limits were better than 20 % expressed as 2 relative SDs (Heidam et al., 2004). The detection limit is 1.5 ng S m^{-3} for both sulfate and sulfur dioxide.

3 The Danish Eulerian Hemispheric Model (DEHM)

The transport and transformation of BC and sulfate to the Arctic originated from anthropogenic and natural sources outside the Arctic was simulated by DEHM (Christensen, 1997; Christensen et al., 2004; Heidam et al., 2004; Skov et al., 2006; Hole et al., 2009; Brandt et al., 2012). The model system consists of a weather forecast model the PSU/NCAR (Pennsylvania State University/National Center for Atmospheric Research) Mesoscale Model version 5 (MM5) modeling subsystem (see Grell et al., 1995). This subsystem is driven by global meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) or National Centers for Environmental Prediction (NCEP). The DEHM model includes 2-way nesting capabilities (Frohn et al., 2002).

In this study the model was set up with a horizontal resolution of 150 km × 150 km south of 60° N and a nested grid of 50 km × 50 km north of 60° N, both model domains with the North Pole in the centre. The vertical resolution was defined on an irregular grid with 29 layers up to approximately 15 km reflecting the structure of the atmosphere. The basic chemical scheme in DEHM includes 67 different species and is based on

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the scheme by Strand and Hov (1994). The chemical scheme has been extended with a detailed description of the ammonia chemistry. Furthermore, reactions concerning the wet-phase production of sulfate have been included. The current model describes concentration fields of 58 photo-chemical compounds (including NO_X, SO_X, VOC, NH_X, CO, O_3 etc.) and several classes of particulate matter, where one class is related to BC. This class comprises of two types of BC components: freshly emitted BC, which is treated as hydrophobic, and aged coated BC, which is treated as hygroscopic material. The transformation from freshly emitted BC to aged, coated BC was treated linearly in the model with a lifetime of freshly emitted BC set to 24 h. Both BC species are assumed to have an upper particle size limit of 1 µm in diameter. The anthropogenic emissions used are based on the Representative Concentration Pathways emissions (RCP emissions) with a $0.5^\circ \times 0.5^\circ$ resolution (Lamarque et al., 2010). These emissions from the EMEP expert database (European Monitoring and Evaluation Programme) are used for the areas over Europe with 50 km × 50 km resolution (Mareckova et al., 2008). Furthermore, the biomass burning emissions are based on the Global Fire Emissions Database version 3 (GFED 3) (van der Werf et al., 2010), which have a horizontal resolution of a $0.5^\circ \times 0.5^\circ$ on a monthly time step. The calculation of the dry deposition velocity is based on the resistance method. The parameterisation of wet deposition is based on a simple scavenging ratio formulation with in-cloud and below-cloud scavenging coefficients for both gas and particulate phase.

There are several uncertainties connected to the model calculations of sulfate and BC. The anthropogenic emissions of SO_2 and BC are given as total yearly values, which have an uncertainty of 20 % or more. These yearly emissions are distributed to daily or hourly emissions using simple time profiles resulting in emissions on shorter timescales having larger uncertainties compared to the total yearly emissions. Furthermore the emissions from biomass burning have even larger uncertainties compared to the anthropogenic emissions. There are also uncertainties associated with predicted three-dimensional wind fields, clouds, precipitation, and turbulence by the MM5 model, especially in the Arctic due to the limited number of meteorological observations inside

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the Arctic. These uncertainties of the meteorological fields could have large influence on the total model uncertainties for a transport over several thousand kilometres from sources in mid-latitudes to e.g. VRS at Station Nord. Finally there are several parameterizations inside the transport model, which increase the uncertainties: the simple scavenging by precipitation (snow or rain), the turbulence parameterization and the bulk representation of the particles by a particle diameter of 1 µm.

4 Results and discussion

4.1 Comparison of BC and EC concentrations

Time series of weekly BC and EC concentrations measured at VRS in the period from May 2011 to August 2013 are presented in Fig. 1a. The maximum concentration of BC was about 0.34 µg m^{-3} during winter 2012/13, while the highest EC concentration was found to be 0.13 µg m^{-3} for the same week. Both the BC and EC weekly minimum concentrations were close to zero. A seasonal variation was observed for BC and EC with highest concentrations during winter and spring and minimum concentrations during summer.

Generally, Fig. 1a shows good agreement between the patterns of BC and EC concentrations, however BC concentrations are often higher compared to EC concentrations. Differences between the two parameters are expected since the BC and EC concentrations are based on two different measurement techniques which both experience several problems due to different artefacts. In Fig. 1b, the correlation between BC and EC is shown resulting in a slope of $y = 0.50$ and a correlation coefficient of $R^2 = 0.64$. In principle, the overestimation of BC in comparison to EC as observed in the time series is reflected in the correlation plot. A further look into seasonal mean values of BC and EC shows that this overestimation is more pronounced during the winter and spring period when Arctic haze is observed (Table 1). The fraction of OC/EC is almost

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constant over the seasons and does not give an additional hint that based on specific emissions the fraction of EC might be underestimated for indicative seasons.

The EC/OC carbon analyzer used in this study is based on a thermal-optical method, which corrects for charring that may otherwise overestimate EC. However, artifacts may also arise if samples contain brown carbon (BrC), which is a part of organic carbon that absorbs in the visible and ultraviolet spectral regions (Kirchstetter and Novakov, 2004; Andreae and Gelencsér, 2006). BrC can be volatilized over a broad temperature range and some of the BrC can be accounted for as EC, hence overestimating the EC concentration (Andreae and Gelencsér, 2006). Another issue is the fact that the combustion temperature of EC and BrC can be lowered in the presence of inorganic species, which are sampled on the filter and can catalyze the oxidation of EC and BrC (Andreae and Gelencsér, 2006). This can result in misinterpretation of EC as OC and hence lead to an underestimation of the EC mass concentration. From inspection of Fig. 1, an underestimation of EC could possibly influence the results, whereas volatilization of BrC on the cost of organic carbon and thus overestimation of EC is not likely to occur.

The difference between BC and EC concentration could furthermore be explained by the assumption that the measured absorption coefficients by the MAAP is ascribed to BC. This might not be entirely correct since a minor fraction of the absorption could be caused by BrC (Andreae and Gelencsér, 2006). Therefore, the higher BC concentration could be explained by a higher content of BrC. Furthermore the default specific absorption coefficient for the MAAP of 6.6 $m^2\,g^{-1}$ could be too low since previous studies suggest an enhancement of the absorption coefficient of aged aerosols due to coating, which can increase the specific absorption coefficient with a factor of two or more (Andreae and Gelencsér, 2006). This would contribute to an overestimation of the BC concentration which could help explain the discrepancy between our BC and EC concentrations. In fact, Petzold et al. (1997) found the specific absorption coefficient varying within a wide range when summarizing published values from a number of different studies including different locations and thus different aging status of the observed aerosol. Additionally, also soil dust is known to have absorbing character (Fi-

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alho et al., 2005), which might enhance the estimated BC concentration when present in the observed aerosol. The MAAP was operated with no size cut-off so that the detection of soil dust cannot be excluded, while the EC measurements had a particle size cut-off of 10 µm in diameter.

Another explanation for occasionally observed differences could be that measurements of BC and EC were not being conducted at the exact same location. The HVS, used to sample EC, was located at Station Nord in the military camp whereas the MAAP was installed approximately 2.5 km from the station ("Flygers hut"). This could explain why the EC concentrations occasionally exceed the BC concentrations since these higher values could originate from local pollution events in the military camp at Station Nord, though it is manned with 5 permanent staffs and consequently has very low emissions.

4.2 Seasonal variation of measured BC and sulfate concentrations

Figure 2 shows the weekly time series of BC and sulfate concentrations measured at VRS between May 2011 and August 2013. Sulfate maximum concentrations were up to 1.746 µg m^{-3} for weekly mean values coinciding with the maximum value during the study period of black carbon concentration at about 0.336 µg m^{-3} (only one data point as indicated in Fig. 2). Minimum sulfate concentrations were close to zero and appeared simultaneously with the minimum of the black carbon concentrations. A clear seasonal pattern was observed for both BC and sulfate with maximum concentrations in winter/spring and minimum concentrations in summer each year (Fig. 2). The observed pattern for sulfate is identical with previous observations at VRS (Heidam et al., 2004).

In comparison, reported BC concentrations from the field stations Alert, Canada (82.5° N 62.3° W, 210 m a.s.l.), and Zeppelin on Svalbard, Norway (78.9, 11.9° E, 478 m a.s.l.), exhibit similar seasonal patterns as observed at Station Nord (AMAP report, 2011). At the Alert field station the highest BC concentrations were found to be up to 0.089 µg m^{-3} and at the Zeppelin field station on Svalbard the highest BC concentra-

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tions were found up to 0.082 µg m^{-3} during winter/spring 2007/08. During the following winter/spring (2008/09) the maximum BC concentration at the Zeppelin field station reached only values up to 0.036 µg m^{-3}. This is in good agreement with values found in this study for a later time period regarding the typical winter and spring maximum values.

Mean concentrations of measured BC at VRS during winter (measurement period: December to February) were about 0.067 ± 0.071 µg m^{-3} compared to the summer (measurement period: June to August) where mean values were about 0.011 ± 0.009 µg m^{-3}. In contrast, the corresponding values for sulfate were 0.485 ± 0.397 µg m^{-3} for winter and about 0.112 ± 0.072 µg m^{-3} for summer during the measurement period. The values are also listed in Table 1.

The ratio between Sulfate and BC is little lower in autumn and winter compared to spring and summer, but does not give a hint that biomass contribution may have a significant impact on the distribution between Sulfate and BC during special time periods. This is different with regard to the ratio between Sulfate and EC where lower values are observed during summer and autumn, which might be indicative for biomass burning during e.g. the summer period as only minor emissions from sulfate are expected from this source compared to BC.

4.3 Comparison of measured and modelled BC and sulfate concentrations

The results of modeled BC and modelled sulfate concentrations from DEHM are presented together with the corresponding measurements observed at VRS in the period from May 2011 to August 2013 in Fig. 3a and b. The examined BC and sulfate concentrations exhibit very similar patterns characterized by higher concentrations in winter/spring compared to summer, which is in accordance with the seasonal cycle of Arctic Haze observed in Arctic regions by several authors (Heidam et al., 1984, 1999, 2004; Iversen et al., 1984; Barrie et al., 1989; Stohl et al., 2007).

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In general, it can be concluded that the seasonal variation of both species is reproduced well by the model. For VRS, the BC measurements state seasonal mean values which are about 1.5 to 2.5 higher compared with the model results during winter and spring. On the other hand the model seems to overestimate the measured sulfate concentrations (seasonal mean values) in autumn, winter, and spring by a factor between 1.3 and 2.3.

Discrepancies seen for BC (Fig. 3a) can be explained by the larger uncertainties of emission inventories used as input for the model of BC compared to emission inventories of sulfur dioxide and sulfate with relation to both the total amount and the temporal and geographical variations. For example, there is little information known about the daily and seasonal activity pattern concerning BC emissions, which is also reflected in the uncertainty of the emission inventories. Emissions from wild and agricultural fires are calculated based on GFED data (Global Fire Emissions Database) on a monthly basis. Emission events rarely last a month and higher resolution data are urgently required, which is expected to improve the agreement between measured and modeled BC concentrations.

Previous studies comparing measured and modeled values for a 10 year period at Station Nord suggested that DEHM in general describes the sulfate concentrations in the Arctic well (Heidam et al., 1999, 2004; Christensen, 1997) which is also apparent from Fig. 3b. Nevertheless, the model performance for the studied period is not as good as for previous years. The reason for that might be changes in the emissions, which not yet have been incorporated in the model. Measurements of sulfate at VRS show a large decrease on more than a factor of two with respect to the yearly mean concentrations from 2008 to 2012, while the model concentrations have only a small variability (±10–20 %). The emissions data in the model were the best available estimate up to date.

The seasonality of air pollutants at Station Nord occurs predominantly because of specific transport patterns of air masses, which are highly related to the location and extension of the polar vortex. Thus, transport of pollution into the Arctic boundary layer from mid-latitudes is much more likely in winter and spring compared to summer. These

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seasonal patterns are well reproduced for BC and sulfate concentrations by the model and measurement results.

Mean concentrations of modelled BC at VRS during winter (December to February) were about $0.040 \pm 0.033\,\mu g\,m^{-3}$ compared to summer (June to August) where mean values were about $0.019 \pm 0.013\,\mu g\,m^{-3}$. In contrast, the corresponding modelled values for sulfate in winter were $0.900 \pm 0.657\,\mu g\,m^{-3}$ and about $0.144 \pm 0.097\,\mu g\,m^{-3}$ for the summer period. The values are also listed in Table 1.

4.4 Atmospheric processing of BC and sulfate in the Arctic

Orthogonal regression was applied separately to the measured and modelled data of BC and sulfate concentrations. The BC and sulfate concentrations were found to correlate with an R^2 correlation coefficient of 0.84 for the measured values (Fig. 4a) and corresponds to a value of R^2 correlation coefficient of 0.85 for the modeled values (Fig. 4b). Pure BC atmospheric particles are known to be primarily emitted, whereas sulfate aerosol is a combination of minor contributions of primary emitted sulfate aerosol and dominated by secondary inorganic aerosol formation favored by the photo-oxidation of sulfur dioxide. For VRS at Station Nord it has previously been shown that anthropogenic emissions of sulfur dioxide and sulfate is the main source of sulfate aerosol by application of the COnstrained Physical REceptor Model (COPREM) and the Danish Eulerian Hemispheric Model (DEHM) (Heidam et al., 2004). The results suggested that photo-oxidation of dimethanesulfide (DMS) and sea-salt sulfate plays a minor role. A recent study by Nguyen et al. (2013) at Station Nord (VRS) also suggests that sulfate aerosol and sulfur dioxide predominantly originate from anthropogenic emissions such as Siberian metal smelters and other long-range transported anthropogenic pollution.

The positive correlation between BC and sulfate concentrations at VRS indicates that they undergo a similar transport pattern despite having partly different sources. However, BC is believed to initially associate with particles in the submicrometer size range. The particles can either grow by condensation or coagulation and in this way

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reach sizes around 0.5 µm where they have the longest atmospheric life times and thus can be transported over the longest distances (Seinfeld and Pandis, 2006; Huang et al., 2010). It should be added that BC from modern fuels (e.g. biomass combustion) is supposed to have sizes of up to a few hundred nanometers when freshly emitted. BC from fossil fuels (e.g. traffic) is rather more emitted in smaller size ranges of up to about 100 nm.

Since BC and sulfate are only partly related to the same combustion sources during winter and spring, we hypothesize that sulfate particles function as transport containers for BC matter as the concentration of sulfate is clearly dominating and mostly formed in secondary processes. Although our study does not investigate the state of mixing of observed aerosols, it is most likely that sulfate and BC are internally mixed as BC appears aged only short time after release, which can be indicated by its loss of hydrophobic character (Swietlicki et al., 2008).

By comparing the scatter plots of measured (Fig. 4a) and modeled (Fig. 4b) values, it is apparent that the DEHM model for VRS results in similar correlation coefficients between BC and sulfate as observed for the measurements as mentioned above. The slopes of the two correlations $y = 5.86$ (measurement results) and $y = 24.15$ (model results) obtained by orthogonal regression are on the contrary rather different from each other stating that from the perspective of measurement results concentrations of sulfate aerosol are by a factor of about six higher compared to concentrations of black carbon aerosol whereas model results state concentrations of sulfate aerosol are by a factor of 24 higher.

Assuming that particles at VRS arrive as internally mixed particles composed of black carbon, sulfate and some other inorganic and organic species, a freshly emitted primary BC particle might experience a substantial diameter growth during long-range transport to the Arctic prolonging its lifetime in relation to dry deposition, because the dry deposition velocity for small particles (50 to 100 nm in size) decreases with increasing diameters up to aproximately 0.5 µm (Slinn and Slinn, 1980, 1982) and therewith increasing its probability to reach the Arctic. Additionally, it is known that a low deposi-

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tion rate is observed during the Arctic winter enhancing the long-range transport of air pollutants.

On the other hand, any up-take of soluble material and implied change from hydrophobic to hydrophilic character also enhances the chance of the particle to get involved in a cloud process and thus the chance for wet deposition. But the main pathway in the lower troposphere for Artic Haze during winter and spring is from sources north of the Arctic Front in northern Eurasia over snow or ice covered surfaces (Stohl, 2006; Christensen, 1997). This atmospheric transport is highly episodic and often related to large-scale blocking events over Sibiria and with only none or small amount of precipiation events during the transport episodes (Raatz and Shaw, 1984; Christensen, 1997). This means even though the increased hydrophilic character of BC tends to decrease the lifetime, the wet deposition is still small during the transport episodes. This finding also indicates that the source areas for BC and SO_2 (precursor for sulfate aerosol) are somewhat similar, especially during the winter and spring time and the removal processes of BC and sulfate are also similar to a certain extent.

5 Summary and conclusions

The characteristics of Arctic air pollution with respect to BC, EC and sulfate aerosol observed at the high Arctic site, Villum Research Station at Station Nord, for a period of two years and three months was investigated in this study to improve our understanding of the particle dynamics in relation to their properties, life times and and mutual dependencies.

BC and EC concentrations followed the same pattern, but measured concentrations values of BC were substantially higher which is explained by individual limits of the different techniques. Measurements together with model results were used to estimate the concentration levels and the seasonal variation of BC and sulfate to the Arctic and to understand the interplay of the two species during transport to the Arctic region. Measured BC concentrations showed average concentrations of 0.067 $\mu g\,m^{-3}$ in

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winter and 0.011 µg m^{-3} in summer compared to values of 0.485 µg m^{-3} in winter and 0.112 µg m^{-3} in summer for sulfate. Corresponding modeled values were 0.040 µg m^{-3} in winter and 0.019 µg m^{-3} in summer for BC and 0.900 µg m^{-3} in winter and 0.144 µg m^{-3} in summer for sulfate.

It was found that BC and sulfate concentrations exhibit very similar patterns, characterized by higher concentrations in winter/spring compared to summer, which is in accordance with the seasonal cycle of Arctic Haze and its annual variation. This finding can be concluded for both, measurement and model results.

A comparison of modeled and measured concentrations of BC and sulfate has been carried out. The results show good agreement between the modeled and measured concentrations of both species with respect to their seasonal pattern indicating that the model is able to describe the strong seasonal variation of both BC and sulfate concentrations. Nevertheless, the relationship in mass ratio between sulfate and BC is observed to be much higher for the model results compared to the measurement results. The most likely reason for this discrepancy is seen in the uncertainty of emission inventories for black carbon that are used as input variable for the model. Little information is known about these emissions with respect to their temporal and geographical variations.

A specific relationship between sulfate concentrations and BC was observed for the entire study period. This indicates that both species undergo a common aging process, which predominantly leads to accumulation of BC and sulfate containing particles following the same transport pattern to the high Arctic. Sulfate aerosol seems to function as transport container for BC matter. In this way, particles are composed of a mixture of carbonaceous and inorganic material and reach sizes enhancing their lifetime giving the possibility to be transported over long distances in the atmosphere reaching eventually e.g. the high Arctic. A low deposition rate during the Arctic winter is most likely supporting this process.

Author contributions. A. Massling prepared the manuscript with contributions from all co-authors. I. E. Nielsen and D. Kristensen analyzed the filters for EC measurements,

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I. E. Nielsen evaluated the MAAP measurements and provided a literature review for the article. Q. T. Nguyen prepared the instruments for analysis and calibrated those. H. Skov and L. L. Sørensen designed the experiments. J. K. Nøjgaard and M. Glasius and supervised the data analysis, J. K. Nøjgaard controlled the carbon analysis. B. Jensen installed the sampling techniques and carried out the experiments. J. H. Christensen developed the model code and performed the simulations. All authors have contributed to revising the manuscript and have approved the final version.

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Table 1. Mean measured and modeled BC and sulfate concentrations together with mean measured EC and OC concentrations during the different seasons at VRS (Station Nord), North Greenland between May 2011 and August 2013.

	BC measured [μgm^{-3}]	Sulfate measured [μgm^{-3}]	BC modeled [μgm^{-3}]	Sulfate modeled [μgm^{-3}]	EC measured [μgm^{-3}]	OC measured [μgm^{-3}]
Summer (Jun, Jul, Aug)	0.011 ± 0.009	0.112 ± 0.072	0.019 ± 0.013	0.144 ± 0.097	0.020 ± 0.022	0.151 ± 0.079
Autumn (Sep, Oct, Nov)	0.022 ± 0.034	0.138 ± 0.120	0.011 ± 0.015	0.317 ± 0.398	0.019 ± 0.020	0.144 ± 0.077
Winter (Dec, Jan, Feb)	0.067 ± 0.071	0.485 ± 0.397	0.040 ± 0.033	0.900 ± 0.657	0.036 ± 0.028	0.202 ± 0.149
Spring (Mar, Apr, May)	0.054 ± 0.029	0.480 ± 0.243	0.022 ± 0.017	0.618 ± 0.582	0.042 ± 0.018	0.245 ± 0.061
Total no. of samples	106	94	119	119	78	78

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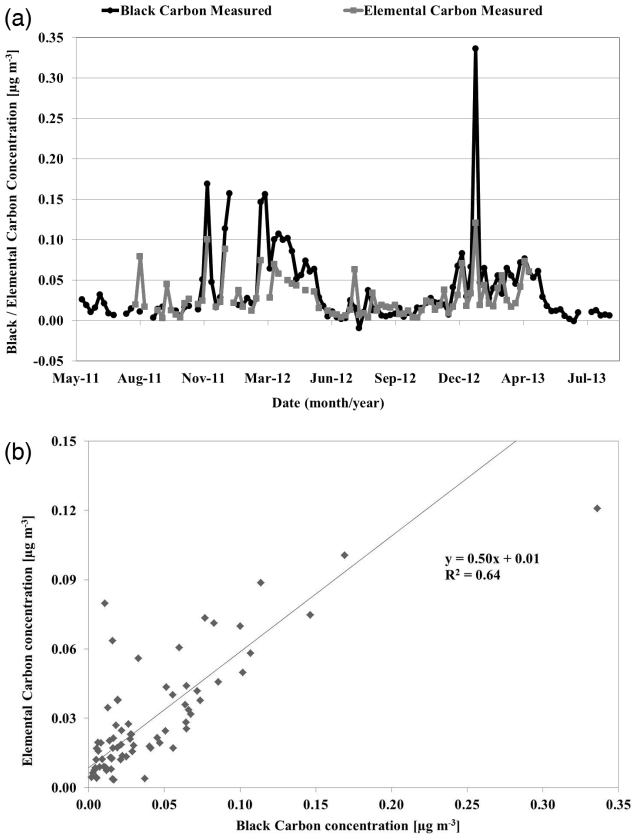

Figure 1. (a) Weekly measured BC and EC concentrations in $\mu g\,m^{-3}$ from May 2011 to August 2013 measured at VRS (Station Nord), North Greenland. Both BC and EC show the same seasonal variation with maximum concentrations in winter and spring and minimum concentrations in summer, **(b)** scatter plot of measured BC and EC concentrations (the slope obtained by orthogonal regression is $y = 0.50$ showing a correlation coefficient of $R^2 = 0.64$).

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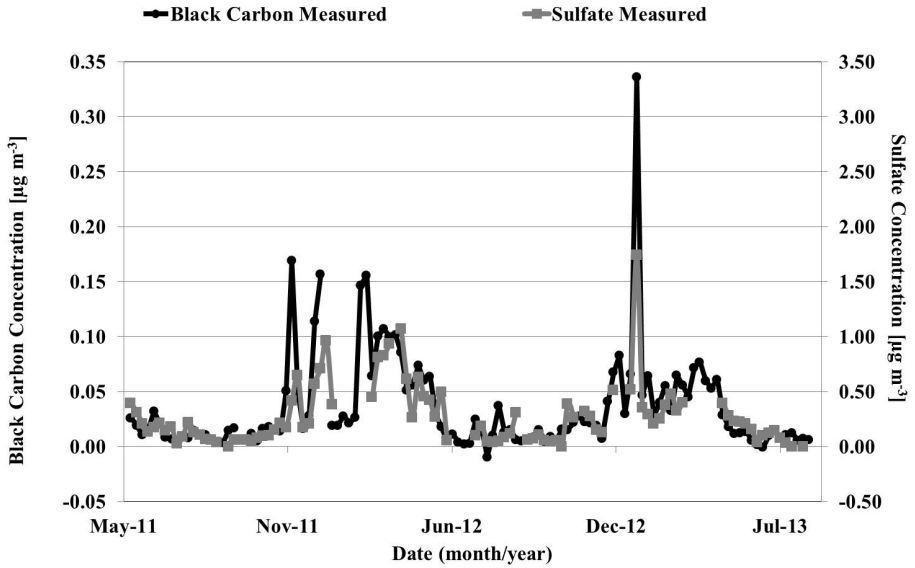

Figure 2. Weekly measured BC and sulfate concentrations in $\mu g\,m^{-3}$ from May 2011 to August 2013 measured at VRS (Station Nord), North Greenland. Here, BC and sulfate concentrations exhibit very similar patterns with maximum concentrations in winter and spring compared to minimum concentrations in summer.

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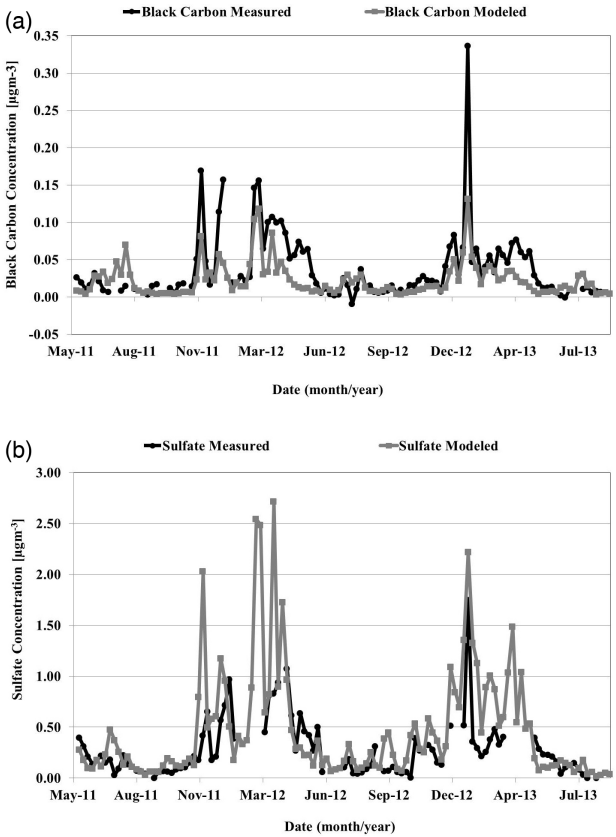

Figure 3. Comparison of model and measurement results from DEHM for **(a)** BC and **(b)** sulfate concentrations at VRS (Station Nord), North Greenland, during the time period from May 2011 to August 2013.

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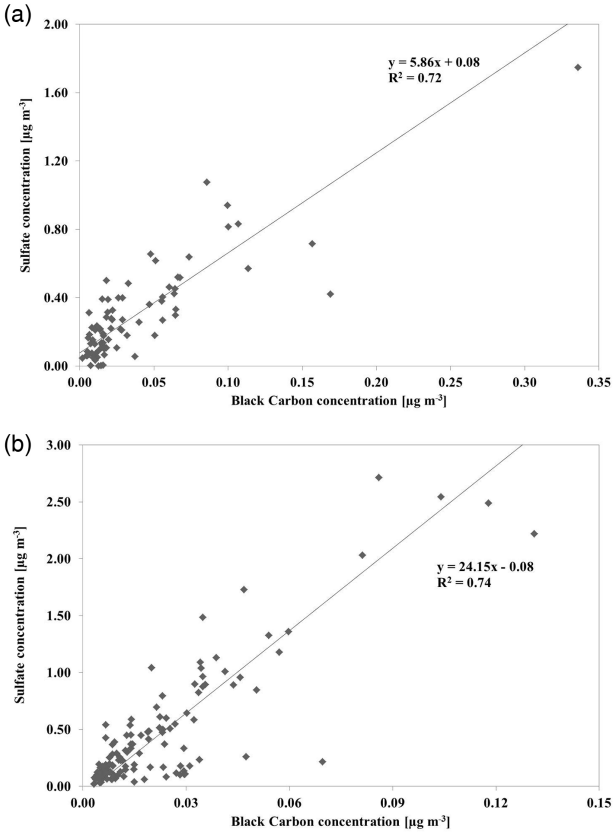

Figure 4. Scatter plot of BC and sulfate concentrations based on **(a)** measurement results (the slope obtained by orthogonal regression between sulfate and BC is $y = 5.86$ showing a correlation coefficient of $R^2 = 0.72$) and **(b)** model results (the slope obtained by orthogonal regression between sulfate and BC is $y = 24.15$ showing a correlation coefficient of $R^2 = 0.74$) during May 2011 to August 2013 at VRS (Station Nord), North Greenland.

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