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ORIGINAL RESEARCH ARTICLE

Aerosol optical properties over Svalbard: a comparison between Ny-Ålesund and Hornsund

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Summary This paper presents the CAMS model based aerosol optical properties calculated for two Spitsbergen fjords, Kongsfjorden (Ny-Ålesund) and Hornsund (Polish Polar Station in Hornsund) measured between 2010 and 2015. A small decrease in Aerosol Optical Depth (AOD) is shown throughout the study period leading to an alteration of the state of the polar atmosphere. However, the potential differences observed between the stations were not statistically significant. While during the studied period no significant differences in chemical composition between the stations were observed, increasing mean values of Black Carbon (BC) were found to be associated with an increasing number of wild forest fires in remote areas producing smoke plumes, which are further transported over vast distances and reach Spitsbergen.

1. Introduction

It is an unquestionable fact that the Arctic climate is being transformed and as a result of these changes it is being warmed at a rate two to three times higher than the global average (ACIA, 2004; IPCC, 2013). The evidence of climate change can also be observed in the sea, in the reduction of the Arctic Ocean ice cover, in the melting of glaciers and permafrost, and in changes in the functioning of biotic elements. However, the atmosphere and its components play a crucial role in the changes in the Arctic system and the leading position in the context of climate response has been attributed to aerosols (Rodríguez et al., 2012). The Arctic area is very sensitive to any climate shifts due to its lack of

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local anthropogenic aerosol sources. Rapidly changing weather conditions, seasonal changes in solar radiation, surface albedo and the supply of polluted air masses from continental regions make the Arctic one of the more important targets of research on climate change (Kaufman et al., 2002; Nagel et al., 1998).

The highly variable distribution, chemical composition and concentration of aerosols have a strong impact on the distribution of the radiative balance of the Earth by modifying the amount of energy passing through the atmosphere (Charlson et al., 1992; Rozwadowska and Gorecka, 2012; Tomasi et al., 2007; Treffeisen et al., 2004). Thus, these small particles can significantly alter the state of the atmosphere both directly and indirectly.

The degree of climatic impact is related to the levels of aerosols originating from both marine and continental environments (the contents of which vary significantly) and depends on air mass sources and trajectories (Petelski et al., 2014). These aerosol types are, in general, poorly accounted for in climate models. It has been long established that better quantification methods of the radiative forcing of different types of aerosols are needed to improve the predictions of future climate changes (Brock et al., 2011). Improving our knowledge and understanding of atmospheric processes, including aerosols, has been the subject of interest of international research activities for decades Such research initiatives include, e.g. Aerosol Robotic Network — AERONET, Maritime Aerosol Network — MAN, The Polar Aerosol Optical Depth measurement network project — POLAR-AOD, Aerosol-Cloud Coupling And Climate Interactions in the Arctic — ACCACIA, Polar Study using Aircraft, Remote Sensing, Surface Measurements, and Models of Climate Chemistry, Aerosols, and Transport — POLARCAT, Impact of Absorbing aerosols on Radiative forcing in the European Arctic — iAREA.

The aerosol climate effect balances the surface-atmosphere radiation directly and indirectly by scattering and absorbing sunlight in the shortwave band. This is the intensity of sunlight scattered back to space, absorbed in the atmosphere and arriving to the surface (caused by natural and anthropogenic forces). In the Arctic, the direct effect usually leads to the warming of the atmosphere and cooling of the surface which depends on the amount of solar radiation passing through the atmosphere, on the properties of aerosol particles and also on the reflective state of the surface beneath. The warming effect in polar regions, mostly over ice and snow, causes a warming effect at the surface (caused by its absorption ability and high albedo, which can exceed 0.85 in visible spectroscopy (VIS)) (Engvall et al., 2008; Tomasi et al., 2007). The indirect effect results in cloud evolution patterns which alter the formation process of warm, ice and mixed-phase clouds increasing the number of droplets and the amount of ice particle concentration (Markowicz et al., 2012).

Since small particles scatter more light per mass unit and stay in the atmosphere longer than bigger particles, aerosol size distribution is another key climate criterion alongside aerosol chemical composition and particle size, which influences the number of cloud condensation nuclei.

Both properties (the chemical composition and particle size) together with the changing annual solar cycle and the diverse conditions of the boundary layers make the Arctic a very complicated system to study. Thus, it is crucial for us to understand and to describe the aerosol processes to be able to determine their properties, and therefore their impact on the climate (Stone et al., 2010).

Climate conditions of Spitsbergen are related to its location, i.e. the northern part of the north hemisphere, where both the Norwegian Current and West Spitsbergen Current have a strong influence on local temperatures causing moderate climate conditions. Cold polar air from the north and west (i.e. the high pressure systems around the Greenland and Polar regions) comes into contact with warm, wet marine air from the south (a low pressure system between Greenland and Svalbard) in the Arctic region (Rozwadowska et al., 2010; Treffeisen et al., 2007). Such conditions cause cyclonic circulations with fronts which are responsible for the formation of clouds, rains, and high speed winds. These are key elements which describe the changeable weather in the Spitsbergen region. Such changeability causes unique weather patterns to appear across various areas of the archipelago.

This paper attempts to identify changes in the Arctic atmosphere by determining the horizontal structure of physical, optical and chemical aerosol properties over Spitsbergen using the Copernicus Atmosphere Monitoring Service (CAMS) model reanalysis. It is an atmospheric composition global model, the European Union's flagship Earth-observation programme. The CAMS model is built by EU-funded Global Earth-system Monitoring using Space and in situ data (GEMS) and series of Monitoring atmospheric composition & climate (MACC) projects at European Centre for Medium-Range Weather Forecast (ECMWF) (Inness et al., 2013). Reanalysis of the Atmospheric Composition includes AOD and the particle chemical composition with the following elements: Black Carbon (BC), Organic Matter (OM), Sea Salt (SS), Sulfates (SU) and Dust (DU) at 550 nm (Morcrette et al., 2009).

Two locations were taken into consideration for the analyses of the aerosol properties. These are — Ny-Ålesund, in the north and Hornsund, in the south. Such a choice of geographical locations provides the right conditions to study how significantly different types of air masses over the study area influence aerosol characteristics.

2. Description of the study area

This paper focuses on the comparison of aerosol conditions in two Spitsbergen locations, Ny-Ålesund and Hornsund (Fig. 1).

Ny-Ålesund (NYA) is the northernmost research station located in Kongsfjorden, and thus it is surrounded by mountains and a tundra system. It hosts fifteen permanent research stations run by agencies from ten different countries, which concentrate their research on environmental sciences, mostly atmospheric. Hornsund (HOR), on the other hand, is located in the south of Spitsbergen, and the Polish Polar station, which provides all-year research facilities, is located there.

Such a choice of stations allows us to observe a variety of air masses in both study areas. Both stations conduct continuous aerosol measurements using ground based-techniques, which are common in the study of atmospheric pollutants, including aerosol particles. These are optical methods, which facilitate the collection of data from one

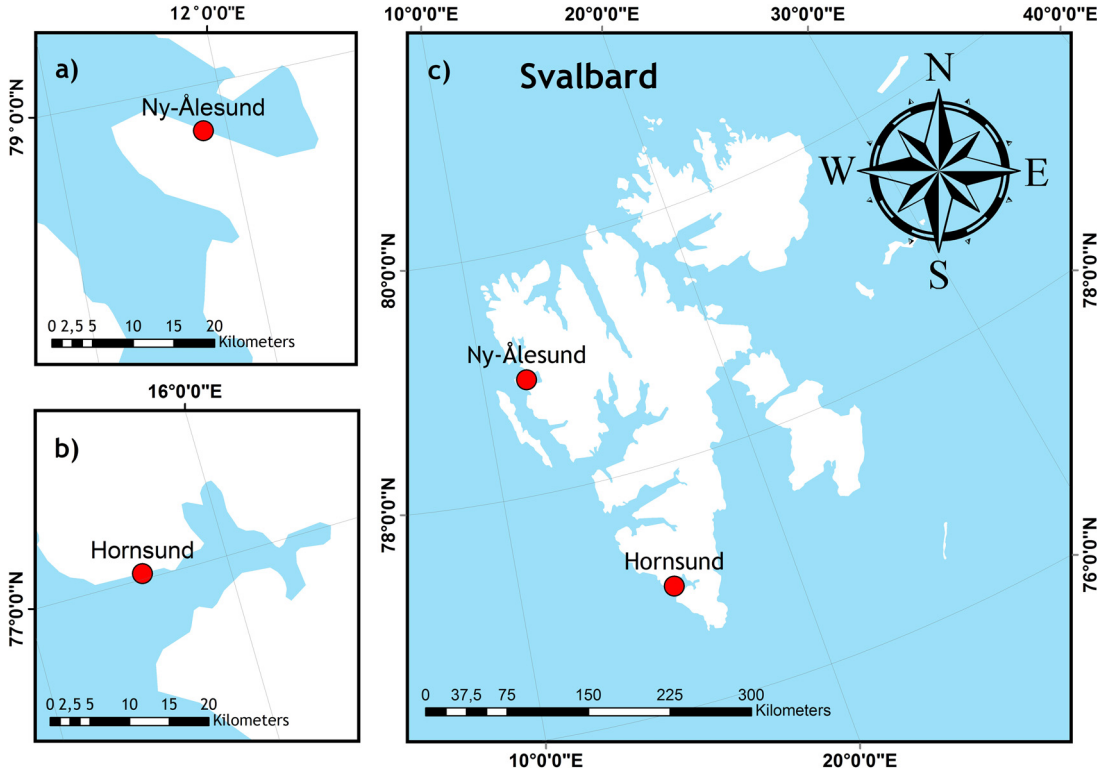

Figure 1 Location of Ny-Ålesund and Hornsund.

location or from a limited area (Dixon, 1998; Drollette, 2000; Labow et al., 1996; Smirnov et al., 2002) and thus, they are very useful in the study of particle optical properties, such as Aerosol Optical Depth (AOD), which is a crucial element in aerosol research (Dubovik et al., 2002; Markowicz et al., 2008; Mazzola et al., 2012; Zielinski, 2004; Zielinski et al., 2012). Satellite remote sensing also facilitates studies of spatial aerosol distribution over a study area, however, it requires the use of good aerosol models.

The comparison of aerosols at both locations for a period between 2010 and 2016 leads to the development of knowledge about temporal and spatial variability of Arctic aerosols through the use of the Copernicus Atmosphere Monitoring Service (CAMS) model reanalysis, which includes in situ datasets.

3. Methodology

Due to the uncertainties in emissions, transport and non-linear physical processes, the modelling and prediction of the state of aerosol particles includes a significant amount of uncertainties (e.g. radiative effects or cloud nuclei formation). Ground-based observation networks, complemented in recent years by satellite sensors which offer a more global view of aerosol distribution, have been crucial in improving our knowledge of this atmospheric component.

ECMWF has been committed to operate the Copernicus Atmosphere Monitoring Service (CAMS) and the Copernicus Climate Change Service (C3S) on behalf of the European Commission until the end of 2020. We used this model for our analyses of the results presented in this article. It includes the ECMWF Integrated Forecasting System (forward model) which was extended by modules of atmospheric chemistry, aerosols and greenhouse gases, and also a data-assimilation module (Bellouin et al., 2008). The forward modules use 12 prognostic variables (11 aerosol mass mixing ratios and one precursor — SO_2) and they include both the satellite and in situ data.

The initial physical parameterizations of aerosol particulate processes mostly used the aerosol approach established by the Laboratoire de Météorologie Dynamique general circulation model (LOA/LMD-Z model) (Boucher et al., 2002; Reddy et al., 2005). Natural particles such as mineral dust and sea salt are set with 3 sizes of bins and for dust, the bin limits are at 0.03, 0.55, 0.9, and 20 μm, while for sea salt — at 0.03, 0.5, 5 and 20 μm (Benedetti and Fisher, 2007).

Dust emissions are based on available characteristics such as winds at 10 m a.s.l., soil moisture, UltraViolet-Visible Spectroscopy (UV-VIS) of the surface albedo or a land area with vegetation during the snow-free conditions. Sea-salt emissions are described by using a source function based on works by Guelle et al. (2001) and Schulz et al. (2004). This approach involves wet sea-salt mass fluxes at 80% relative humidity to be integrated for the three size bins between 2 and 4 μm. Sources for the other aerosol types which are linked to emissions from domestic, industrial, power generation, transport and shipping activities, are taken from the SPEW (Speciated Particulate Emission Wizard; http://www.hiwater.org/spew.htm), and EDGAR (Emission Database for Global Atmospheric Research; http://edgar.jrc.ec.europa.eu/) annual- or monthly-mean climatologies. Emissions of OM, BC and SO_2 linked to fire emissions are obtained using the

GFAS (on MODIS) satellite observations of fire radiative power, as described in Kaiser et al. (2012). Removal processes are also included, and these are: dry deposition with turbulent transfer to the surface, gravitational settling, and wet deposition including rainout by large-scale and convective precipitation as well as wash-out of aerosols within and below clouds (Morcrette et al., 2011). Finally, effects of hygroscopy are described for OM and BC particles (Morcrette et al., 2011).

The CAMS reanalysis of reactive gases (Inness et al., 2013), aerosols and atmospheric composition reanalysis (covers tropospheric and stratospheric reactive gases and aerosols) as well as the meteorological fields are presented in one consistent data set. The meteorological observations assimilation is based on ECMWF RD setup, which include the satellites, sondes and surface measurements. The assimilated observation also includes AODs, recovered from the MODIS instruments onboard the Terra and Aqua satellites over the ocean and dark land surface. The assimilation in the CAMS system is based on satellite atmospheric composition retrievals. The advantages of that is, the subset of trace gases and total aerosol with sufficient accuracy is limited to the place of observations, while satellites cover almost the whole world. Each aerosol type is corrected in relation to the level of its original contribution to the total aerosol mass (Benedetti et al., 2009). These results have been validated by independent observations from the AErosol RObotic NETwork (AERONET).

At the latitude on which Svalbard lies, photometric studies are limited to spring-summer months, since the rest of the year (from late September to early March) is mostly dark or fully dark (polar night). The sunphotometer facilitates the collection of data on cloudless days to avoid cloud contamination. In cases when there are some scattered clouds present, an effort is made to keep at least an angular distance of 30° between the Sun and the closest cloud patch.

Observations are essential in the process of describing the ever changing components of the atmosphere and when it comes to aerosol characteristics, long-term observations are a necessity. In general, particulate optical properties are obtained using ground-based methods. The model database (set to the same parameters) is a supplementary set. This allows us to gather the elements needed to describe aerosol optical parameters, such as AOD and the Ångström Exponent (AE).

The Aerosol Optical Depth is a dimensionless, wavelength dependent parameter which refers to the weakening of direct sunlight passing through the atmosphere. It is a function of the concentration of particles, their size distribution and their chemical composition. AOD is a measure of the state and type of aerosol particles (e.g., urban haze, smoke particles, desert dust, sea salt), which are present in a column of air, measured over a distance between the instrument (Earth's surface) and the top of the atmosphere (WMO, 1994). Thus, the AOD is described as an integrated extinction coefficient τ over a vertical column of unit cross section (see formula (1)). The extinction coefficient is the fractional depletion of radiance per unit path length (also called attenuation, especially in reference to radar frequencies).

$$\mathrm{AOD} = \tau = \int_0^l \epsilon(\lambda, h)\, dh, \tag{1}$$

where ϵ is an extinction coefficient, λ is a wavelength, h is the height ("top") of the atmosphere.

For further interpretations, we used all available AODs from the CAMS model at the following wavelengths: 469, 550, 670, 865 and 1240 nanometers (nm), however, in further analyses we concentrated mostly on AODs at 550 nm.

The AE describes dominating particles in relation to their size. The size distribution of aerosols can be estimated from spectral aerosol optical depth, typically from 440 nm to 870 nm. The negative slope (or first derivative) of AOD with wavelengths in a logarithmic scale is known as the AE. Values of AE greater than 2.0 indicate the presence of fine mode particles (e.g., smoke particles and sulfates), while values of AE near zero indicate the presence of coarse mode particles such as desert dust (Eck et al., 1999). The coefficient characterizes the degree of atmospheric turbidity due to aerosols and is equal to the AOD for a wavelength (λ) equal to 1 μm. The AE is calculated from a minimum of two wavelengths using the following formula:

$$\mathrm{AE}(\lambda_1, \lambda_2) = \frac{\ln \mathrm{AOD}(\lambda_1) - \ln \mathrm{AOD}(\lambda_2)}{\ln \lambda_1 - \ln \lambda_2}. \tag{2}$$

Using the CAMS obtained AODs we calculated AE for two wavelengths: 469 nm and 865 nm, which is the closest proxy to the typical AEs analyzed by other researchers.

The data set was sequenced into analyses of weekly, monthly, seasonal and annual changes in horizontal direction. Distribution, identifying aerosol sources, partial chemical composition, types and size distribution were analyzed with the CAMS model and meteorological data.

Additionally, information on meteorological data (wind speed and direction) has been collected from the weather services of the World Meteorological Organization (WMO) for both stations: Hornsund (01003) and Ny-Ålesund (01007). Wind direction was divided into 16 cases (with a 22.5° step). The main direction, and monthly analyses for the period 2010–2015 are useful when connecting sources of aerosols and in determining local emissions.

4. Results and discussion

The Arctic presents particular challenges when assessing aerosol impact due to large variations in aerosol concentrations, and their varying chemical, physical and optical properties.

Polar aerosols were characterized by Tomasi et al. (2012), who presented a time series of AODs at a number of sites, for the period 1977–2010, including in Ny-Ålesund for the period of 1991–2010. Climatologically, AODs (500 nm) greater than 0.1 during the spring and very low AODs during the summer months persist over the Arctic, and unusually high values of AODs are typically not observed during Arctic summers. Diurnal average AODs (500 nm) varied from 0.12 to 0.25 during the winter-spring time (result of Arctic Haze events) (Rahul et al., 2014). Herber et al. (2002) reported values of AOD (532 nm) during the summer at 0.046 ± 0.012, with over 90% of AODs (532 nm) between 0.022 and 0.070 in summertime. Therefore, the winter-spring AODs of over 0.08 are relatively high due to Arctic Haze episodes, and those below 0.065 characterize clean-air cases. Analyses of time series of AOD measurements carried out in Ny-Ålesund

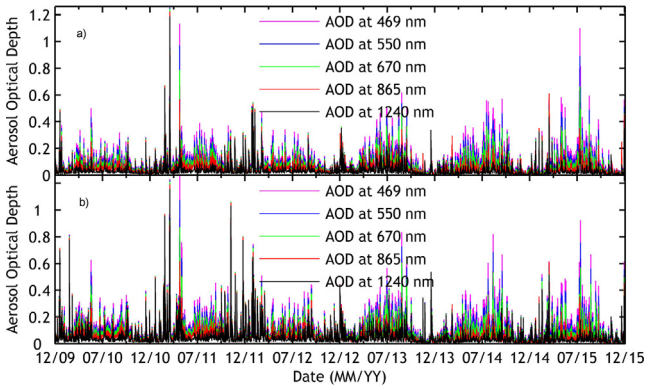

Figure 2 3-hour Aerosol Optical Depth (whole spectrum) at two sites: a) Ny-Ålesund and b) Hornsund based on reanalysis from the CAMS model for the period 2010—2015.

show a noticeable increase in AOD over the last ten years, changing the estimations of the long-term average variability of this parameter from 2.0% per year found by Tomasi et al. (2007) in the 1977—2006 period to nearly zero in the last ten years.

Results from the CAMS model analyses were concentrated on optical and chemical properties. Taking into account the AOD in all spectra, we did not find significant differences between the stations in the period 2010—2015 (Fig. 2). Slight changes in absorption are visible at the beginning of 2010 and in the winter of 2011 but they are mostly concentrated in the water vapor absorption channel — 1240 nm. These changes at longer wavelengths are connected with a low sensitivity to the fine-mode aerosols and relatively well-known absorption properties of dust (almost non-absorbing), given the bigger errors in AOD retrieval. Each peak is almost the same, with small differences in values, which are mostly higher in Hornsund (Fig. 2b).

As mentioned above, there are differences in the water vapor channel, but taking into account the extinction in 550 nm, the higher values of AOD are seen in Hornsund, with exception to 2015, when higher values are observed in Ny-Ålesund (Fig. 3). During the summer of 2015, a very intensive

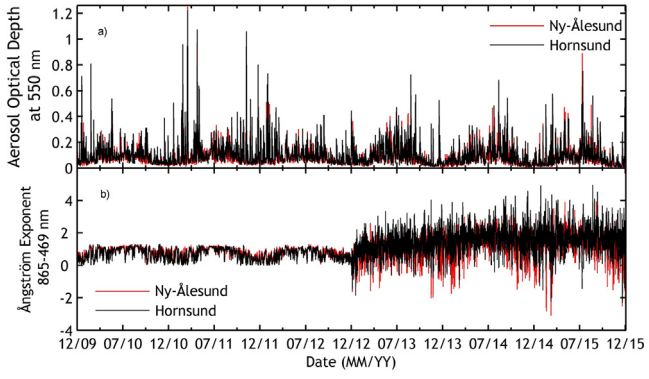

Figure 3 3-hour a) Aerosol Optical Depth at 550 nm and b) Ångström Exponent for 865—469 nm in Hornsund (black line) and Ny-Ålesund (red line) based on reanalysis from the CAMS model during a period 2010—2015. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

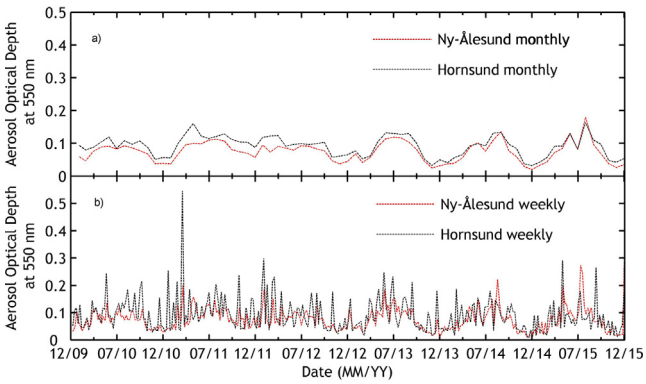

Figure 4 (a) Monthly and (b) weekly means of Aerosol Optical Depth at 550 nm in Ny-Ålesund (red dashed line) and Hornsund (black dashed line) based on reanalysis from the CAMS model during a period 2010—2015. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Arctic Event occurred which was related to massive biomass burning (BB). According to the Canadian Wildland Fire Information System (http://cwfis.cfs.nrcan.gc.ca) forest fires started in late April in Canada. Slight differences seen on the upper plot of Fig. 3 could be explained by the topography of both stations. The event was observed with a delay of several hours between the two stations. This delay was caused by wind circulation, and the speed and direction of winds which transport polluted air-masses. The Ångström Exponent is generally stable during 2010—2012, with a predominance of large particles in the atmosphere. Elevated values of AOD in 2010 and 2011 do not change the composition of the atmosphere (see Fig. 7). After 2012 the Ångström Exponent values changed significantly, increasing in Hornsund and decreasing at the same time in Ny-Ålesund. Sudden, large differences in the extinction in 469 nm and 869 nm, and the resulting differences in the Ångström Exponent, appeared at the beginning of 2012 due to the use of different types of data. From 2010 to 2012 results from reanalysis were used and then, for 2013—2015 — near-real-time data from the CAMS model was applied. The values of the Ångström Exponent above 1.3 in both locations are directly linked to the increasing numbers of recent BB episodes advecting pollution in to the Arctic.

Furthermore, monthly and weekly means of AOD confirm that the mean values are higher in Hornsund than in Ny-Ålesund (Fig. 4). The high values of AOD are mostly related to air-masses from mid-latitudes carrying anthropogenic pollution. From this point, we can conclude that pollution generating events from urbanized areas affect Hornsund more than Ny-Ålesund. It can be confirmed by circulation pathways and local orography. Isaksen et al. (2016) found that during the spring and winter, when polluted air masses are transported mainly from Europe and Asia, the temperature anomaly is more often observed in Hornsund than in Ny-Ålesund. This is strongly connected with anticyclonic types of circulation, which transport air masses from Europe and the Atlantic Ocean. The orography, in addition, has a great impact on the observations of pollution; in Ny-Ålesund during advection from a northern direction a local production of sea-salt is initiated, which could dry-out the polluted air masses.

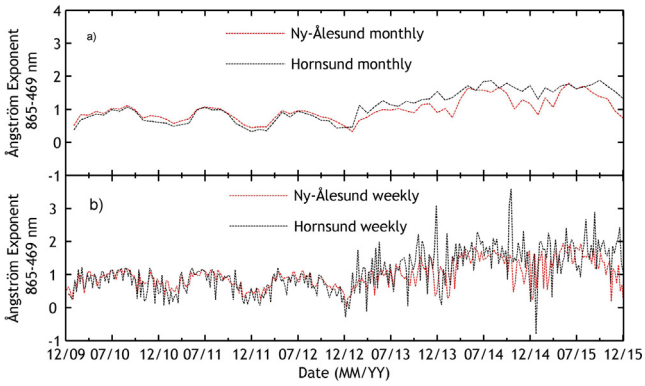

Figure 5 (a) Monthly and (b) weekly means of Ångström Exponent for 865–469 nm in Ny-Ålesund (red dashed line) and Hornsund (black dashed line) and Ny-Ålesund (red dashed line) based on reanalysis from the CAMS model during a period 2010–2015. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Additionally, mountains around the station cause changes in the route of transported air masses (Lisok et al., 2016). Fig. 5 shows weekly and monthly means of AE. The Hornsund station displays higher peaks, with strong a discrepancy especially in 2014. Annual means of this parameter show discrepancy between the stations, higher values in 2010–2012 in Ny-Ålesund, and lower in 2013–2015 (Tables 1 and 2).

Arctic Haze is among most significant phenomena, which occur in the Arctic, however, recent observations of air pollution show that it is mostly related to biomass burning events (Turetsky et al., 2011; Westerling et al., 2006). Over a period of last 10 years, high AODs were observed in particular years (2005, 2006, 2008 and 2011). They occurred as a result of strong aerosol events such as the Arctic Haze or were advected with air masses from BB sources. It is obvious that average AODs show an increasing trend over Svalbard for the period of 2000 and 2012 (Pakszys et al., 2015). This may indicate increasing atmospheric pollution in the region, which may potentially lead to a significant warming at ground level (Tomasi et al., 2015).

Fires influence climate systems on temporal and spatial scales as a result of emissions of trace gases and aerosols (Bowman et al., 2009; Marlon et al., 2008; Power et al., 2008; Ward et al., 2012). Numbers of large biomass burning cases have been increasing over the period of the last 100 years. Unfortunately, it is obvious that anthropogenic activities are the main factor in this process (Brönnimann et al., 2007; Mtetwa and McCormick, 2003). This situation can be related both to climate changes and agricultural fires. These events are a significant global source of atmospheric gases and particles, which influence the chemistry of the atmosphere and thus the climate of the Earth (Beringer et al., 2003; Crutzen et al., 1979; Langenfelds et al., 2002; Page et al., 2002).

Some events in the past have been well described in literature, but they are based on different models and measurements. The Arctic was strongly impacted by aerosol particles from wild fires in Canada in July 2004 (Stohl, 2006; Stohl et al., 2006) and as a result of the Kasatochi and Sarychev volcanic activity, which began in August 2008 (Hoffmann et al., 2010) as well as in July 2009 (Tomasi

Table 1 Annual mean of Aerosol Optical Depth (AOD) at 550 nm, Ångström Exponent for 865–469 nm, five chemical components at 550 nm: Sea Salt, Dust, Organic Matter, Black Carbon and Sulphate in Ny-Ålesund based on reanalysis from the CAMS model.

Ny-Ålesund						
	2010	2011	2012	2013	2014	2015
AOD 550 nm	0.0708	0.0841	0.0747	0.0746	0.0695	0.0743
AE 865–469	0.8696	0.7806	0.7443	0.8959	1.3378	1.3183
SSAOD 550 nm	0.0181	0.0277	0.0250	0.0230	0.0078	0.0102
DUAOD 550 nm	0.0173	0.0188	0.0194	0.0046	0.0010	0.0011
OMAOD 550 nm	0.0041	0.0048	0.0044	0.0089	0.0048	0.0100
BCAOD 550 nm	0.0012	0.0012	0.0009	0.0023	0.0014	0.0015
SUAOD 550 nm	0.0301	0.0315	0.0250	0.0357	0.0545	0.0516

Table 2 Annual mean of Aerosol Optical Depth (AOD) at 550 nm, Ångström Exponent for 865–469 nm, five chemical components at 550 nm: Sea Salt, Dust, Organic Matter, Black Carbon and Sulphate in Hornsund based on reanalysis from the CAMS model.

Hornsund						
	2010	2011	2012	2013	2014	2015
AOD 550 nm	0.0899	0.1113	0.0943	0.0876	0.0804	0.0843
AE 865-469	0.7820	0.7038	0.6594	1.1347	1.6258	1.6194
SSAOD 550 nm	0.0320	0.0481	0.0422	0.0326	0.0143	0.0184
DUAOD 550 nm	0.0174	0.0194	0.0194	0.0049	0.0011	0.0012
OMAOD 550 nm	0.0045	0.0053	0.0044	0.0094	0.0052	0.0085
BCAOD 550 nm	0.0013	0.0013	0.0009	0.0026	0.0015	0.0015
SUAOD 550 nm	0.0348	0.0373	0.0274	0.0382	0.0583	0.0547

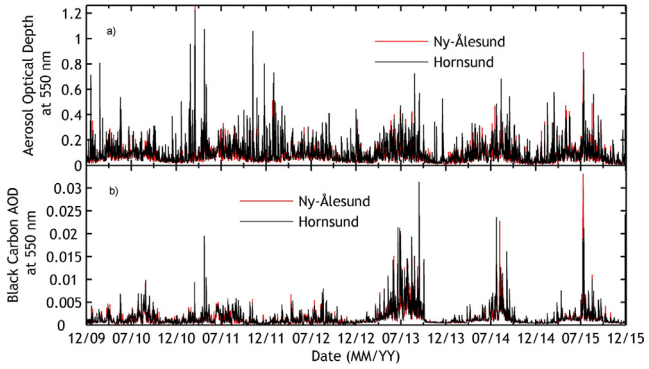

Figure 6 3-hour a) Aerosol Optical Depth at 550 nm and b) Black Carbon Aerosol Optical Depth at 550 nm in Ny-Ålesund (red line) and Hornsund (black line) and Ny-Ålesund (red line) based on reanalysis from the CAMS model during a period 2010—2015. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

et al., 2012), respectively. In May 2006, agricultural fires in Eastern Europe resulted in the highest pollution levels in the Arctic region ever recorded (Myhre et al., 2007; Stohl et al., 2007). In July 2015 Markowicz et al. (2016b) reported about intense Canadian fires over the European Arctic, which was one of the most spectacular biomass burning events in the last decade. It also shows greater averages of AOD at 550 nm (during summer) — 0.0843 (0.1222) in Hornsund than 0.0743 (0.1177) in Ny-Ålesund (Tables 1 and 2, 5 and 6). These fires are also visible in Fig. 6. Following high values of AOD, which indicate potential events in the Arctic (above 0.1, but looking for extremes, above 0.6), we can also see changes in Black Carbon (BC) values. BC is very absorptive at all spectrum ranges, and even more so at longer wavelengths. Dust, on the other hand, becomes less absorptive as the wavelength is increased. Strong values of BC are related to the BB events, due to the fact that it is a component of forest fires. Also, values of AE representing smaller particles, correspond with the size of BB particles. These values are three times higher than the multiannual average this year (0.0015, Tables 1 and 2). These small particles can reach high latitudes much easier, and be transported over vast distances.

Emissions of trace gases and aerosols during fires have significant impact on the climate on a temporal and spatial scale. They are related to growing anthropogenic activities and agricultural fires over the world. Such events have an impact on the chemistry of the atmosphere. Variations in increasing anthropogenic emissions of SO_2 from coal-burning events and industrial activity in Europe and Asia since 2000 have led to the formation of sulphate particle loadings, which have been subsequently transported to the Arctic. This increase is clearly seen over the last years (Fig. 7). These strong peaks in 2012 are probably primarily related to the values of AOD in that period in both stations, and thus also to the strong wind blowing through the fjords at the beginning of the year (Tables 3 and 4). BC and dust are clearly major absorbers in the solar spectrum; they both show large seasonal and interannual variability (Fig. 7). However, their participation in the chemistry of the atmosphere has been decreasing significantly since 2013. There are no significant differences in the chemical composition between the stations, though, sharp increases in each individual value is present at both stations. Regarding Tables 1 and 2 as well as Fig. 7, only the sea-salt component shows some visible variations. From 2010 to 2012 its participation is twice as high in Hornsund than in Ny-Ålesund, and then in 2013—2015 the discrepancy is weaker, but there are still higher values in Hornsund than in Ny-Ålesund. SSAOD values show strong seasonal variations due to a strong correlation with wind speed (see Fig. 7, Tables 3 and 4). Dust emissions show differences smaller than an order of magnitude, the biggest

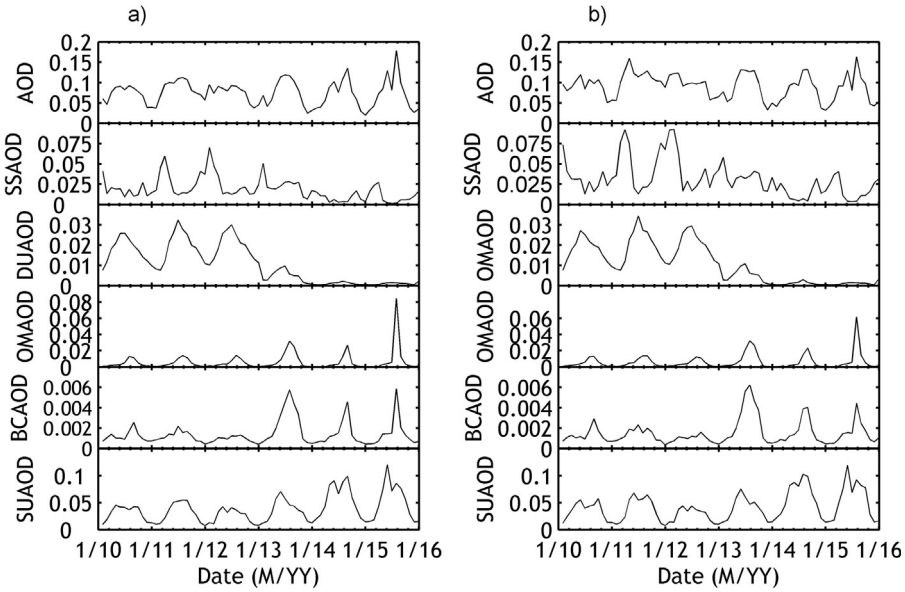

Figure 7 Monthly mean values of AOD at 550 nm, Ångström Exponent for 865—469 nm, 5 chemical components at 550 nm: Sea Salt, Dust, Organic Matter, Black Carbon and Sulphates in (a) Ny-Ålesund and (b) Hornsund from reanalysis from the CAMS model for 2010—2015.

Table 3 Monthly means of wind velocity [m s^{-1}] and direction [0—360°] for Ny-Ålesund, for a period of 2010—2015 based on the WMO database.

Ny-Ålesund mean values wind velocity and direction												
	2010		2011		2012		2013		2014		2015	
	v	Dir.	v	Dir.	v	Dir.	v	Dir.	v	Dir.	v	Dir.
1	4.94	179.01	3.84	173.02	5.71	163.10	4.36	165.83	3.52	180.90	5.07	164.22
2	3.64	158.03	5.14	160.64	5.31	175.15	4.01	162.68	4.53	156.26	5.86	185.21
3	4.11	175.48	5.37	190.00	5.76	163.00	3.83	154.20	4.94	187.68	6.16	169.34
4	3.33	173.42	5.19	172.11	3.37	179.30	3.53	178.94	3.94	200.72	5.31	159.82
5	3.16	188.33	3.33	197.44	4.27	196.90	3.24	196.97	3.53	195.50	2.49	195.13
6	2.67	200.32	2.76	237.49	2.52	214.11	3.84	220.23	2.85	223.72	3.55	211.84
7	2.58	151.07	3.29	157.21	3.21	160.42	3.39	173.96	2.50	178.25	3.44	158.37
8	3.01	183.97	3.42	156.15	4.26	174.73	3.79	159.13	2.47	149.61	2.69	153.42
9	2.55	202.04	2.60	183.89	4.05	165.62	4.13	157.40	4.33	200.73	3.47	189.93
10	4.15	178.09	4.75	170.05	3.24	176.80	3.88	192.81	5.46	161.86	3.88	180.87
11	3.26	177.72	5.04	181.31	4.36	173.38	4.99	180.84	4.46	189.60	6.64	174.85
12	5.06	180.95	5.72	166.23	4.39	172.67	5.34	166.07	6.27	173.97	5.77	154.78

Table 4 Monthly means of wind velocity [m s^{-1}] and direction [0—360°] for Hornsund for a period of 2010—2015 based on the WMO database.

Hornsund mean values of wind velocity and direction												
	2010		2011		2012		2013		2014		2015	
	v	Dir.	v	Dir.	v	Dir.	v	Dir.	v	Dir.	v	Dir.
1	4.94	179.01	3.84	173.02	5.71	163.10	4.36	165.83	3.52	180.90	5.07	164.22
2	3.64	158.03	5.14	160.64	5.31	175.15	4.01	162.68	4.53	156.26	5.86	185.21
3	4.11	175.48	5.37	190.00	5.76	163.00	3.83	154.20	4.94	187.68	6.16	169.34
4	3.33	173.42	5.19	172.11	3.37	179.30	3.53	178.94	3.94	200.72	5.31	159.82
5	3.16	188.33	3.33	197.44	4.27	196.90	3.24	196.97	3.53	195.50	2.49	195.13
6	2.67	200.32	2.76	237.49	2.52	214.11	3.84	220.23	2.85	223.72	3.55	211.84
7	2.58	151.07	3.29	157.21	3.21	160.42	3.39	173.96	2.50	178.25	3.44	158.37
8	3.01	183.97	3.42	156.15	4.26	174.73	3.79	159.13	2.47	149.61	2.69	153.42
9	2.55	202.04	2.60	183.89	4.05	165.62	4.13	157.40	4.33	200.73	3.47	189.93
10	4.15	178.09	4.75	170.05	3.24	176.80	3.88	192.81	5.46	161.86	3.88	180.87
11	3.26	177.72	5.04	181.31	4.36	173.38	4.99	180.84	4.46	189.60	6.64	174.85
12	5.06	180.95	5.72	166.23	4.39	172.67	5.34	166.07	6.27	173.97	5.77	154.78

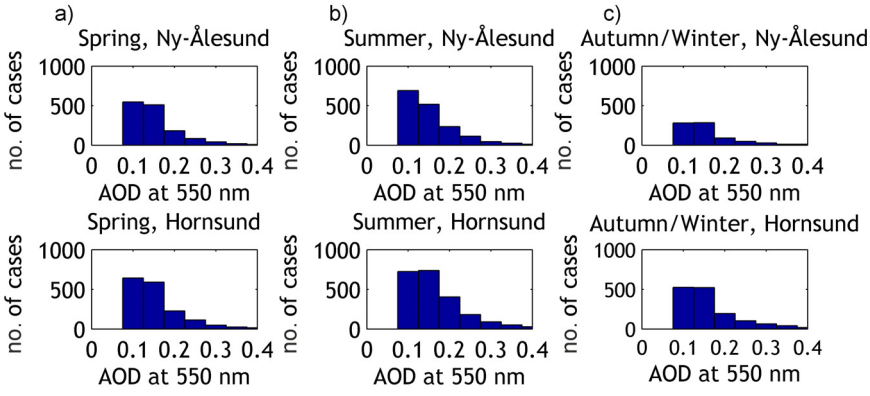

Figure 8 Relative frequency histograms of AOD (550 nm) from CAMS database > 0.1 at Ny-Ålesund (upper plots) and Hornsund (lower plots) from 2010 to 2015 performed into (a) spring, (b) summer and (c) autumn-winter season.

Table 5 Seasonal mean of AOD at 550 nm, Ångström Exponent (440—870 nm or 865—469 nm) from the CAMS database in Ny-Ålesund and Hornsund for a period 2010—2015.

		Ny-Ålesund		Hornsund	
	Season	AOD	AE	AOD	AE
2010	Spring	0.0880	0.9427	0.1036	0.8886
	Summer	0.0885	1.0614	0.1049	1.0099
	Autumn and winter	0.0526	0.7309	0.0750	0.6071
2011	Spring	0.0954	0.9036	0.1220	0.8423
	Summer	0.1040	1.0039	0.1238	0.9780
	Autumn and winter	0.0679	0.6003	0.0993	0.4888
2012	Spring	0.0874	0.9409	0.0957	0.8798
	Summer	0.0828	0.8550	0.1014	0.7735
	Autumn and winter	0.0641	0.5861	0.0900	0.4872
2013	Spring	0.1008	0.9322	0.1139	1.1281
	Summer	0.1048	0.9608	0.1275	1.1811
	Autumn and winter	0.0436	0.8431	0.0532	1.1138
2014	Spring	0.0867	1.5980	0.0910	1.6954
	Summer	0.1070	1.5557	0.1224	1.8074
	Autumn and winter	0.0409	1.0896	0.0528	1.4947
2015	Spring	0.0982	1.6059	0.1034	1.7096
	Summer	0.1177	1.5819	0.1222	1.7185
	Autumn and winter	0.0392	1.0320	0.0545	1.5210

differences (0.006) occur in 2012, DUAOD has a value of 0.0194 in Hornsund and 0.0194 in Ny-Ålesund. Organic matter values have slightly bigger values in 2011 (0.0048) and 2013 (0.0094) in Hornsund, while sulfates — are always greater by about 0.0040 in Hornsund which is about 10% of the value.

Furthermore, a slight increase in stratospheric AODs has recently been observed due to the transfer of sulphate aerosol particles and gases between the troposphere and low stratosphere at tropical latitudes, which has also had impact on polar regions (Tomasi et al., 2015).

Table 6 Seasonal mean of AOD at 550 nm $>$ 0.1, Ångström Exponent (440—870 nm or 865—469 nm) from the CAMS database in Ny-Ålesund and Hornsund for a period 2010—2015.

		Ny-Ålesund		Hornsund	
	Season	AOD	AE	AOD	AE
2010	Spring	0.1491	0.9957	0.1632	0.8645
	Summer	0.1294	1.1089	0.1352	1.0141
	Autumn and winter	0.1657	0.5073	0.1834	0.3967
2011	Spring	0.1569	0.9797	0.1944	0.8354
	Summer	0.1503	1.1177	0.1531	1.0276
	Autumn and winter	0.2056	0.2933	0.2288	0.2653
2012	Spring	0.1360	1.0552	0.1407	0.9116
	Summer	0.1376	0.9517	0.1502	0.7219
	Autumn and winter	0.1893	0.3410	0.2003	0.2942
2013	Spring	0.1604	0.9771	0.1704	1.2195
	Summer	0.1664	0.8934	0.1911	1.2813
	Autumn and winter	0.1460	0.4475	0.1689	0.8721
2014	Spring	0.1517	1.6667	0.1481	1.7560
	Summer	0.1810	1.6359	0.1733	1.9647
	Autumn and winter	0.1371	1.1455	0.1467	1.5996
2015	Spring	0.1631	1.6886	0.1617	1.7825
	Summer	0.2027	1.7255	0.1891	1.9155
	Autumn and winter	0.2002	0.7098	0.1827	1.3137

An Aerosol Optical Depth below 0.1 at 550 nm is usually classified as clean-air in the Arctic, air which was not advected from mid-latitudes. We divided the whole dataset into three seasons: spring, summer and autumn-winter combined. Due to the fact that in situ aerosol measurements can only be carried out during the polar day, these two seasons are mostly statistically irrelevant in comparison to other results. We also wanted to determine where the strongest events occurred, and so we had to single out cases where the AOD was larger than 0.1. AOD $<$ 0.1 in Ny-Ålesund represents 78% of the whole dataset while in Hornsund it is 69%. Then, we divided the AOD values into 13 classes from 0.1 to 1.3. The AODs $>$ 0.4 are not present in Fig. 8, because their percentage is far too low in Ny-Ålesund, where they represent 2.67% of cases without values above 0.1 and in Hornsund it is 2.92%. According to the first paragraph in this chapter, greater AODs persist especially during the spring (Tables 5 and 6). This suggests that the aerosols which are advected to the Artic are

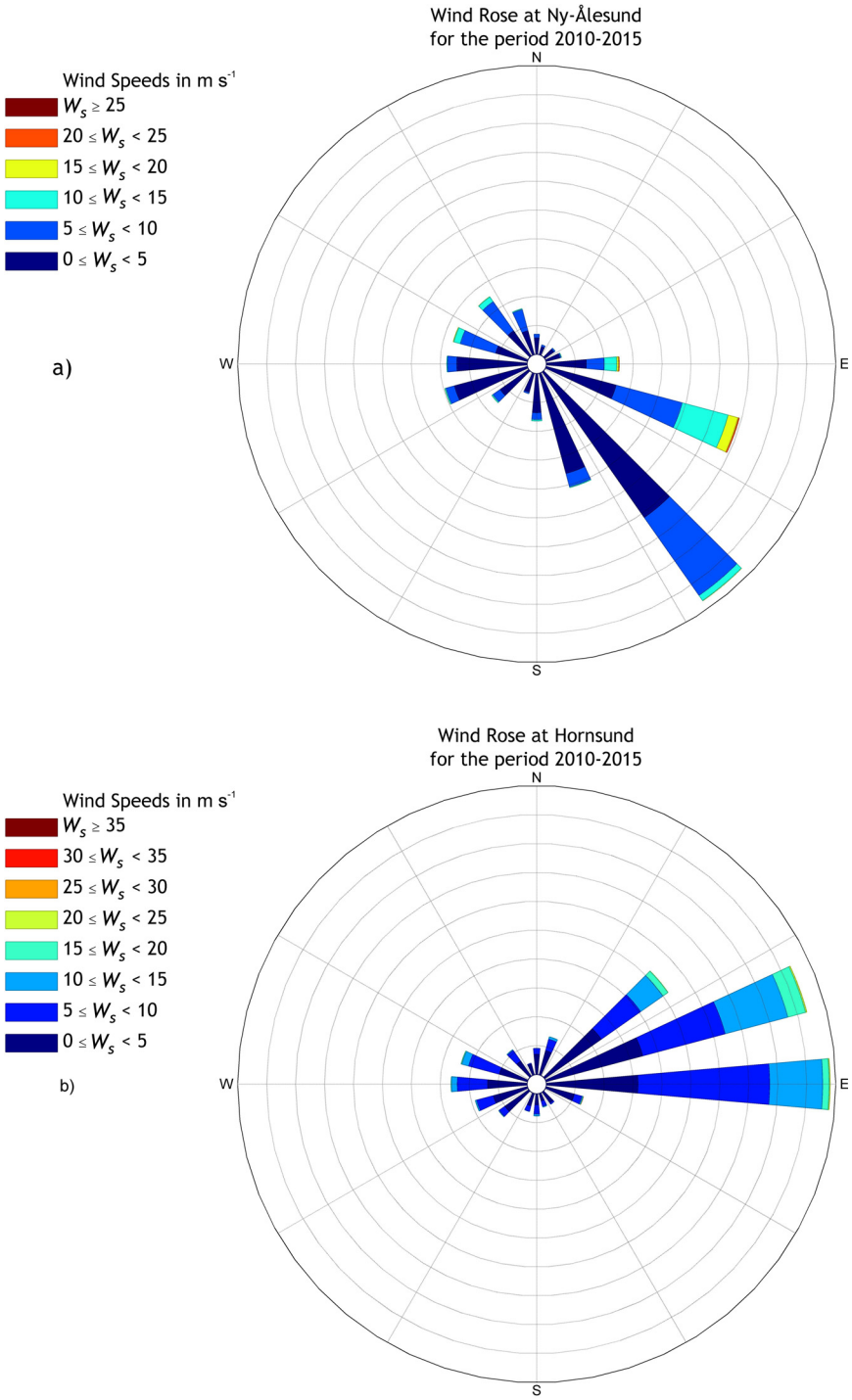

Figure 9 Wind rose (wind speed [m s^{-1}] and direction [0—360°]) for (a) Ny-Ålesund and (b) Hornsund for a period of 2010—2015 based on the WMO database.

enriched by aerosols of continental and anthropogenic origin in midlatitudes. Results without clean-air values explain also higher values in winter and early spring, which result from the haze conditions in the atmosphere, or common BB events. Furthermore, individual year mean values for each season are higher at the Hornsund station. The exception for higher values at the Hornsund station is the instance of small differences in 2014 and 2015 — but only when we take into account the threshold for extreme AODs (AOD $>$ 0.1). We can conclude that during the Haze events differences between the two stations decrease, which is related to the changing conditions of the atmosphere due to transportation of air to the Arctic. In general, seasonal variations of both parameters (AOD and AE) resemble the multi-year pattern.

The main factor regulating the circulation of the atmosphere over the two stations is the orography of the surrounding area. Hornsund receives eastern direction of advection, while the inflow in Ny-Ålesund is from the south-east. It is quite closely related to the influx of aerosols. Strong winds from the south and south-west in Hornsund (i.e. associated mostly with the wind along the fjord) cause the domination of marine aerosols, especially during the period 2011—2012. Due to large areas of open water west of Spitsbergen, emissions of sea-salt and organic aerosols are expected to be dominant. However, the state of the surface waters around Spitsbergen influences the type and the intensity of the regional atmospheric circulation and thus affects the frequency of winds with specific directions, which are recorded at both stations. The presence of a low pressure system west of Spitsbergen causes an increase of eastward winds, particularly on the west coast (Rogers et al., 2005; Tsukernik et al., 2007) (Fig. 9). Although, the frequency of winds from certain directions at the Hornsund station depends on the type and intensity of the regional atmospheric circulation (Marsz and Styszyńska, 2007). In addition, the wind regime, and especially its structure, is heavily influenced by local conditions, especially the position of the station relative to the main elements of the orography. This causes that the wind direction recorded at the stations often does not indicate the actual direction of the flow of air masses.

5. Conclusions

Analyses of the time series of modelled AODs from the CAMS model carried out for Ny-Ålesund and Hornsund show a noticeable decrease in AOD over the study period, leading to the alteration of the state of the polar atmosphere. It is less than 1% (at a linear trend) for each half-year of the model results at both locations. Potential differences observed between the two stations are not statistically significant, it is always a 2% difference between values, and it is always higher in Hornsund for each half-year of observations.

Annual and monthly means of AOD confirm that the mean values are higher in Hornsund than in Ny-Ålesund. The Hornsund station has higher peaks of AE values, with a strong discrepancy in 2014, still, annual means of this parameter represent a big discrepancy between the stations, higher values in 2010—2012 in Ny-Ålesund, and lower in 2013—2015. Results of seasonal means show the highest values in winter and in early spring, which correlates with haze conditions in the atmosphere, or common BB events. Also, individual yearly mean values are higher at the Hornsund station in all seasons.

In general, the increasing values of AOD are related mostly to air masses carrying anthropogenic pollution from midlatitudes. This is most often caused by the Artic Haze, but recently more and more events are related to biomass burning (Markowicz et al., 2016a,b; Moroni et al., 2017; Tomasi et al., 2015). Such events reach both stations at different times, which is related to the circulation and orography of the surrounding area.

There are no significant differences in chemical composition between the stations, although, sharp increases in individual values appear at both stations. Increasing mean values of BC and OM are associated with the increasing number of agricultural forest fires reaching Spitsbergen. During the Haze events differences between the two stations decrease, this is related to the changing conditions of the atmosphere due to the transportation of air to the Arctic. Nevertheless, taking into account the AOD in all spectra we did not find any significant differences between the stations in the period 2010—2015.

Modelling and prediction of aerosol properties is associated with a large degree of uncertainty due to the unpredictable character of the processes which involve aerosols. Nevertheless, the CAMS model is quite successful at reproducing the temporal variability of aerosols and the dominant aerosol types over the two chosen stations, as described in this paper. The results obtained from modelling with the CAMS (Copernicus Atmosphere Monitoring Service) were used to prove that the Arctic model could correctly estimate Arctic events and provide accurate aerosol information for the area of Svalbard. Previous comparisons with the MODIS derived AODs, the NAAPS model and in situ data shows that all models overestimate the values of AOD. However, including the AOD from the MODIS observations significantly increases the consistency between the model and the NAAPS derived AODs. Nevertheless, the CAMS model provides a good reconstruction of real values of AOD, especially during the occurrence of changes in the atmosphere and the subsequent changes in the composition and concentration of aerosols (Markowicz et al., 2016a).

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enabled the development of a large range of products dedicated to analysing the composition of the atmosphere and is also an essential asset for a better understanding of climate change.

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