Atmospheric black carbon in Svalbard (ABC Svalbard)

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1. Introduction

Black carbon (BC) is a component of submicrometer aerosol particles, characterized by atmospheric residence times of a few days up to a week. This relative longer life-time, compared to coarse particles, allows BC transport from source regions to remote locations. Once in the Arctic, BC impacts the regional radiation balance by absorbing incoming solar radiation (direct effect), altering cloud distribution and their radiative properties, and reducing snow and ice surface albedo after deposition. In addition, BC at mid-latitudes can still warm the Arctic, affecting the transport of heat towards higher latitudes, with the strongest impacts in summer and in the upper troposphere (Sand et al. 2013).

BC radiative direct effect in the Arctic is enhanced compared to mid-latitude impacts, due to the prolonged solar light exposure during summer and the high reflectivity of lower atmosphere and surface, characterized by low level clouds and snow/ice cover, respectively (Quinn et al. 2011). The top of the atmosphere radiative forcing due to BC direct effect calculated using different modelling tools and approaches ranges between +0.12 and +0.80 W m^{-2} (Quinn et al. 2011; Quinn et al. 2015 and references therein). Differences among results are mainly driven by BC atmospheric burden (Quinn et al. 2011), particle atmospheric residence time (Wang et al. 2014), cloud distribution, and treatment of radiative processes (Quinn et al. 2015). In addition, current model uncertainties are strongly affected by lack of constraints on BC vertical distribution and a simplified description of the BC radiative properties (Koch and Del Genio, 2010; Boucher et al. 2013).

In addition to direct radiative forcing, BC aerosol particles can alter the Earth's energy budget participating to cloud formation. Although freshly emitted BC particles are hydrophobic, atmospheric processing promotes BC particle coating by soluble components, that eventually make BC particles hydrophilic enough to act as cloud condensation nuclei (CCN) (Park et al. 2005). Promoting cloud nuclei formation, BC might increase cloud thickness and cloud residence time, leading to a negative radiative forcing (Boucher et al. 2013). On the other side, atmospheric heating induced by cloud interstitial BC can favour cloud evaporation (Jacobson et al. 2010), suppressing precipitation and thus reducing BC wet removal. Conversely, the ability of BC particle to act as ice nuclei (IN) is still under debate (Xu et al. 2019). Climate response driven by BC-cloud interaction is characterized by large uncertainties, mainly driven by limited number of modelling studies and limited observational constraints (Bond et al. 2013; Xu et al. 2019).

After removal from the atmosphere through wet and dry deposition, BC can still affect the climate by darkening snow and ice surface (albedo reduction) and promoting their melting. Even a small number of BC particles has a significant impact on snow and ice, since BC has a mass absorption cross-section up to five times higher than snow. In addition, ice and snow crystals reflect solar radiation, increasing the light optical path, and enhancing the probability

of interaction between light and BC embedded in snow and ice (Quinn et al. 2011). Finally, snowpack warming promotes metamorphism, which leads to larger snow crystal formation and thus further albedo reduction (Ginot et al. 2014; Hadley and Kirchstetter 2012). These effects can trigger a feedback mechanism: earlier snow melting during the warm season decreases the surface albedo and increases the fraction of solar energy trapped by the atmosphere. To estimate BC albedo forcing, models need to simulate BC concentration and light absorption in the upper layers of the snow surface. BC concentration in snow depends on BC concentration in snowfall, BC settled through dry deposition, snow melting/sublimation, and BC runoff (Doherty et al. 2014). In addition, the BC mass absorption cross section in snow depends on particle age (Schwartz et al. 2013). Using a multi-model approach and BC observations, Jiao et al. (2014) calculated that the regional BC albedo forcing was equal to +0.17 W m^{-2} (Jiao et al. 2014). Based on direct observations of BC in snow samples, Dang et al. (2017) drew comparable results, with BC albedo forcing in the Arctic ranging between 0.06 and 0.5 W m^{-2} considering old and fresh snow, respectively.

Although recent improvements, models that are used to quantify BC climate impacts and related temperature changes in the Arctic often underestimate the actual BC concentrations and fail in reproducing the amplitude of its seasonal variation (Winiger et al. 2017). Actual sources of BC in the Arctic are still characterized by some degree of uncertainty. The integration of chemical transport models and aircraft observations indicated that anthropogenic sources dominated BC in the Arctic lower troposphere, with Russian emission accounting for the largest BC share (Wang et al. 2011; Popovicheva et al. 2017). Sand et al. (2016) calculated that BC is responsible for the increase of Arctic temperature by 0.48 K, with the highest impacts from domestic emissions from Asia and flaring from Russia, while observations in the Siberian Arctic would suggest an overestimation of flaring emissions (Winiger et al. 2017). Finally, radiocarbon characterization of carbonaceous aerosol indicated that the actual emission inventories tend to underestimate the contribution of biomass burning (Winiger et al. 2017; Winiger et al. 2019). Future BC emission scenarios are even more unclear, since projections depend on changing local anthropogenic activities in the Arctic and on changing transportation pattern of BC from low-latitudes (Winiger et al. 2017). Future changes in wildfires frequency in warming climate are also undetermined.

To summarize, the description of BC impacts on climate and air quality in the Arctic requires an accurate understanding of BC optical and microphysical properties, BC aerosol-cloud interaction, wet and dry deposition, and sources (Mahmood et al. 2016; Winiger et al. 2017). The analysis of BC spatial and temporal variability can help to understand how changes in climate and anthropogenic activities affect BC concentration and can support the analysis of BC climate-relevant properties. The specific aims of this chapter are: i) to quantitatively describe temporal variability of BC concentrations at different altitude sites based on long-term measurements, ii) and to deploy available short-term observations to describe horizontal and vertical variability of BC concentrations.

2. Overview of existing data

2.1 Long-term observations at Svalbard

BC is defined operationally, based on the measurement techniques employed for its quantification (Lack et al. 2014). Long-term observations at Svalbard deploy optical techniques to infer BC concentration: aerosol particles are deposited on a filter and change in light transmission through the filter is measured. These techniques allow the quantification of equivalent black carbon (eBC), namely the amount of strongly light absorbing carbon, with optical properties similar to those of soot, that would lead to the same absorption signal (Andreae and Gelencser, 2006). eBC mass concentration is then derived by multiplying the measured light absorption coefficient by an appropriate mass absorption cross section (MAC), which needs to be specified when eBC is reported (Petzold et al. 2013).

The quantification of eBC through filter-based optical measurements relies on a few assumptions. First, light absorption is assumed to be due exclusively to BC particles, ignoring the contribution of organic aerosol (brown carbon) and dust. Observations indicate that brown carbon and dust absorptions have a stronger wavelength dependence than BC, and are higher in the UV and visible part of the spectrum (Bergstrom et al. 2007; Russell et al. 2010). As a consequence, this report focuses on measurements at wavelength larger than 600nm, where BC light absorption dominates over other species (Kirchstetter et al. 2004). In addition, liquid-like organic aerosol can spread across the filter substrate after collection, altering filter optical properties and particle morphology, and consequently decreasing accuracy of eBC quantification (Subramanian et al. 2007; Lack et al. 2008). Such a bias depends on the type of sampled air masses and can be relevant (larger than 10%) in polluted environments (Lack et al, 2008). This artefact is likely negligible in the Arctic. Finally, to convert absorption into eBC concentration, an accurate MAC value is required. MAC of BC depends on the particles' diameter, morphology (fractal or compact shape), and coating by non-absorbing materials, thus it varies while the particles reside in the atmosphere and is higher far from the source regions (Sharma et al. 2004; Bond et al. 2006). Measurements of BC MAC in the Arctic are consistently higher than those of bare BC particles (Sharma et al. 2017; Zanatta et al. 2016; Zanatta et al. 2018), and range between 7.5 and 9.9 $m^2\ g^{-1}$ at 550 nm. Due to the variability of MAC with aerosol age, here we compare long-term measurements of light absorption coefficients without converting them into eBC concentration.

Long-term light absorption measurements at Svalbard are performed at Gruvebadet and Zeppelin. Gruvebadet observatory (78.918°N, 11.895°E; 61m above sea level) is located 800m south-west of the Ny-Ålesund research village. The Zeppelin observatory (78.908°N, 11.881°E; 474 m above sea level) is located at the top of the Zeppelin mountain, about 10

km from the coast and 5 km from Gruvebadet observatory. The Zeppelin Observatory is owned and managed by the Norwegian Polar Institute and is part of the Global Atmospheric Watch network.

At Zeppelin aerosol light absorption measurements were performed with a single wavelength aethalometer AE-9 in 1998 and in 1999, operating at 880 nm. Starting from 2001, light absorption has been measured continuously at seven wavelengths, ranging from 370 nm to 950 nm, with a multi-wavelength aethalometer AE-31. Both instruments worked at 30-minute time resolution (Eleftheriadis et al. 2009). Light attenuation measurements were corrected for filter transmissions according to Backamn et al. (2017) and using a multiple scattering correction factor C equal to 3.25. Light absorption at 525 nm has been measured with a Particles Soot Absorption Photometer (PSAP) since 2002. Since June 2015 an Aethalometer AE33 is running in parallel with the AE31. Work is currently in progress to evaluate the performance of the two instruments, with respect to other parallel absorption measurements. It is aimed to consolidate the use of loading and multiple scattering correction factors and provide quality assured absorption coefficient time series at the Arctic sites. At Gruvebadet aerosol light absorption is measured at three wavelengths with a Radiance Research PSAP at 1-minute time resolution (Bond et al. 1999), since 2010. Data are then averaged over 1-hour period. Absorption coefficients are measured at 467 nm, 530 nm, and 660 nm, with a precision ranging between 20 and 25%. Measurements are performed generally from April to September, with a limited number of data during the winter season. Measurements are corrected according to Bond et al. (1999) for filter transmission, flow, and sampling filter area. Data are not corrected for aerosol scattering, while shadowing effect is considered negligible due to the low aerosol loading. At both sites absorption coefficients are normalized at standard pressure and temperature conditions (1 atm and 0 °C).

Figure 1 (panels a and c) shows the time series of daily-averaged Zeppelin and Gruvebadet aerosol absorption coefficients at 660 nm, while panels 1b and 1d report the seasonal capture of light absorption at the two sites. The wavelength of 660 nm is chosen to minimize the interference of brown carbon and dust, and since it is a common wavelength to aethalometer and PSAP instruments. Although the variability range of the two time-series is comparable, Gruvebadet shows often short episodes of high absorption coefficient values, likely due to the influence of local emission sources at Ny-Ålesund village and harbour, constrained in the lower layers of the troposphere and not affecting the high-altitude site. The annual average absorption coefficient at Zeppelin from 2005 to 2018 was 0.13 Mm^{-1}. Over the period 1998-2007 Eleftheriadis et al. (2009) measured an annual average eBC concentration of 39 ng m^{-3}. Assuming a MAC at 660 nm in the range of 6.2-8.2 (extrapolated at 660 nm from Zanatta et al. 2018 and assuming an absorption angstrom exponent of 1), the average eBC observed during the period 2005-2018 is equivalent to 16-20 ng m-3, in agreement with the decreasing tendency during the 2000s reported by

Stone et al. (2014) for multiple sites in the Arctic. The average absorption coefficient over spring and summer, when measurements are representative of both sites, were 0.15 Mm^{-1} and 0.28 Mm^{-1} at Zeppelin and Gruvebadet, respectively, indicating higher concentration of BC at the lower altitude observatory.

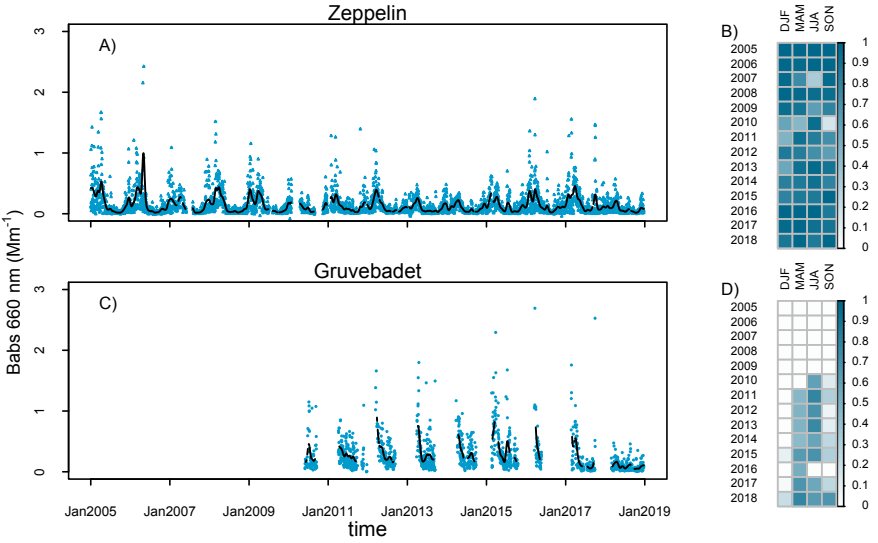

Figure 1. Time series of daily average aerosol light absorption coefficients at 660 nm from aethalometer measurements at Zeppelin (A) and PSAP measurements at Gruvebadet (C); the black line in each panel indicates the seasonal trend from the application of a Kolmogorov-Zurbenko filter. On the right, the seasonal data coverage of hourly light absorption coefficient measurements at Zeppelin (B) and Gruvebadet (D) is shown.

The black lines in Figure 1 indicate the seasonal trend of the light absorption coefficient at 660 nm, calculated with the Kolmogorov-Zurbenko (KZ) filter on daily average data. $KZ_{(m,p)}$ filter is a low-pass filter obtained by applying a moving average with a window length of m days repeated for p iterations (Wise and Comrie, 2005). To isolate the seasonal trend, m and p were here set equal to 15 and 5, respectively. Aerosol absorption coefficients show a clear seasonality, with the highest values in spring, and the lowest records in summer and fall. The observed trend is due to the enhanced transport of pollution from mid-latitudes in winter and spring (Sharma et al. 2004) and higher efficiency of wet removal in summer (Shen et al. 2017). The Gruvebadet seasonal trend showed maxima that were synchronized with those observed at Zeppelin, although with values up to 3 times higher. Notably in 2018, when both sites were characterized by significant lower concentrations of eBC compared to previous years, the seasonal trends overlapped well.

2.2. eBC Spatial variability

A climatological approach to BC and eBC distribution within Svalbard is difficult because of scarcity of long-term measurements. Nevertheless, short-term observations have been reported for several locations across the islands. eBC measurements were performed during cruises (ship measurements, e.g. Ferrero et al. 2019) and short-term intensive field campaigns (e.g. Zhan and Gao 2014; Chen et al. 2016; Ferrero et al. 2016, Lisok et al. 2016). Existing observations indicate the importance of local sources to explain spatial variability of eBC at local scale. Major land-based anthropogenic sources are located in the central part of the Spitsbergen. They are related to the settlements of Barensburg, Longyearbyen, Sveagruva, Ny-Ålesund and mining activities at Barensburg (up to 2007), Longyearbyen and Sveagruva. Occasionally, local dust storms can also be a source of absorbing aerosols (Dörnbrack et al, 2010), although it is not clear if they play a significant role in the overall aerosol characteristics of the Svalbard area.

Results of eBC (photoacoustic extinctiometer) observations in Longyearbyen, carried out within the iAREA2014 campaign (the Impact of Absorbing aerosols on Radiative forcing in the European Arctic) in spring 2014, indicate a significant impact of the local anthropogenic pollutions (Lisok et al. 2016). The absorption coefficient showed a distinct diurnal cycle with the day time maximum about 3-4 times larger than the background night values, which implies local emissions between 05:00 and 21:00 UTC. The values measured during night-time (0.6 ± 1.2 Mm^{-1}, 26 Mar - 14 Apr) were close to Ny-Ålesund levels (0.8 ± 0.4 Mm^{-1}).

The research vessel r/v Oceania (Institute of Oceanology Polish Academy of Sciences) has carried out atmospheric aerosol measurements along Svalbard every summer since 1987. In the years 2011 and 2012 (Ferrero et al. 2019), 2014, 2015, 2016, 2018, and 2019 measurements of eBC and/or EC were also performed. The EC concentrations measured in 2011-2012 (35±1 and 25±1 ng m^{-3}, respectively) were close to the eBC concentrations measured during the AREX2018 (ARctic EXperiment) cruise (Ferrero et al. 2019). The data from 2014-2016 AREX campaigns need correction for elevated relative humidity of filter samples (40-60%). Measurements which allow to estimate the impact of relative humidity on the eBC values and to optimize a correction algorithm are planned for AREX2020. During AREX2018 a considerable increase in eBC concentration near Svalbard was measured at latitudes 76-78° N (about 60-100 ng m^{-3}) when compared to seas north and south of these latitudes (10-20 ng m^{-3}), with the largest concentration near the southern coast of the archipelago (75.5-76.5 ° N). The most elevated concentrations located south of Svalbard suggest that the Black Carbon emission from marine transport may be one of the main sources. Ship emission impact on eBC is most probably limited to coastal regions and should not influence the interior of Svalbard.

The contribution of local sources, both land-based and ship emissions to the mean concentration of atmospheric eBC over Svalbard is still an open question. However, the observed east–west gradient of elemental carbon (EC) concentrations in snow over Spitsbergen (Forsström et al. 2009) suggests that the impact of the long-range transport on the atmospheric BC concentration is dominant. The snow EC gradient is the outcome of a combination of the elevated BC in air masses advected to Svalbard from the eastern sector, "the orographic effect of the archipelago, and the efficient scavenging of the carbonaceous particles through precipitation".

Recently new techniques have been developed to study the spatial distribution of eBC in local scales. Spolaor et al. (2017) presented an innovative approach to characterize concentration of atmospheric aerosol particles and air mass layering along the elevation profile of glaciers. This methodology has been employed during the BC-3D campaign (Cappelletti et al. 2019) in the Spitzbergen and Hornsund regions at Svalbard. The general observed phenomenology points at an accumulation of eBC in the lower sectors of the glaciers. Low weight and fast response sensors (microaethalometers) and a miniature Diffusion Size Classifier (miniDisc) deployed on a snowmobile were used. Measurements by means of small unmanned aerial system (UAS) carrying microaethalometers are also promising in fjord/local scales (e.g. Chiliński et al. 2019).

2.3 eBC vertical distribution

Only few experimental data on eBC vertical distribution are reported in the literature. These results have been obtained mainly with remote techniques (sun-photometers) or by in-situ observation (soot photometers and microaethalometers) deployed on aircraft, helicopters, or tethered balloons (Schwarz et al. 2010; Spackman et al. 2010; Kupiszewski et al. 2013; Bates et al. 2013; Ferrero et al. 2016; Mazzola et al. 2016; Markowicz et al. 2017).

The springtime PAM-ARCMIP (Stone et al. 2010) and HIPPO (Schwarz et al. 2010) campaigns showed high BC concentrations close to the ground, below the thermal inversion, but also dense pollution and BC at high altitudes over the Arctic. Interestingly, the PAM-ARCMIP results show a decrease of eBC compared to past measurements (i.e., AGASP; Hansen and Novakov, 1989). In addition, the HIPPO campaign revealed that in the lower troposphere the eBC vertical gradient can change seasonally from positive to negative (Schwarz et al. 2013). Spackman et al. (2010) reported eBC located mainly in the Arctic free troposphere with a positive gradient in the lower troposphere.

Regular campaigns of aerosol vertical profiles have been conducted at Ny-Ålesund at the Gruvebadet observatory in the periods 2011-2012 and 2014-2018 and at the AWIPEV observatory in the period 2015-2017 by deploying aerosol instrumentation on tethered

balloons that are able to sound the Arctic troposphere continuously at high resolution up to about 1.5 km (Moroni et al. 2015; Ferrero et al. 2016; Moroni et al. 2016; Mazzola et al. 2016; Markowicz et al. 2017). The field campaigns were manly performed in spring, and in fall in 2014 (Mazzola et al. 2016). Recently, the first winter vertical profiles in the Arctic have been reported (Nakoudi et al. 2019). eBC measurements were mainly based on the AE51 Magee Microaethalometer, a light, single wavelength (880 nm) filter-based instrument. In the 2019 winter campaign a PSAP was also deployed on the AGAP payload and intercompared with the AE51.

Spring and summer eBC profiles were compared for the period 2011-2012 (Ferrero et al. 2016). Homogeneous profiles have been observed only for 15% of the cases in spring, while they dominate (37%) in summertime. 20% of the spring profiles showed an increase of the eBC concentration with elevation, with frequent layers of BC observed around and above 1 km of altitude and attributed to long-range transport during the Arctic Haze. Averaged spring eBC concentrations exceed 5 times the summer ones. When big ships were present in the Kongsfjord strong increases (up to 40-fold) have been observed for BC concentration. This increase was observed mainly at lower altitude and for short times, i.e. before ship plume dispersion took place (Ferrero et al. 2016).

3. Unanswered questions

Long-term trends of atmospheric BC concentration show a clear seasonality, associated with long (years) and short time-scale (hours) variability. The dynamics of BC entrainment in the Arctic boundary layer is still a poorly understood process and may have a wide variability depending on local conditions. The differences in light absorption observed between Gruvebadet and Zeppelin can help to investigate such a dynamic. The two sites are located at different altitudes, but at close distance. A deeper analysis of BC (or light absorption) variability on short and long time scale, together with vertical profile measurements of BC, meteorological variables and particle number concentration (Mazzola et al. 2020) can help to better understand the impact of local sources and long-range transport on BC vertical structure. Cyclonic disturbances, low clouds dynamics and orographic effects may be some of the important factors to be studied in order to reduce the overall large uncertainties. In addition, extending the data coverage at Gruvebadet to the winter months would be beneficial for this analysis.

The year-to-year variability of light absorption observed at the two long-term monitoring sites can support a deeper investigation of all the factors affecting BC variability at Svalbard, and in the Arctic in general. BC atmospheric concentration in the Arctic region is controlled by BC emissions at high and middle latitudes, local meteorology and large-scale circulation, and BC removal efficiency through wet and dry deposition, which in turn are controlled by BC microphysical properties and meteorology. A better understanding of all the factors controlling BC atmospheric concentration and vertical distribution is a key element to improve the model ability to describe BC climate impact and reduce uncertainty of future climate scenarios.

Emission inventories indicate that implementation of climate and air quality policies in Europe and United States have efficiently contributed to fossil fuel BC emission reductions over the last 40 years. At the same time, emissions of BC from wood burning from residential heating in winter and exceptionally high emissions of BC from forest fires in summer have increased over the last 20 years due to a larger use of biofuels and increased frequency of wildfires (Chin et al. 2014). Although these changes are expected to translate non-linearly into changes in atmospheric concentration, the limited temporal and spatial coverage of existing atmospheric BC measurements cannot track these trends, specifically relevant for atmospheric and climate model validation. Sediments and ice are historical archives of atmospheric composition changes. The use of such archives requires an accurate knowledge of the BC deposition fluxes and all the mechanisms able to modify BC concentration in snow, including melting-freezing cycles and wind scouring of surface winter snow. BC long-term measurements, together with glaciers and snow monitoring activities (Schuler et al. 2020) are extremely valuable tools to better understand the link between atmosphere and cryosphere composition.

In addition to primary or processed combustion particles, secondary particles from nucleation events represent a potentially increasing source of particles in the Arctic, triggered by the sea ice melting. Long-term BC monitoring could be useful to identify the presence of anthropogenically influenced air masses during the year, linking the analysis of new particle formation events with air mass origin (Sipilä et al. 2020).

4. Recommendations for the future

- Improving the comparability and accuracy of atmospheric BC measurements, by understanding the differences among different monitoring methods (PSAP, MAAP, aethalometer, SP2) and by improving the existing correction algorithm for aerosol light absorption measurements with optical techniques. The development of a correction algorithm specifically designed and tested in the Arctic conditions, i.e. low aerosol loading and high single scattering albedo, is advisable. Experiments where different measurement techniques are co-located will support the achievement of these goals.
- Increasing space and time coverage of BC vertical profile measurements, especially during winter. Developing efficient methodology for continuous monitoring of the vertical profiles of optical properties of aerosols on a global and regional scale would improve the knowledge of BC climate impact in the Arctic.
- Promoting dry and wet deposition measurements of BC on snow surface and supporting simultaneous and long-term BC measurements in the atmosphere and cryosphere to develop reliable parameterizations of BC wet and dry deposition, BC impacts on snow and ice albedo, and to support the use of historical BC records in snow and ice to reconstruct atmospheric BC trends. The development of common discussion platforms and integrated database is recommended.
- BC-cloud interaction is a key factor affecting BC climate impact. Starting from 2015 light absorption coefficient of eBC at Zeppelin is measured with a Multi-Angle Absorption Photometer (MAAP) both inside cloud droplets and in the cloud interstitial aerosol. Similar measurements able to quantitatively describe the ability of BC particles to act as cloud condensation nuclei and ice nuclei, together with the analysis of atmospheric processes altering such ability in the Arctic, are needed.

5. Data availability

Data discussed in this report include: long-term measurements of aerosol light absorption coefficients measured at Zeppelin and provided by NCSR Demokritos (http://ebas.nilu.no/), and long-term measurements of aerosol light absorption coefficients at Gruvebadet and provided by ISP-CNR (Italian Arctic Data Centre IADC database).

Dataset	Parameters	Period	Location or area	Dataset landing page	Comment
Gruvebadet time series	Light absorption coefficients	2010-07-01 2018-12-31	Svalbard, Gruvebadet observatory	http://iadc.cnr.it/cnr/metadata_view.php?id=75	Data are available upon request. Contact: stefania.gilardoni@cnr.it
Zeppelin time series	Light absorption coefficients	2005-01-01 2018-12-31	Svalbard, Zeppelin observatory	http://ebas.nilu.no/	

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