



Aerosol black carbon in the European Arctic: Measurements at Zeppelin station, Ny-Ålesund, Svalbard from 1998–2007

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Received 19 August 2008; revised 13 October 2008; accepted 1 December 2008; published 24 January 2009.

[1] An aerosol black carbon (BC) data set from ongoing measurements at Zeppelin station (474 m asl), near Ny-Ålesund (10 m asl), Svalbard is reported for the period 1998–2007. Annual average and median BC concentrations were 39 and 27 ng m⁻³, respectively, while monthly averages ranged from a maximum ~80 ng m⁻³ in February/March to a minimum 0–10 ng m⁻³ from June to September. BC concentrations were calibrated by elemental carbon thermo-optical analysis. PSCF trajectory analysis indicated that BC concentrations at Zeppelin were mainly influenced by source regions in northern and central Russia. Since 2001, a small decreasing trend in BC concentration (–9.5 ng m⁻³ per decade) for the period to 2007 has been observed which is, however, similar to the detection limit. **Citation:** Eleftheriadis, K., S. Vratolis, and S. Nyeki (2009), Aerosol black carbon in the European Arctic: Measurements at Zeppelin station, Ny-Ålesund, Svalbard from 1998–2007, *Geophys. Res. Lett.*, *36*, L02809, doi:10.1029/2008GL035741.

1. Introduction

[2] Black or elemental carbon (BC, EC) aerosol is one of a number of atmospheric constituents considered to be an important contributor to climate change in Arctic regions. It has been shown that BC aerosols can decrease the albedo of snow/ice, resulting in a global estimated radiative forcing of $+0.10 \pm 0.10 \text{ W m}^{-2}$ [Hansen and Nazarenko, 2004]. However, long-term BC measurements have so far only been conducted at few Arctic locations. Sharma *et al.* [2006] reported updated BC data sets for the Global Atmosphere Watch (GAW) stations at Alert (82.39°N, 62.3°W; 210 m asl) and Barrow (71°N, 156.6°W; 8 m asl) over the 1989 to 2003 period. In contrast, measurements in the European Arctic at Ny-Ålesund, Svalbard (10 m asl; 78°54′N, 11°53′E; GAW site) were only conducted for sporadic periods from 1979 to 1990, and continuously from 1990 to 1992 [Heintzenberg and Leck, 1994]. In order to determine a high-resolution, long-term BC aerosol climatology for the European Arctic, results from ongoing measurements at Zeppelin station (474 m asl) near Ny-Ålesund, are reported here. The seasonal variation in BC for the period from August 1998 to spring 2007 is described with respect to potential source regions estimated by means of a Potential Source Contribution Function (PSCF) model based on calculated air mass trajectories [Polissar *et al.*, 2001, and references therein].

2. Methods

[3] A brief description of the aerosol sampling technique is first considered. A common inlet (snow-hood and stainless steel duct) is used to sample ambient air, which attains laboratory temperature (~21°C) before reaching the measurement instruments, hence ensuring measurement at a relative humidity RH < 20%. Losses due to diffusion were estimated at <4% due to the high flow rate (>35 lpm) in the inlet line, and hence corrections are not made. As the inlet is not size-specific, the total aerosol size distribution is sampled even during in-cloud periods. Aethalometer BC measurements were conducted from August 1998 to July 1999 with an AE-9 model (peak $\lambda \sim 880 \text{ nm}$; Magee Sci., manufacturer calibration $\alpha_{AP} = 19 \text{ m}^2 \text{ g}^{-1}$). After a major renovation of Zeppelin, BC measurements only continued in May 2001 but this time with the newer AE-31 model (7-wavelength; only $\lambda = 880 \text{ nm}$ reported here). Both instruments measured with a time resolution of 30 minutes. A detection limit of $\sim 10 \text{ ng m}^{-3}$ ($3 \times$ standard deviation) was determined from measurements of filtered (particle-free) ambient air. Local pollution events, most probably from occasional cruise ship visits to Ny-Ålesund, were identified and flagged by means of a basic objective algorithm. As a result of local pollution (0.2% of all cases), and instrument failure etc (7.4%), the BC data set is at present 92.4% complete.

[4] BC and EC are operationally defined as the light absorbing and thermal refractory fraction of carbonaceous aerosol, respectively. Corrections were applied to the AE-9 and AE-31 measurements in order to obtain a data set of equivalent BC values based on a thermo-optical EC method. The aethalometer mass absorption coefficient (α_{AP}) relates the optical attenuation through the filter (pre-fired quartz fiber) with the BC concentration. This factor needs to be empirically determined for each site, and a thermal EC analysis was conducted on a representative set of AE-9 filters and thermo-optical analysis on samples obtained in parallel to AE-31 measurements [Nyeki *et al.*, 2005b, and references therein]. Site-specific values $\alpha_{AP} \sim 15.2 \text{ m}^2 \text{ g}^{-1}$ ($n = 11$) and $\alpha_{AP} \sim 15.9 \text{ m}^2 \text{ g}^{-1}$ ($n = 23$) were determined for the AE-9 and AE-31 (for $\lambda = 880 \text{ nm}$), respectively. This calibration implicitly takes the aethalometer wavelength sensitivity into account. As the definition and measurement of BC and EC is still an unresolved issue, it should be noted that the above α_{AP} values are current “best-estimate” values. The uncertainty is reported as $\pm 8\%$, being the standard error of the AE-31 vs thermo-optical BC calibration. A more extensive EC thermo-optical analysis will be conducted in a future study. Further uncertainty may arise from the EC analysis method. For instance, Junker *et al.* [2006] found that a thermal and thermo-optical method gave different α_{AP} values in a study of the 1989–2003 BC data

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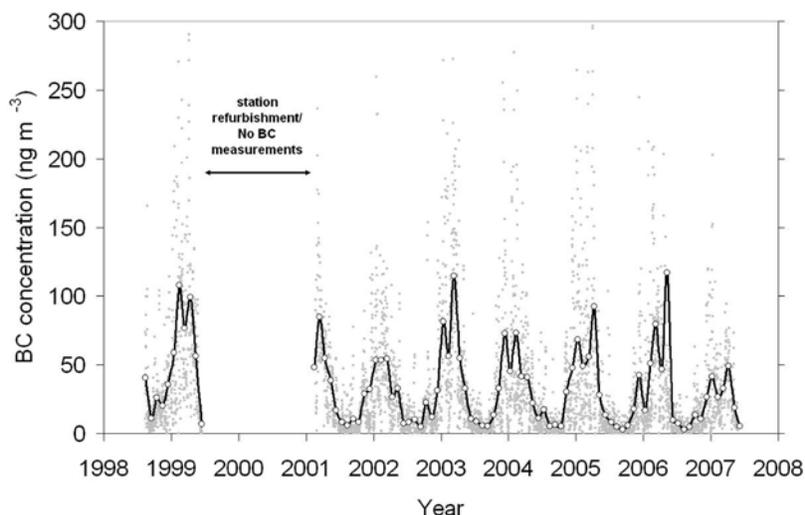


Figure 1. BC time-series at Zeppelin station from 1998 to 2007. 12-hr (gray points) and monthly (black line) averages are shown.

set (Aethalometer AE-8 and AE-9 models) at Mace Head, Ireland. Uncertainty in the absolute BC value derived may be similar or even larger than the uncertainty given above. BC data reported here are for standard conditions (1013 hPa, 273.15 K). BC 30-minute data were used to construct 12-hr average values at 06:00 and 18:00 UTC.

[5] Back-trajectory analyses were conducted using the NOAA HYSPLIT model [Draxler and Rolph, 2003] with a $1^\circ \times 1^\circ$ resolution. The model was run for Zeppelin altitude using twice daily 5-day back-trajectories at 06:00 and 18:00 UTC. A PSCF model [Polissar *et al.*, 2001, and references therein] was used to determine potential source regions of measured BC concentration at Zeppelin. The end-points of back-trajectories from May 2001 to the present were found to have origins not further south than 42°N . End-points are considered the locations of the air mass calculated by the model for every hour past the start time. The northern hemisphere from 42° to 90° was divided into $3^\circ \times 3^\circ$ grid cells and the number of end-points found to correspond to BC concentrations exceeding the 80th percentile BC value, were considered to represent potential emission source areas with their contribution to the measured BC at Zeppelin defined as follows: If n_{ij} is the number of endpoints found in a cell and m_{ij} the points for which the BC concentration criterion is fulfilled, then the conditional probability for this cell to be a source of BC is $\text{PSCF}_{ij} = m_{ij}/n_{ij}$. A weighting function equivalent to that used by Polissar *et al.* [2001] was used to reduce the uncertainty in the results introduced by geographical areas with sparse trajectory coverage. End-point population thresholds were introduced by taking into account the size of the data set. This approach excluded data from the analysis which were not statistically significant. PSCF probabilities have not been calculated for regions north of 72.5°N as this region is predominantly oceanic.

3. Results and Discussion

3.1. Aerosol BC Time-Series

[6] Black carbon measurements from August 1998 to July 2007 are shown in Figure 1 as 12-hr and 1-month averages. The winter-spring months exhibit frequent epi-

sodes of high BC concentrations (maximum $\sim 300 \text{ ng m}^{-3}$) lasting as long as several days. In comparison, summer-autumn BC concentrations were generally $< 20 \text{ ng m}^{-3}$ with occasional episodic BC events. The highest 12-hr BC concentrations (up to 1020 ng m^{-3}) were measured from 1–6 May 2006, and were attributed to agricultural fires in Eastern Europe [Stohl *et al.*, 2007]. The annual average (median) BC concentration from Figure 1 for 1998 to 2007 is $39 (27) \text{ ng m}^{-3}$ and compares with lower model estimates of $\sim 21 \text{ ng m}^{-3}$ (only 2002 to 2003 available) using the TM5 chemistry transport model coupled to the M7 aerosol dynamical model (E. Vignati *et al.*, manuscript in preparation, 2008). A similar seasonal cycle has been observed at Alert and Barrow as reported by Sharma *et al.* [2006]. The annual BC average at Alert and Barrow of $\sim 25 \text{ ng m}^{-3}$ from mid-1998 to 2004 is lower than at Zeppelin, although a better comparison awaits a more fully overlapping data set at all three stations. Studies of trace selenium and vanadium aerosol concentrations, related to fossil fuel burning, also agree with the BC cycle [Maenhaut *et al.*, 1989]. The seasonal cycle in these and other atmospheric trace constituents is generally attributed to the seasonal position of the arctic polar front. During the winter, the front lies at about 50°N hence allowing long-range transport from major industrial regions in Europe, Russian and N. America into the Arctic [e.g., Treffeisen *et al.*, 2004]. Once in the region, air masses are likely to experience little scavenging in the stable, atmosphere of the northern hemisphere winter. The circulation pattern is different in summer, when the front lies further to the north (about 70°N) hence preventing polluted air masses from effectively reaching the Arctic [e.g., Polissar *et al.*, 2001]. In addition, frequent low-level stratus cloud-cover leads to drizzle, which in turn removes aerosols by wet scavenging.

[7] The 1998 to 2007 (AE-10 and AE-31 instruments), and the 2001 to 2007 (AE-31 only) data sets were examined for trends by de-seasonalising and simple linear regression analysis. Decreasing trends of -19.7 and -9.5 ng m^{-3} per decade were found, respectively. The latter result is more reliable as it corresponds to a continuous data set. However,

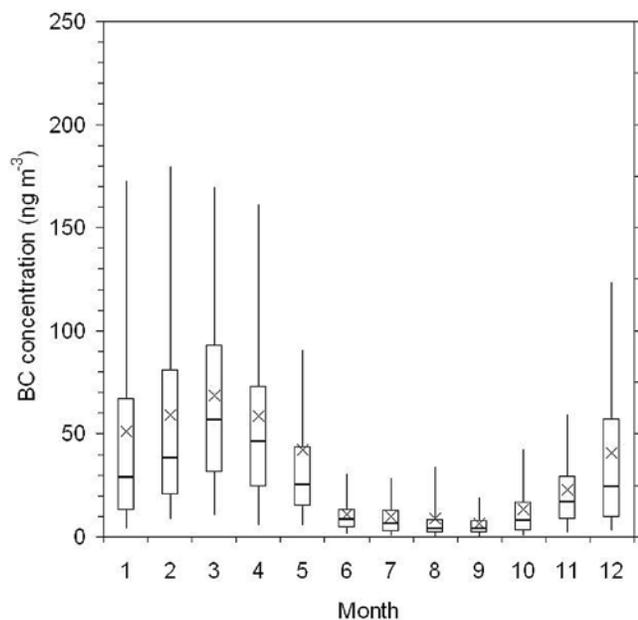


Figure 2. Box-and-whisker plot of monthly BC concentrations illustrating the mean (cross), and 5, 25, 50, 75, 95th percentiles for 1998 to 2007.

as the 2001–2007 trend is similar to the detection limit, it cannot be regarded as significant until a longer data set is available. However, a small increasing trend at both Alert and Barrow was observed from about 1999 to spring 2003 [Sharma *et al.*, 2006]. Prior to this period, large decreases in equivalent BC concentration by up to 54 and 27% were observed from 1989 to spring 1999 at Alert and Barrow, respectively. Sharma *et al.* [2006] considered several aspects to be responsible for the overall decline in EBC concentrations. The first concerned a reduction in BC fossil fuel emissions in the former USSR from 1989 to 1994. The second concerned changes in atmospheric circulation patterns, most notably the North Atlantic Oscillation (NAO). A positive (negative) NAO⁺ (NAO⁻) phase is defined when a larger (smaller) gradient in the standard sea level pressure between the Azores high and Iceland low occurs resulting in greater (less) zonal circulation across the North Atlantic and Europe. It has been observed (NOAA NAO data, <http://www.cpc.noaa.gov>) that during the 1980s to 2007 (covering BC data sets from this and other studies) NAO⁺ phases have occurred with a $\sim 74\%$ frequency as opposed to $\sim 26\%$ for NAO⁻ during winter and spring months (DJFM). Using a Lagrangian particle dispersion model, Eckhardt *et al.* [2003] demonstrated that air pollution transport to the Arctic was enhanced during NAO⁺ phases. Supporting evidence was found by Sharma *et al.* [2006] for BC at Alert and Barrow where NAO⁺ was 40% higher than NAO⁻ (1990 to 2002; DJFM). While DJFM values of 61 ($n = 6$) and 47 ng m⁻³ ($n = 1$) were observed at Zeppelin during NAO⁺ and NAO⁻ respectively, the influence of the NAO on the BC time-series is at present uncertain.

[8] Monthly BC concentrations are shown in Figure 2 as a box-and-whisker plot. Higher average than median values occur for all months, and a frequency distribution which is approximately log-normal was found (not shown). Maximum monthly averages range from ~ 68 ng m⁻³ in February

to April to a minimum of ~ 7 ng m⁻³ for June to September, representing a factor ~ 10 difference. Our observations are similar to those of Heintzenberg and Leck [1994] who reported EC ~ 11 ng m⁻³ in summer and 93 ng m⁻³ in winter from 1990 to 1992 at Zeppelin. This comparison of EC and BC concentrations is possible as our BC data set has been calibrated against an EC thermal method. Heintzenberg and Leck [1994] also measured at the Gruevbadet sea level site near Ny-Ålesund from 1979 to 1990. Lower values of 5 and 66 ng m⁻³ were found for summer/autumn and winter/spring, respectively. Whether this difference was due to changes in the long-term BC trend and/or measurement altitude is difficult to answer. However, it raises the question to what extent boundary layer (BL) measurements at sea level can be compared to those at Zeppelin which is considered to represent free tropospheric (FT) conditions. Sodar studies [Beine *et al.*, 2001] and web-camera observations of the BL cloud top (<http://www.nilu.no/niluweb/services/zeppelin/>) during late spring/summer suggest that Zeppelin is frequently representative of BL conditions. On the other hand, simultaneous measurements of the aerosol scattering coefficient (σ_{SP}) at Zeppelin and at sea level during spring/summer 2001 [Treffeisen *et al.*, 2004; Nyeki *et al.*, 2005a] indicated lower σ_{SP} values at Zeppelin by ~ 13 –66% suggesting that FT conditions were prevalent. Sea level measurements will be more affected by shallow ground inversions and katabatic winds from surrounding glacier sheets, especially during non-summer months. Mountain stations are often influenced by BL conditions, which may result in a diurnal variation in trace gas and aerosol parameters. Analysis of the entire 30-minute BC data set only gave a small diurnal variation during the summer months (JJA) of about $\pm 10\%$ about the average and median. However, these results are inconclusive as the variation is less than the ± 1 standard deviation range. Sharma *et al.* [2002] also found a small diurnal variation in summer but gave no further details. The above observations do not suggest that BC concentrations are higher at lower altitude, but highlights the fact that assessing whether Zeppelin is influenced by BL or FT conditions must also be based on aerosol source region and lifetime, meteorological conditions, solar irradiation etc. In conclusion, the body of evidence suggests that aerosol measurements at Zeppelin station may be considered to be generally representative of FT conditions.

[9] An interesting aspect in the annual cycle in Figure 2 is the period of low BC concentration lasting from summer to autumn. The onset of this period is more clearly seen in the 12-hr (not shown) instead of the monthly time-series. A steady decrease from March until the end of May occurs, after which a narrow BC concentration range ~ 0 –10 ng m⁻³ with an inter-quartile range ~ 10 ng m⁻³ is observed until the beginning of October. A similar seasonal cycle in BC concentration is also observed at Alert and Barrow [Sharma *et al.*, 2006]. A strong decrease in BC concentration occurs several weeks earlier at both sites in comparison to Zeppelin (Barrow, beginning of April, and Alert, mid-April) while BC remains low until October at all three sites. These observations are attributed to well-established atmospheric circulation patterns [Eneroth *et al.*, 2003; Sharma *et al.*, 2006].

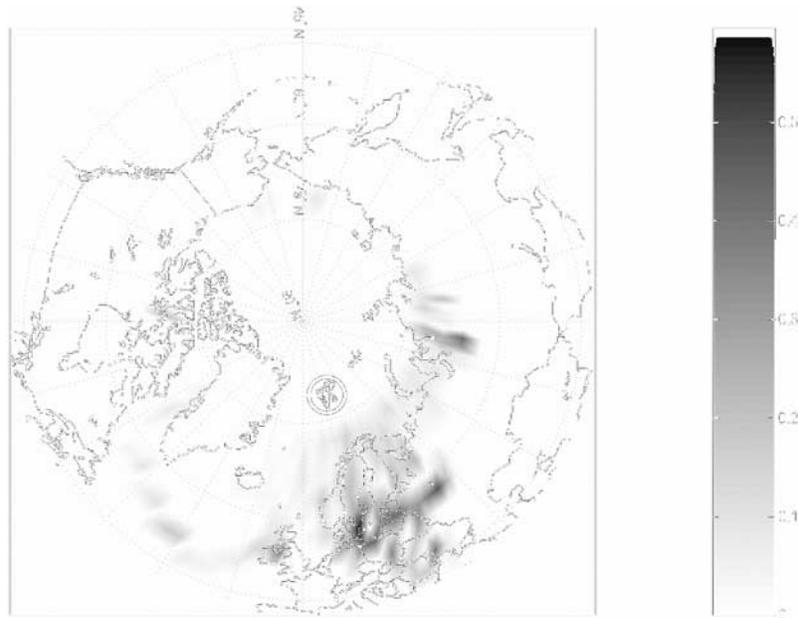


Figure 3. PSCF probability map of BC aerosol for the summer half-year (May–October) at Zeppelin station (black circle) for 1998–2007.

3.2. Determination of Aerosol BC Source Regions

[10] A trajectory analysis determined that air masses mainly originated from the Arctic ($>80^{\circ}\text{N}$) and Russian (30° – 130°E) sectors, and accounted for 40% and 31% of trajectories, respectively. This agrees well with the circulation patterns for Svalbard described in the previous section.

[11] In order to ascertain the source regions of BC at Zeppelin, the PSCF model was applied to back-trajectory data for the period of observation. Summer and winter PSCF maps appear in Figures 3 and 4, respectively. The summer map illustrates that Northern Europe is in general a

low PSCF area, although several regions of elevated PSCF appear in Scotland/Ireland, southern Sweden, and Russia (Kola Peninsula and Noril'sk). These regions are industrialized but elevated PSCF values are mainly due to a small number of BC episodes in the 1998–2007 database. This was confirmed by omitting episodes from a further PSCF analysis. In contrast, the winter map exhibits high PSCF regions in a broad swath covering the Kola Peninsula and Archangel'sk region to the Krasnoyarsk region in central Russia. A comparison of Figures 3 and 4 with BC PSCF maps for Alert [Polissar *et al.*, 2001] reveals that source

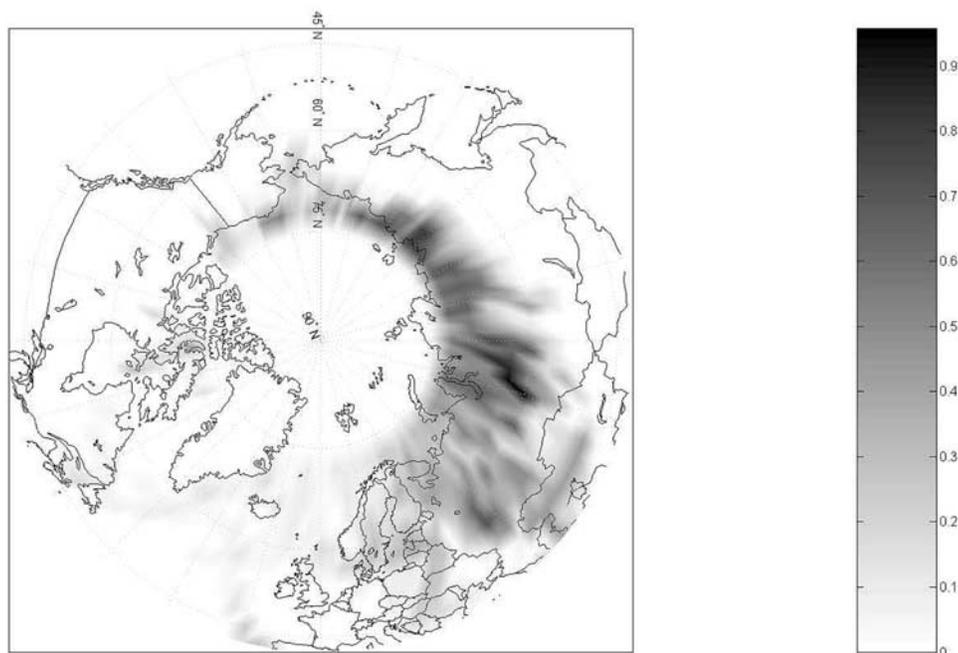


Figure 4. Same as Figure 3 except for winter half-year (November–April).

regions are broadly similar. However, subtle differences exist, for instance Figure 4 illustrates elevated PSCF in north-east Russia which is a tundra region with little heavy industrial or oil/gas production. Several other general aspects may also lead to differences, such as: 1) the different geographical locations of Alert and Zeppelin and hence regional circulation patterns, 2) the shorter length of the present BC data set, and 3) the use of 5-day rather than 10-day back-trajectories, which means that BC source regions from low latitudes are not captured in our analysis. An overview of the trajectory points allocated in the grid cells down to 42°N reveals that although there is adequate coverage of trajectory points down to 50°N over the western Atlantic and eastern Europe regions, other regions have sparser coverage. For instance, there are essentially no trajectory points further south than 66°N in the northern hemisphere sector from 90 to 270°E. It is therefore possible that potential source areas for the above-described regions indicate air masses high in BC concentration in Figure 4 which may in fact be transported from more southern latitudes (below 50°N) in East Asia, as postulated by *Stohl* [2006].

4. Conclusions

[12] Black carbon concentration, based on a thermo-optical calibration, for the 1998–2007 period at Zeppelin station, Svalbard, is reported. Annual average and median BC concentrations were 39 and 27 ng m⁻³, respectively, while monthly averages ranged from a maximum ~80 ng m⁻³ in February/March to a minimum 0–10 ng m⁻³ from June to September. PSCF trajectory analysis indicated that BC concentrations at Zeppelin were mainly influenced by source regions in northern and central Russia. Continuous data, available since 2001, indicated a decreasing trend in BC concentration (–9.5 ng m⁻³ per decade) up to mid-2007.

[13] **Acknowledgments.** The authors gratefully acknowledge NOAA ARL for use of HYSPLIT. Thanks are also extended to the staff at Norwegian Polar Institute, NILU, and PSI (AE-9 loan). Funding from the EU LSF programs is kindly acknowledged.

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