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[1] During the winter and spring the North American Arctic is impacted by anthropogenic black carbon (BC) in "Arctic Haze" pollution from sources mainly located in Europe and Russia. This black carbon, while suspended in the atmosphere and in surface snow, has a significant effect on radiative forcing of the Arctic atmosphere. Routine ground-level observations of aerosol black carbon by optical absorption have been made at a Canadian Arctic location, Alert (82.5°N, 62.5°W), Nunavut since 1989. A 3-year intensive study was conducted to compare BC obtained by the thermal analysis and optical absorption methods, so that the seasonal variations in the "operational" absorption cross sections of the aerosol could be determined. A time series analysis indicated that black carbon concentrations undergo a strong seasonal variation superimposed upon a long-term trend. The latter shows a decrease of about 55% in BC concentrations between 1989 and 2002 at Alert. Factors responsible for these trends such as changes in emissions and atmospheric transport support the hypothesis that BC emissions from the former USSR are mostly responsible for the observed decreasing trend. Transport from other sectors such as North America and Europe are not as prevalent at Alert. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 3319 Meteorology and Atmospheric Dynamics: General circulation; 4857 Oceanography: Biological and Chemical: Pollution; 4801 Oceanography: Biological and Chemical: Aerosols (0305); *KEYWORDS:* black carbon, emissions, Arctic

1. Introduction

[2] Black carbon (BC) is a substantial component of the carbonaceous aerosols which are produced as a result of incomplete combustion of fossil fuels and biomass burning worldwide. It is important in the atmosphere as it affects air chemistry processes, air quality, and climate. Submicrometer BC particles have a long atmospheric life and may be transported over long distances from source regions to the remote locations such as the higher latitudes. A combination of the long-range transport mechanism as governed by the atmospheric circulation patterns to the Arctic with lower precipitation events during the winter/spring time leads to higher levels of Arctic pollution, often referred to as "Arctic Haze" [*Barrie*, 1986, 1996; *Hansen et al.*, 1984]. Thus higher concentrations of particles are observed in the Arctic during this period including particulate sulfate and carbonaceous aerosols [*Sirois and Barrie*, 1999; *Polissar et al.*, 1999; *Quinn et al.*, 2002].

[3] Recent models [*Jacobson*, 2001] and observations [*Novakov et al.*, 2000; *Ramanathan et al.*, 2001] indicate that heating of the atmosphere by BC particles could be a major process in radiative forcing of the atmosphere. This effect is even more prominent when pollution is long range transported over locations with high surface albedo such as the Arctic [*Blanchet*, 1994].

[4] There have been several studies conducted previously in the Arctic. Most of these studies were episodic or short-term surface-based aerosols measurements, mainly conducted in the European Arctic [*Heintzenberg*, 1982; *Covert and Heintzenberg*, 1993; *Bigg and Leck*, 2001] except for the *Heintzenberg and Leck* [1994] study which reported measurements for 48 months. These short-term experiments have focused on the physical and chemical properties of arctic aerosols with influence from the Atlantic air masses as well as the anthropogenic continental air masses. Wintertime and summertime black carbon concentrations were as high as 330 ng m^{-3} and as low as 5 ng m^{-3}. Moreover, there has been some extensive work carried out at the site of Point Barrow (71.2°N, 156.3°W) in northern Alaska, focusing on the long-term trends in the aerosol measurements [*Polissar et al.*, 1998a, 1998b, 1999]. It also explored the possible sources impacting this site. Pollution at Point Barrow has been related to long-range transported anthropogenic sources, sea-salt aerosol (Pacific and the Arctic Ocean influence), and particles with relatively high black carbon content.

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Figure 1. Map showing the location of the sampling location. Alert is located at 82°27′N and 62°31′W on the edge of the Lincoln Sea at the northeastern tip of Ellesmere Island in the Canadian Arctic. This site is unique as potential source regions such as North America, the former USSR, and Europe influence it.

[5] Since May 1989, continuous ground-based, hourly measurements of particulate black carbon have been made at the Canadian Baseline laboratory at Alert (82.5°N, 62.5°W; see Figure 1), Nunavut. Seasonally adjusted site-specific attenuation for 3 years were applied to the 13 year BC time series to obtain the best BC concentrations. This site is centrally located in the Arctic and thus is influenced by northern hemispheric pollution as well as long-range transport from countries surrounding the Arctic. The main focus of this paper will be to investigate the possible trends in the 13 years of BC concentrations in relation to trends in BC emissions in the source regions. However, other influences such as atmospheric circulation, pathways or removal processes are not excluded and will be explored.

2. Methods

2.1. Aethalometer

[6] The aethalometer AE-6 (Magee Scientific [*Hansen et al.*, 1984]) measures the attenuation of light transmitted through particles that accumulate on a quartz fiber filter. A vacuum pump draws air through the instrument so that the particles continuously accumulate on the filter while being illuminated by visible light (incandescent light source). The effective operational wavelength of the aethalometer is 880 nm [*Bodhaine*, 1995; *Lavanchy et al.*, 1999]. The light transmitted is detected by two photo-diodes: one through the sample spot and the other through a blank (unsampled) portion of the filter called the reference spot. The change in attenuation is obtained as a function of

time and relates to the BC concentration in g m^{-3} as follows:

$$BC = \frac{-A \cdot 100 \cdot \ln\frac{I_2}{I_1}}{k \cdot Q \cdot (t_2 - t_1)}, \tag{1}$$

where I_1/I_2 is the ratio of the intensities of the sample beam to the reference beam at times t_1 and t_2 (in seconds); Q (m^3 s^{-1}) is the sample flow rate through the filter; A (m^2) is the area of the exposed spot on the filter; and k is the specific attenuation coefficient. A recommended value for k of 19 m^2 g^{-1}, which is based upon calibrations during instrument development and theoretical calculations, is typically used. This k accounts for absorption by BC and additional light attenuation assumed to be caused by the layering of particles or their close proximity on the filter, which induces multiple scattering as well as reflection effects. The detection limit of the aethalometer is dependent on the stability of the optics. For a 1-hour integration the noise in the light intensity corresponds to 2 ng m^{-3}. We have defined the detection limit as twice the standard deviation in the noise or 5 ng m^{-3}.

[7] The measurement of BC by this technique is controversial since this optical technique is based on light absorption of atmospheric particles sampled on filters and Mie theory to estimate BC mass concentrations in the atmosphere. However, it has been shown that k of atmospheric BC varies with composition of the aerosol, which varies in time and space [*Liousse et al.*, 1993; *Petzold and Niessner*, 1995; *Lavanchy et al.*, 1999; *Ruellan and Cachier*, 2000; *Sharma et al.*, 2002]. Thus a site-specific attenuation k(S.S.), as defined by *Sharma et al.* [2002], is required and can be obtained by knowing how much thermally derived elemental carbon (EC) is present in the samples. To address the ambiguity in the use of terms BC or EC in this paper, we will be using the term BC since this paper focuses on the light absorption measurements. Usually, the graphitic content of the sample as obtained from the thermal analysis is referred to as EC but here it will be called BC$_{therm}$. The BC concentrations determined from equation (1) were compared to the BC$_{therm}$ to derive seasonally adjusted k values, k(S.S.). This will be extensively used in the paper.

[8] A 3-year study was conducted at Alert to determine the relationship between light absorption measurements of an aethalometer and thermally derived black carbon concentrations (BC$_{therm}$). Seasonally adjusted site specific attenuation, k(S.S.), was then determined by using the ratios of BC by aethalometer and BC$_{therm}$ derived from the thermal method, as described in section 2.2. A subset of this data from the first year of the measurements has been investigated [*Sharma et al.*, 2002]. In this paper, we use all the data collected from November 1998 to March 2002 to determine any inter- and intra-annual variabilities in k(S.S.). These seasonally adjusted k(S.S.) are then used with the 13 year time series of aethalometer absorption measurements to obtain the best estimates of BC concentrations. Any long-term uncertainty in the k(S.S.) cannot be determined, and thus it is assumed that the seasonally

adjusted k(S.S.) are valid for 13 years of measurement corrections within some uncertainty.

2.2. Thermal Analysis: Two-Step Thermal Method

[9] The samples for the thermal analysis were collected on Pallflex quartz filters (47 mm, 2500QAT-UP filters from Gelman Sciences) at 30 slpm (standard liters per minute) for 2 weeks. This is the integrated sampling time required for the thermal analysis for the period 1998 to 2002. The details of the thermal technique can be found in *Cachier et al.* [1989]. Briefly, a decarbonation step in hydrochloric acid (HCl) fumes was applied to the sample filters prior to analysis. During the precombustion step, organic material is removed from the filter under a flow of pure oxygen at 340°C for a period of 2 hours. In step 2 the remaining carbon (black carbon) was determined by coulometeric titration (by using a Strohlein Coulomat 702C analyzer) of CO_2 evolved from the combustion of the sample at 1100°C. In order to achieve lower blank levels (5% of filter load), the filters were preheated at 700°C for black carbon and 400°C for the total carbon (TC). The organic carbon was determined by subtracting BC from TC. The precision and accuracy of this technique is better than ±10% and the limit of detection is 3 μg C.

[10] An absolute measurement technique for measuring BC_{therm} still remains illusive due to the complex characteristics of the aerosols. One reason for this is the lack of well defined universal temperature to differentiate between the elemental carbon and organic carbon analysis. The accuracy in BC_{therm} is also compromised due to other interferences such as presence of alkaline metals [*Novakov and Corrigan*, 1995; *Yu et al.*, 2002]. However, despite all these uncertainties, BC_{therm} is related mainly to graphitic carbon content and is presently the best available technique as its precision is excellent and trends deduced in this paper are valid. This technique has been compared to response of other laboratories in an intercomparison study [*Schmid et al.*, 2001; *Currie et al.*, 2002] and the response was in the "acceptable range" (not different from the mean response).

2.3. Aerosol Sulfate

[11] The detailed descriptions of the sample collection and chemical analysis have been given by *Sirois and Barrie* [1999]. Seven-day aerosol samples were collected on 20 × 25 cm Whatman 41 filters using a high-volume sampler. Once samples were back in the laboratory, they were cut into eight aliquots to be used for chemical analysis. Sulfate is analyzed by liquid ion chromatography (IC). The accuracy and precision of this technique is better than ±10%. The treatment of blanks and detection limit method are found in *Sirois and Barrie* [1999].

2.4. Back Trajectory Clusters

[12] The 10-day backward trajectories used in this analysis were provided by the Canadian Meteorological Centre model. A brief description of the model can be found at http://www.cmc.ec.gc.ca/cmc. A cluster analysis using a Hartigan's K mean algorithm was used to sort the trajectories into 6 groups that have similar pathways to Alert [*Hartigan*, 1975; *Dorling et al.*, 1992; *Sirois*, 1999]. In this technique, initial clusters are created and then trajectories are moved to the cluster with a mean backward trajectory closest to it. As the cluster means are a function of the trajectories that belong to the cluster, the process of allocation and mean calculation is an iterative one. The process is continued until no trajectories are moved from one cluster to another. In the present study, we used the mean Euclidean distance (after projection) between two trajectories as defined by *Hartigan* [1975] and *Dorling et al.* [1992]. This was done in the hope of being able to separate between back trajectories that remain mostly within the Arctic Circle and the ones coming from emission regions outside of it.

2.5. Time Series Analysis

[13] A geometric time variation model was used to describe the temporal variation of the daily concentrations [*Sirois*, 1997]. It included a long-term trend, a seasonal variation, and an auto-regressive component that were correlated in time. The parameters of the model [*Sirois*, 1997] were estimated by maximizing the log-likelihood function of the temporal model (see *Harvey and Phillips* [1979] and *Kohn and Ansley* [1985] for the definition of the likelihood function for a regression model with auto-regressive disturbances). The maximum-likelihood technique using the Kalman filter [*Kohn and Ansley*, 1986] was considered in this study [*Sirois*, 1997].

2.6. Trajectory Analysis of BC Measurements

[14] The potential source areas for the BC concentrations observed at the sites were investigated using the 10-day back trajectories and the observed BC concentrations. We used the technique developed by *Seibert et al.* [1994], in which one first associates each trajectory with an observed concentration and next superimposes a grid on the region of interest. Then one assigns to each grid cell the concentration of all the trajectories passing through the grid and then calculates the geometric mean of those values assigned.

2.7. Black Carbon (BC) Emissions Maps

[15] To analyze the BC concentration trend observed at Alert from 1989 to 2002, annual BC emissions by fossil fuel combustion were computed for the regions surrounding the Arctic. Previous studies indeed showed that former USSR, Europe and North America constitute the major sources impacting Alert during wintertime. Emission maps were calculated by applying the method developed by *Cooke et al.* [1999], for the submicron particles, to the United Nations energy data set. This database provides the consumption and transaction of 23 different fuel types (e.g., hard coal, lignite brown, diesel, etc.) for more than 200 countries worldwide until 1998. The 1990s' emission maps for Asia are more accurate than the 1980s' maps initially built by *Cooke et al.* [1999], since they now include the numerous countries (15) that were part of the former USSR (Ukraine, Belarus, Georgia, Armenia, Lithuania, Estonia, Latvia...). For every country a specific emission factor was applied to each fuel type consumed. Its value also takes into account if this is a developed, semideveloped, or developing country.

3. Results and Discussion

3.1. Quality Assurance of Black Carbon Measurements

[16] This section focuses on the quality control and assurance steps taken to compensate the BC concentration
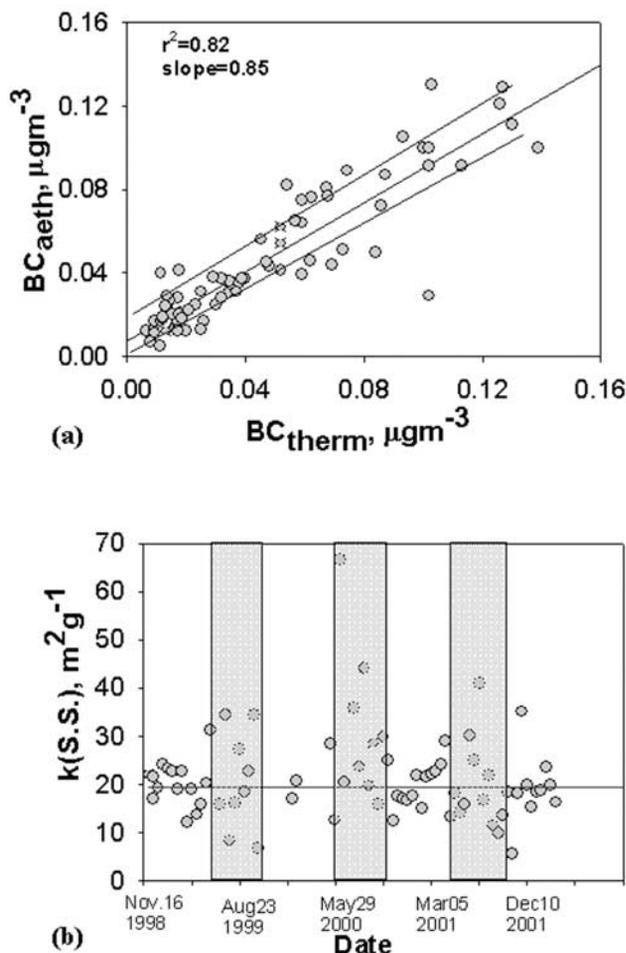
Figure 2. Relationship between (a) BC obtained from thermal versus optical technique showing a regression line and 1σ confidence limits and (b) the correction applied to the specific attenuation coefficient. The mean values for the site-specific attenuation were used to convert the light absorption measurements for the summer (highlighted) and winter.

data measured at Alert for 13 years. For the quality control measures the data were placed into the Research Data Management and Quality Control Systems or RDMQ (a data quality control package developed by Meteorological Service of Canada). Algorithms were developed to formulate the flagging process for the data validation. Data were invalidated from the data set whenever the instrument malfunctioned or local vehicle exhaust was present. Between 75% and 95%, data were recovered for the 13 years by using this quality control process.

[17] While improving the accuracy of the BC mass concentration estimates, the main focus was to take into account seasonal changes in the site specific attenuation coefficient, k(S.S.), which is used to convert optical absorption to black carbon mass. It has been shown previously that in Canada, k(S.S.) is variable from one location to another [*Sharma et al.*, 2002]. This is most likely due to the age, type, and composition of the aerosols. A subset of the data (9 months of measurements) taken at Alert was used in a previous paper to illustrate this point. In this paper, results

from November 1998 to April 2002 are presented to include any interannual variability that might exist in the data set.

[18] Figure 2a displays the relationship between BC obtained by aethalometer, BC_{aeth}, (using manufacturer recommended specific attenuation coefficient of 19 m^2 g^{-1}) and BC_{therm} obtained by the thermal technique, at Alert. On average, both techniques agree well (as shown in Figure 2a) with a correlation coefficient of $r^2 = 0.82$, a slope of 0.85 ± 0.10 (N = 85) and an intercept of 0.09 ± 0.02 suggesting that 19 m^2 g^{-1} might be applicable to the aethalometer data at Alert. However, a regression such as this is heavily influenced by the few points at the upper end of the distribution.

[19] Figure 2b shows a time series of k(S.S.), derived from aethalometer absorption, BC_{aeth} and BC_{therm} [*Sharma et al.*, 2002]. During the polluted Arctic haze period from November to April, the median value of k(S.S.) was 19.8 ± 0.9 m^2 g^{-1} but during the non-Arctic haze period from May to October it was significantly higher and much more variable (28.8 ± 2.5 m^2 g^{-1}). During the winter and spring, aerosols are aged, internally mixed, and anthropogenic, while during the summer the area experiences a minimum of man-made sources. The aerosols influenced by regional sources in that area consist of biogenic sulfur derived from the DMS-rich waters along the fringes of the pack ice area, primary sea salt, and organic particles from wind-induced bubble bursting aerosols are more influenced by regionally produced aerosols of marine biogenic sulfur derived from dimethyl sulfide, marine sea salt, organic particles from wind-induced bubble bursting, and dust from land areas with minimum anthropogenic origin [*Maenhaut et al.*, 1996; *Sirois and Barrie*, 1999; *Norman et al.*, 1999; *Leck and Bigg*, 1999; *Leck et al.*, 2002]. During the summer, wet removal is greater, and long-range transport is much weaker [*Iversen*, 1996] leading to very low concentrations. The large scatter during summer may also be due to greater analytical uncertainty. There is no trend found in the 3 years of k(S.S.) values and unless there are drastic changes in BC sources and other aerosol sources, there is no reason for drastic changes in k(S.S.) Thus, on this assumption, these values were applied to absorption measurements of the aethalometer to correct for the effects of real aerosols.

[20] Figure 3a shows the time series of 6-hour average BC concentrations from 1989 to 2002 at Alert. There is a marked monotonic decrease over this 13-year period; 60% for all data, 56% for the winter/spring (October–May), and 54% for the summer (June–September). However, the BC concentrations at the end of the period seem to be stabilizing. In support of the BC impact on the climate, Figure 3b shows a time series of sulfate, another important component of the aerosols in the Arctic. The trend analysis of sulfate has previously been published for the data between 1980 and 1995 [*Sirois and Barrie*, 1999]. Here a similar time period as for BC concentrations has been chosen. Since BC and sulfate are mostly from the same combustion sources during the winter and spring, there is a similar decrease in sulfate of 56% during the winter, 29% for all data, and 9% for the summer over this 13-year period. There is a large difference in the wintertime and summertime trends. This difference could be attributed to a reduction in the anthropogenic and biogenic emission sources, but a more definite answer still requires further modeling investigation. The
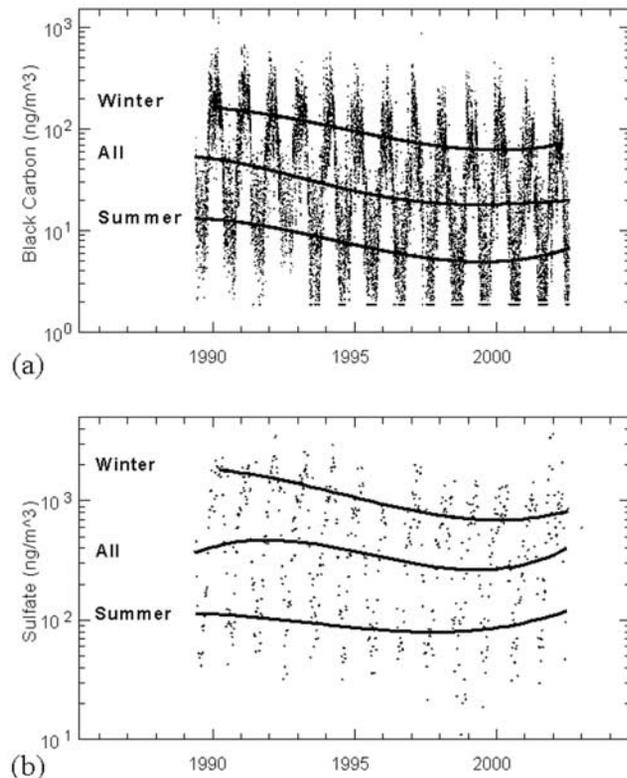
Figure 3. Times series of (a) corrected black carbon concentrations (on a logarithmic scale) and (b) sulfate concentrations from 1989 to 2002 at Alert showing a downward trend in the measurements.

decrease in sulfate could also be related to the decrease of sulfur in the fuel content in Europe during the 1990s [*Boucher and Pham*, 2002].

[21] There is a distinct seasonal variation in BC (Figure 4) with highest concentrations from January to April (about 96 ng m^{-3}) and lowest from June to September (about 8 ng m^{-3}). On average, concentrations during the summer were 12 times lower than those in winter. In comparison, the wintertime and summertime BC averages of 80 and 8 ng m^{-3}, respectively, were measured both at Spitzbergen [*Heintzenberg and Leck*, 1994] and Barrow [*Bodhaine*, 1995]. As mentioned above, the seasonal variation can be affected by seasonal differences in transport and removal. In addition, BC emissions in the source regions can change. Thus the relative contribution of transport and variation in emissions to the BC trends are explored in the following sections.

### 3.2. Atmospheric Transport and Cluster Analysis

[22] Alert is an unique location as this site is impacted by BC emissions from various sources in the lower latitudes as governed by atmospheric transport (Figure 1). In order to identify different source regions of emissions which are likely to impact Alert, the cluster analysis techniques described earlier were applied to the full data set. These sectors were generated on the basis of the most represented trajectories coming from that sector excluding the overlaps between the two clusters. The back trajectories were available every 6 hours, and 6-hour averages of BC concentrations were used corresponding to the time of trajectories.

[23] Figure 5 shows the probability density function of the 10-day back trajectories arriving at Alert from the potential source regions as shown by the 6 different clusters. Here the clusters are represented by contours. Specific labels were assigned to these clusters: AL for Alaska, C-R as central Russia, EU as Europe, ARC as Arctic, E-R as eastern Russia, and N-A as North America. Forty-one percent of the 10-day back trajectories were in the Arctic sector, while the frequency of transport from the Russian, European, and North American sectors was in the range 8 to 27%. The rest was from the Alaskan sector. There were some intra-annual differences found in the transport for all the sectors. For the wintertime and summertime (the latter shown in brackets), 23% (13%) of the transport was from the former USSR (Union of Soviet Socialist Republics) sector, 5% (10%) from AL, 10% (6%) from EU, 30% (17%) from N-A, and 32% (55%) from the ARC sectors, respectively. Transport decreased by about 40% from winter to summer for all sectors except for the Arctic and Alaska.

[24] Figure 6 shows the distribution of hourly mean BC mass concentrations for winter (Figure 6a) and summer (Figure 6b) at Alert for each trajectory sector. The BC concentrations were higher during the winter than the summer with the most striking difference found for the former USSR sector. The highest BC concentrations were associated with transport from the sectors E-R and C-R during the wintertime. The highest geometric mean was 134 ng m^{-3}. In comparison, the summertime BC concentrations of 7 ng m^{-3} are similar in all sectors. This is also supported by the concentration gradient map showing the potential source receptor relationship in Figure 7. Each concentration grid represents the mean BC mass concentration observed at Alert that is associated with trajectories passing over that grid. During the winter the BC concentration is highest for trajectories from the former USSR where the major industries (point sources such as Norilsk in Northern Siberia and in Kola Peninsula in European Russia), and numerous population are located, thereby bringing in higher levels of BC to the Arctic. The boundary layer during the winter is very stable and stratified due to low temperatures and no sunlight. Also, the cloud formation and frequency are minimal thereby minimizing the scavenging of particles. Thus the pollutants stay in the boundary layer
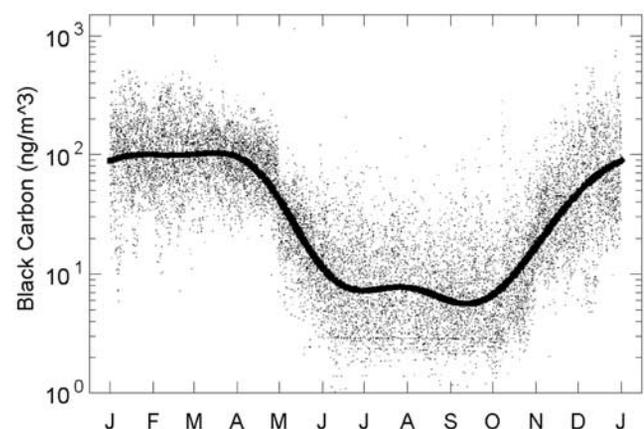
Figure 4. Seasonality in the corrected black carbon measurements for the data from 1989 to 2002.
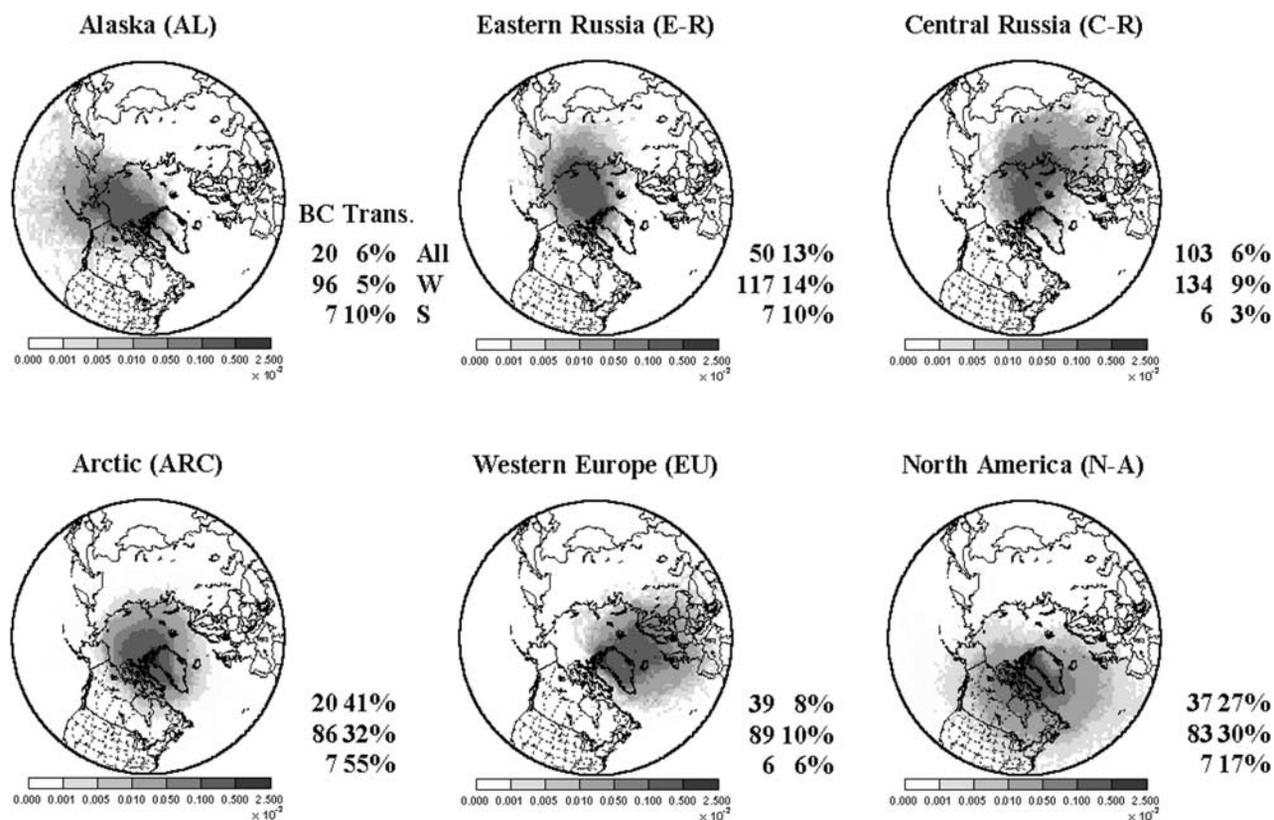
Figure 5. Sector analysis showing the probability density for 10-day back trajectories between 1989 and 2001 at Alert. Also given are the median BC concentrations (ng m^{-3}) for all year, winter, and summer for the sectors with the frequency of the transport from each sector.

for longer periods of time. However, trajectories during the summer were localized to the Arctic representing more homogeneous air masses among all sectors. Scavenging of aerosols by precipitation is also higher during the summer and could also contribute to lower BC concentrations. The 95% Multiple Kruskal-Wallis test (Figure 6) confirms that in summer, all sectors were not significantly different but that in winter, E-R and C-R sectors were significantly higher than all of the other sectors.

[25] It should be noted that there is an uncertainty error associated with the calculations of the back trajectories especially due to the sparseness of data in the remote regions such as the Arctic. This error usually increases as one get further back in time. However, this analysis still has some strength in identifying the source regions which when combined with the emissions inventory data can yield reliable indication of origin of BC.

3.3. Black Carbon Emissions

[26] So far, the observations at Alert are related to the transport of the emissions from different source regions to this receptor site. Is the declining trend in BC also due to changing emissions in the region of origin? To explore this, averaged annual BC emissions by fossil fuel combustion were computed for 1989–1998 by using the methodology of *Cooke et al.* [1999] for the regions surrounding the Arctic. The regions used in the calculations include former USSR, Europe, and North America since the magnitude of transport to Alert is the most frequent from these sectors.

Figure 8 shows the mean annual emissions of BC in Europe, North America, and the former USSR from 1989 to 1998. The greatest change in the BC emissions is in the former USSR sector as compared to the other two. A significant decline in the BC emissions occurred for the former USSR sector. This is most likely due to economic slowdown as a result of the collapse of the former USSR in 1991. Impact of similar economic slowdown has also been observed in the emissions of other gases such as sulfur dioxide from the smelters in the former USSR over the same period of time [*Idimechev et al.*, 1995; *Poznyakov*, 1993]. There is a great uncertainty in the BC emissions for both 1991 and 1992 for the former USSR as not all the data might have been reported for these two years. Therefore these should be used with great caution. Similar lack of data has also been noticed for sulfur dioxide for 1991 and 1992 [*Alexeyev*, 1996]. In contrast to the BC emissions in the former USSR, those in the North American and European sectors increased slightly.

[27] Figure 9 compares for each year from 1990 to 1998, the transport weighted BC emissions, the BC concentrations corresponding to the sectors as indicated by the back trajectories, and also the transport frequency to Alert from North America, Europe, and former USSR sectors. All values have been normalized to 1990 numbers. The results in all three sectors show that BC mass concentrations have decreased by as much as 60% from 1990 to 1998. There is a greater decrease in the BC concentrations in the North American and European sectors by as much as 10% than
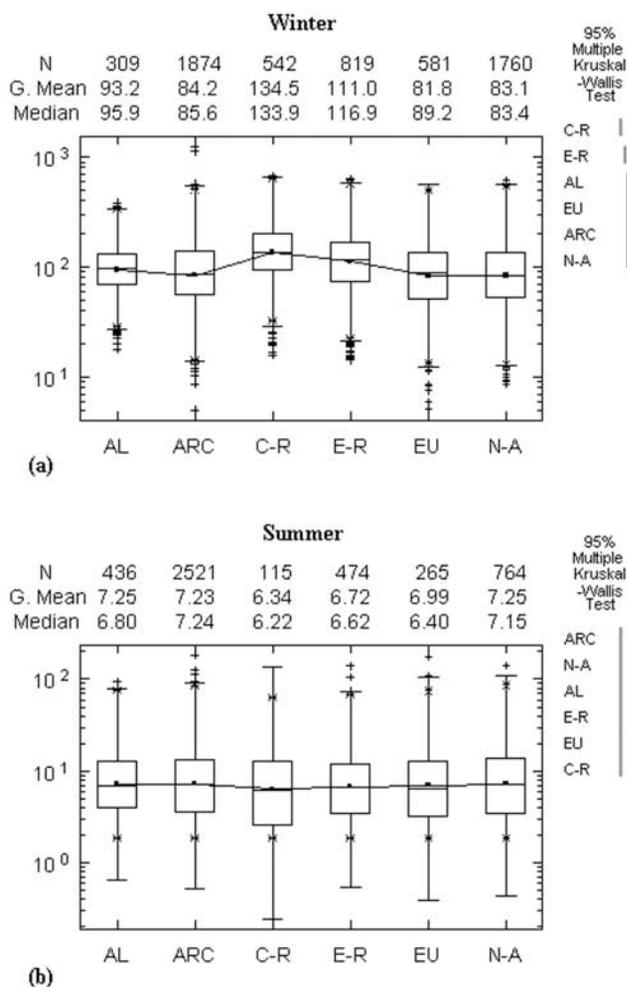
Figure 6. Distribution of 6-hourly mean BC concentrations associated with the back trajectories in the 6 clusters for the (a) winter and (b) summer showing the median, 25th percentile, and 75th percentile.

in the former USSR sector. Figure 9a shows that for the former USSR BC concentrations decreased by 55% from 1990 to 1998, corresponding to an 80% decrease in the transport weighted BC emissions. Over the same period of time, the transport of BC was variable with peaks (+20%) in 1993 and 1994 and the lowest in 1996 (−35%). Both BC concentrations and transport weighted BC emissions looked quite similar in 1996 and 1997. The patterns for the North American and European sectors are very different from the Russian sector as shown in Figures 9b and 9c. The trends in the BC concentrations have decreased over the same time period, but the trends in transport weighted emissions from these regions and the frequency of transport from these sectors to Alert do not correspond to decrease in the BC concentrations. Thus change in the transport weighted BC emissions alone cannot explain the trend in the observed BC concentrations from North American and European sectors. However, the processes occurring in the air masses and the atmospheric circulation en route to Alert might explain some differences. Transport along marine pathways might have contributed to greater removal by clouds and precipitation. Thus pollutants from the North American

sector are largely removed before it reaches the Arctic. This is mainly due to "strong lifting" of the air masses which could take the pollutants to higher altitudes limiting the impact on the Arctic boundary layer. For instance, Figure 10 shows the average height above the ground as a function of time back for each mean trajectory for the winter and summer for the 6 clusters, as discussed in the previous section and shown in Figure 5. There is a striking difference in the height of the trajectories in the three sectors, Alaska, Europe, and eastern Russia during the winter and summer. In winter from these three sectors, the point of origin 10 days
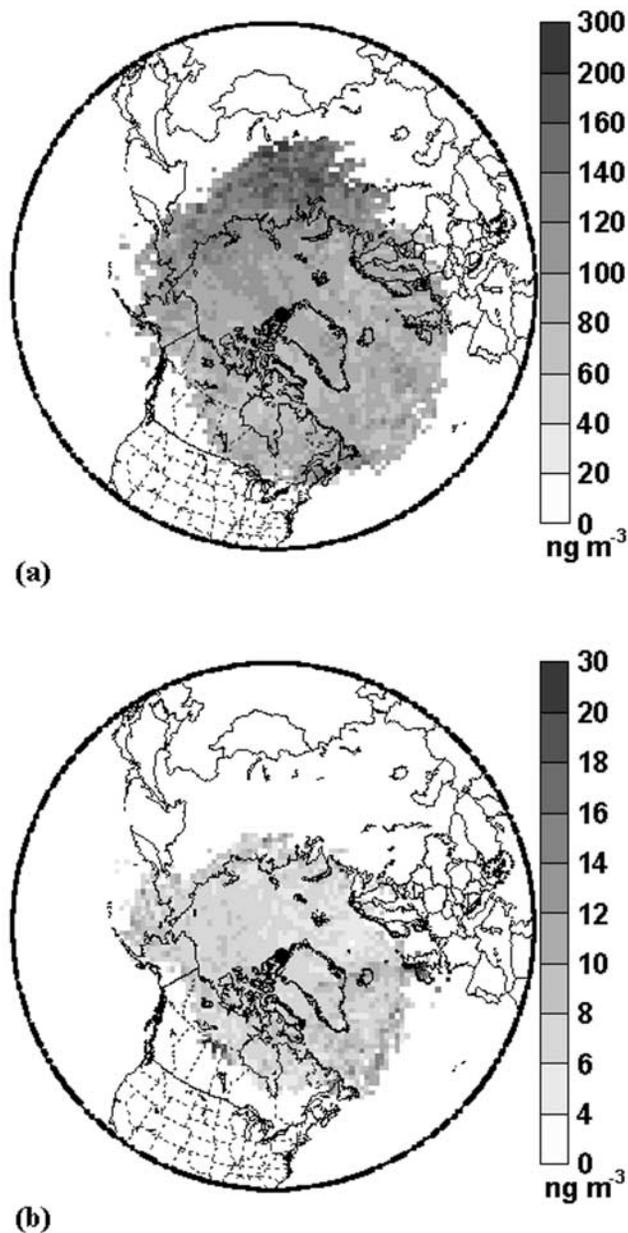
Figure 7. Upwind areas identified by trajectories for which BC measurements at Alert are high and low from 1989 to 2001 as generated by the *Seibert et al.* [1994] analysis for (a) winter (January to April) and (b) summer (June to September). For details, see text. Note the difference in scales between the two plots.
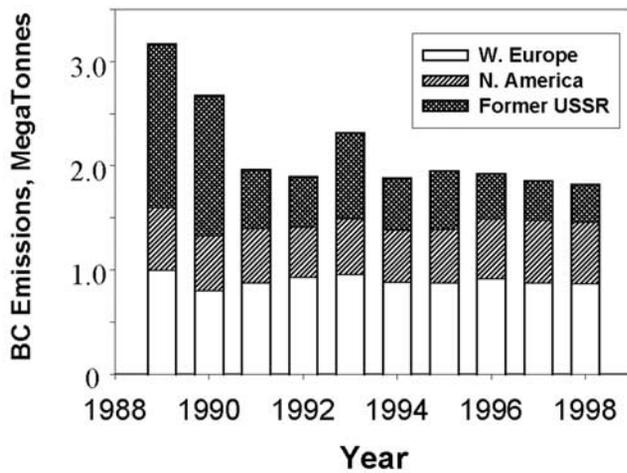
Figure 8. Average annual BC emissions for 1989–1998 for western Europe, North America, and the former USSR as computed by *Cooke et al.*'s [1999] method.

back in time was located at a height of 1.5–2.5 km perhaps due to influence of the convection over the North Atlantic and North Pacific Oceans. Other sectors had trajectories traveling at lower heights. Thus the differences in the parcel heights might be responsible for the transport of the emissions as it has a bearing on how ground-level pollutant release affects the air mass. On the other hand, *Eckhardt et al.* [2003] has shown that due to the enhanced Icelandic flow the North American emissions do not get transported directly to the Arctic but are transported more efficiently to Europe where they combine with European emissions and are eventually transported to the Arctic [*Bottenheim et al.*, 2003].

[28] Hence a source of black carbon has a potential for influencing the Arctic that depends on transport frequency, transport pathway, removal, and emissions. Northern hemispheric transport modeling applied to the Arctic has shown [e.g., *Barrie et al.*, 1989; *Iversen*, 1996; *Christensen*, 1997] that on a per unit of pollution basis the emissions regions of the former USSR, located at a high latitude not far from Alert, have relatively more effect on lower tropospheric air pollution in the high Arctic than emissions of North America and Asia. European emissions have an Arctic polluting potential that falls in between these two extremes. More recent modeling efforts have confirmed this [*Klonecki et al.*, 2003].

4. Conclusions

[29] A 13-year data set of aerosol optical absorption measurements at Alert in the Canadian high Arctic has been quality assured and converted to a time series of black carbon aerosol mass concentration using results of a 3-year intensive study of the specific absorption coefficient of the Aethalometer instrument used. The instrument specific absorption coefficient was determined to be $19.8 \pm 0.9 \ m^2 \ g^{-1}$ but during the non-Arctic haze period from May to October it was significantly higher and much more variable $28.8 \pm 2.5 \ m^2 \ g^{-1}$. The analysis of BC mass concentrations from 1989 to 2002 shows a monotonic decrease regardless of time of year totaling 55%. The average concentration in the Arctic haze season, November

to May, is 12 times higher than in the remaining period, June to October. The highest BC mass concentrations during the Arctic haze time were associated with the 10-days back trajectories from the former USSR sector as climatologically persistent Siberian high-pressure system drives the air northward into the Arctic. Analysis of trajectory pathways and emission trends indicates that the decrease in BC concentrations in air from the former USSR is mainly due to variations in the emissions and the circulation patterns. However, the decreasing BC concentrations in the North
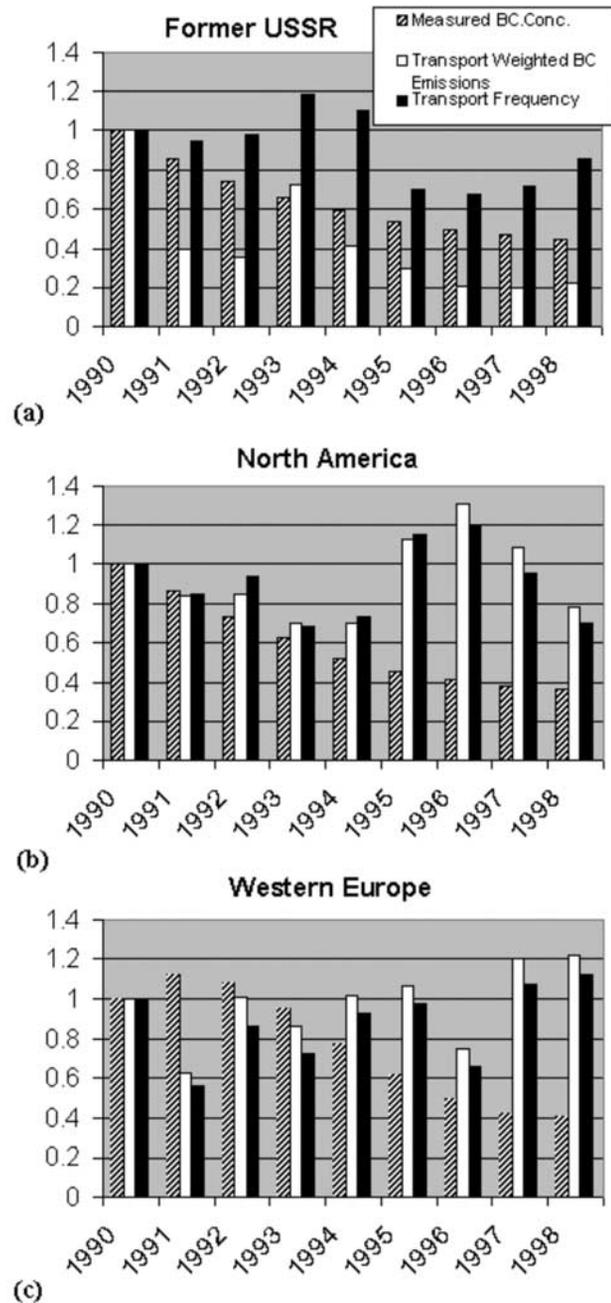
Figure 9. Changes relative to 1990 of BC air concentration geometric means, BC emissions, and transport frequency to Alert from (a) the former USSR, (b) North American, and (c) western European sectors.
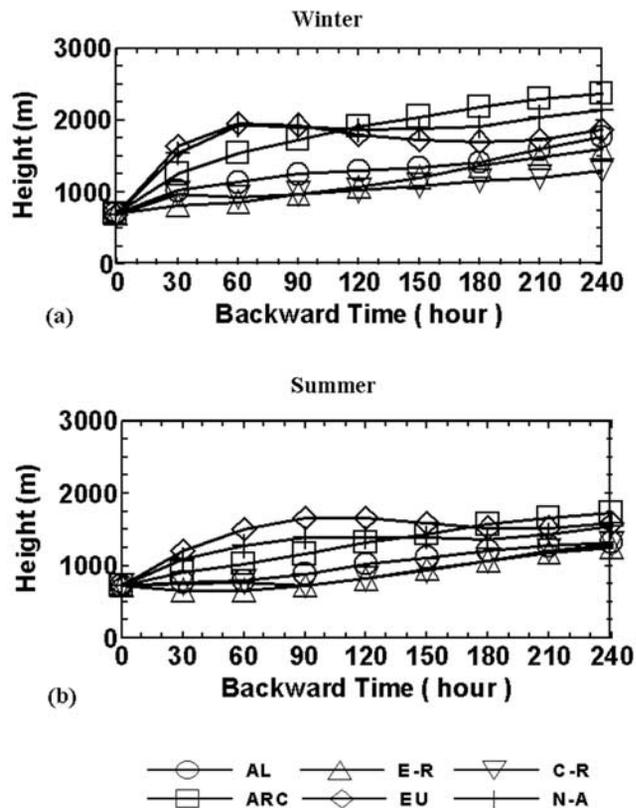
Figure 10. Average height above the ground as a function of time back for the 6 clusters for the 10 days back trajectories for (a) winter and (b) summer.

American and European sectors are not associated with increasing BC emission trends in those source regions. Thus transport and removal variations play a major role in these sectors, and the changes in prevailing sectors might also be related to the air parcel heights. These results suggest that since there is a decrease of 55% in the BC concentrations over the last decade, the warming of the lower troposphere by BC at polar sunrise has decreased monotonically by a total of 55% at Alert between 1989 and 2002. However, by assuming the absorbing material to be internally mixed with hydrophilic reflecting material should further reduce this effect. The next step in this analysis will be to use a climate model with natural and anthropogenic aerosols as active constituents in it to assess the climate effects of anthropogenic aerosols in the Arctic.

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