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Locations and preferred pathways of possible sources of Arctic aerosol

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Abstract

The chemical composition of particles collected at Alert, Northwest Territories, Canada, show strong, persistent seasonal variations. In a previous study, a 2-way/3-way mixed factor model was performed on the weekly average concentrations of 24 aerosol components measured over the period from 1980 to 1991. The Multilinear Engine (ME), a new mathematical technique, was used to obtain the solution. The two modes of the 2-way model consist of the source composition profiles and mass contributions over the 11 yr, while for the three modes of the 3-way model, source profiles, mass contributions variations over the weeks within a year, and the year-to-year variation over the 11 yr within the measurement period. Five 2-way and two 3-way factors were found to provide a good fit to the data and were easily interpreted. In this investigation, potential source contribution function (PSCF) analysis was applied to the source contributions derived from the ME analysis by incorporating meteorological information in the form of 5-d air parcel back trajectories. The potential locations and/or the preferred pathways of these possible sources were then determined by the PSCF analysis. © 1999 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Studies of Arctic air pollution have been of increasing interest for the past 3 decades. Particulate matter from mid-latitudinal industrial sources has been observed in the Arctic for many years. The existence of anthropogenic particles and their influence on visibility has been called Arctic haze. The pollutant concentrations in the Arctic have a strong seasonal variation characterized by a summer minimum and a winter maximum. Furthermore, during the winter, much of the particulate matter is mostly of man-made origin, while in the summer, the lower concentrations have substantial contributions from natural sources (Barrie, 1986; Barrie and Hoff, 1985). It has been demonstrated that Arctic haze consists of various components that may represent aerosol carried by different air masses or from different sources (Heidam, 1981). Therefore, a relatively simply physical structure in the Arctic aerosol might be assumed (Barrie and Hoff, 1985; Barrie and Barrie, 1990). Arctic haze is important since it may result in changes in the solar radiation balance as well as visibility changes in this region. Thus, the investigation of the seasonal variations and long-term trends in these aerosol parameters as well

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as the identification of possible sources and transport pathways of aerosol is important. The results may aid in understanding the effects of Arctic air pollution on the polar ecosystem and on global climate, and developing an appropriate strategy for reducing the extent of Arctic air pollution.

Receptor modeling apportions the measured aerosol concentrations to their sources (Hopke, 1991) based on measurements made of various chemical species and/or physical properties for a series of samples over time. Multivariate methods can be applied to problems where there is no *a priori* knowledge of the sources and their chemical properties. A variety of such methods have been applied to airborne particle composition data (Henry, 1991). Very recently, a mathematical tool called the Multilinear Engine (ME) was invented by Paatero (1998) for solving the general multlinear problem. In ME, any multilinear modeling problem can be represented by a set of equations, each of which approximates one data value by a sum of products of unknowns (factor elements). These unknowns in the prescribed model structure are then estimated for the data values.

ME is very flexible and provides a general framework for different multilinear models including *pure* bilinear and trilinear models as well as mixed 2-way/3-way models. ME permits weights for individual data value based on measurement uncertainties, treatment of uncertain data like missing values and values below detection limit and non-negativity constraints for the factors. ME was successfully used to solve a mixed 2-way/3-way model describing particle concentration data collected at Alert, Northwest Territories, Canada (Xie et al., 1998). Five 2-way and two 3-way factors were found to provide a good fit and could be readily interpreted as different possible particle sources. These probable sources have identifiable compositional profiles and distinctive seasonal variations.

Although the receptor modeling can identify the possible sources in terms of their chemical nature, seasonal variation and estimate the importance or contribution of these sources, they are unable to determine the locations of sources and the pathways by which the aerosol materials are transported to the receptor site. To overcome this shortcoming, meteorological information needs to be incorporated into the receptor model together with the chemical compositional information. Meteorological information can be used in receptor modeling in a variety of ways (Hopke, 1997). One such method is the potential source contribution function (PSCF) (Ashbaugh et al., 1985; Malm et al., 1986). PSCF and its modification have been used for determining possible source origins and preferred pathways of several individual chemical species of Arctic aerosol (Cheng et al., 1993; Hopke et al., 1995).

In this investigation, the factors (possible sources) obtained by the ME analyses (Xie et al., 1998) were regarded as components of aerosol constituents which have common emission origins or transportation pathways to the Arctic, and PSCF analysis was applied to the calculated factor scores. The locations or preferred pathways of the possible sources of Arctic aerosol can then be obtained. The chemical measurements and back trajectory data are described first. Then the results from ME are briefly reviewed. Following the description of PSCF analysis for the factors from ME, the results of PSCF are provided and interpreted.

2. Sampling and chemical composition determination

2.1. Chemical data

Airborne particulate samples were collected at Alert, Northwest Territories, Canada (latitude 82.3 N, longitude 62.5 W) by the Atmospheric Environment Services (AES) of Canada. Samples have been taken on a weekly basis since July 1980. The samples were analyzed for major ions by ion chromatography (IC) and for trace elements by instrumental neutron activation analysis (INAA) and inductively coupled plasma emission spectroscopy (ICP). Details of sampling and chemical analysis were presented by Barrie and Hoff (1985) and Barrie et al. (1989a). These data are summarized by Xie et al. (1998). Data for 24 chemical species in up to 532 weekly samples obtained between September 1980 to August 1991 were used in the analysis.

2.2. Trajectory data

The three-dimensional AES trajectory model (Olson et al., 1978) was used to construct the air parcel movement in the horizontal as well as vertical directions. The movement was described by segment end points of coordinates in terms of latitude, longitude, and height of each point. Five-day backward trajectories were calculated every 6 h four times a day at 00, 06, 12 and 18 UTC for 925 and 850 hPa levels. The model used three-dimensional objectively analyzed wind fields at four pressure levels: 1000, 850, 700, and 500 hPa from Canadian Meteorological Center. Cubic spline interpolation was used to obtain winds at intermediate levels in the vertical direction and between horizontal grid points.

3. Data analysis

3.1. The multilinear engine and mixed 2-way/3-way modeling

The system of equations of a mixed 2-way/3-way model used in the previous ME analysis can be expressed as following:

$$X = TG + ABC + E \tag{1a}$$

or

$$X_{ijk} = \sum_{q=1}^{Q} T_{ikq}G_{jq} + \sum_{p=1}^{P} A_{ip}B_{jp}C_{kp} + E_{ijk}$$

$$\left(\begin{array}{l} i = 1,, I \\ j = 1, \dots, J \\ k = 1, \dots, K \\ p = 1, \dots, P4 \\ q = 1, \dots, Q \end{array}\right). \qquad (1b)$$

In the above equations, the three-way data array **X** is approximated by the sum of a Q factor 2-way PCA model with score and loading factors **T** and **G** and a P factor 3-way trilinear factor model with **A**, **B**, and **C** factors in three modes. For Alert data, matrix **T** contains time variations (source contributions) over the entire 11 yr, and **G** provides the corresponding concentration profiles of the sources derived from 2-way modeling. **A** and **C** are factors corresponding to time variation during a year and over the 11 y, respectively, and the outer product of **A** and **C** is equivalent to **T** in the two-way results. **B** is the concentration profiles of the sources appeared in the 3-way model, i.e. the 3-way counterpart of **G**. **E** denotes the un-modeled part of **X**. Since there is a 3-way part in the model, the data itself needs to be presented in the form of a 3-way data array as well and 3 indices are needed in Eq. (1b). For the same reason as in the presentation of data array **X**, the logically 2-way matrix **T** is represented with 3 indices as well. The ME algorithm and the details of the ME analysis of Alert data have been previously described (Paatero, 1998; Xie et al., 1998).

3.2. Factors from ME analysis

The ME analysis found five 2-way and two 3-way factors fit and could be interpreted for the Alert data (Xie et al., 1998). Fig. 1 present the five 2-way factors of the mixed model. Figure 1 shows the **G** factors normalized to have concentration units, which provide the quantitative information of chemical composition of the sources, i.e. source profiles. Fig. 2 shows the source contribution factors (**T** factors).

Fig. 3 presents the 3-way factors of the mixed model. The upper part shows the source contributions which are the outer product of the two modes of time variation factors, **A** and **C**, in the 3-way model. In the lower part of the figure, the composition factors **B** are shown, which are the sources profiles of the sources.

The nature of the seven sources obtained by the mixed model are summarized in Table 1. The first 2-way factor is associated with anthropogenic activities and the second one is attributed to wind blown soil dust. Constituents from sea salt dominate the third 2-way factor. The fourth 2-way factor contains mainly SO_4^{2-} and the majority of H^+. The fifth 2-way factor contains almost all of iodine, while the first 3-way factor has the majority of Br^-. A common feature of a maximum appearing around March and April just after polar sunrise in these acidic, iodine and bromine factors suggested the involvement of photochemistry. The second 3-way factor denotes a biogenic component of the Arctic aerosol which is characterized by the presence of methanesulfonate (MSA), a tracer for non-sea salt SO_4^{2-} of natural marine origin.

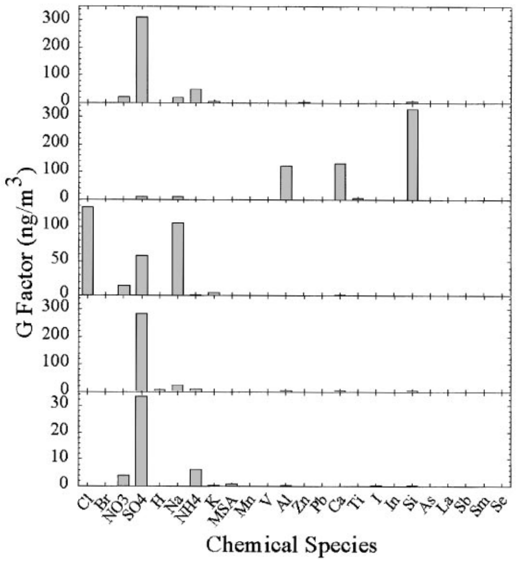

Fig. 1. **G** factor (compositional profile) of 2-way modeling.

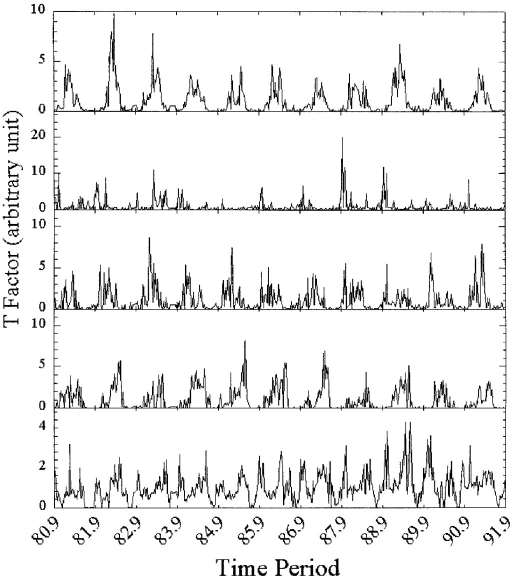

Fig. 2. **T** factors (time variations) of 2-way modeling.

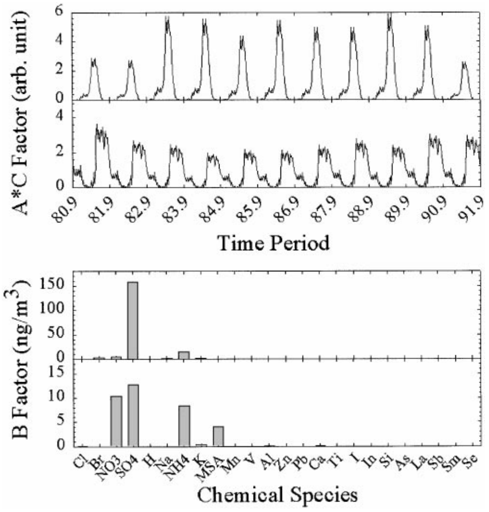

Fig. 3. *Upper*: Outer product of **A** and **C** factors (time variations) of 3-way modeling. *Lower*: **B** factor (compositional profile) of 3-way modeling.

The factors are described in greater detail by Xie et al. (1998).

3.3. PSCF analysis

As mentioned before, although the receptor modeling techniques may identify the possible sources in terms of their chemical composition and seasonal variation, they are unable to determine the specific location of sources and their preferred pathways. These tasks will be performed by a PSCF analysis.

The concept of PSCF is to calculate a conditional probability function describing the spatial distribution of probable geographical source locations by using trajectories. The possible source region is subdivided into a gridded i by j array and the total number of trajectory segment endpoints over the whole study period T is N. If n_{ij} endpoints fall into the ijth cell, the probability of an event, A_{ij} can be given by

$$P[A_{ij}] = n_{ij}/N \tag{2}$$

The probability, $P[A_{ij}]$, represents the relative residence time of randomly selected air parcel on the ijth grid which denotes the potential transport of material to the receptor site. If there are m_{ij} endpoints among n_{ij} corresponding to the trajectories that arrived at the receptor site with pollutant concentration exceeding a pre-specified value, then the probability of this event, B_{ij}, is given by

$$P[B_{ij}] = m_{ij}/N \tag{3}$$

where $P[B_{ij}]$ reflects the relative residence time in a grid cell for these contaminated air parcels. In this study the median concentration unless otherwise stated is selected as the criterion value. The potential source contribution function (PSCF) is then defined as a conditional probability:

$$\mathrm{PSCF} = P[B_{ij} | A_{ij}] = \frac{P[B_{ij}]}{P[A_{ij}]} = \frac{m_{ij}}{n_{ij}} \tag{4}$$

Grid cells with high PSCF values are then interpreted as potential source areas. It should be emphasized that the PSCF result does not yield the emission rate for a pollutant but rather the preferred source region or transport pathways to the site.

The weekly ground concentrations may result from material transport from different heights. Therefore, a total probability of material transfer from multiple vertical heights to the samples must be calculated from the conditional probabilities computed for each pressure level (Cheng et al., 1993). The heights used in this study were at the 925 and 850 hPa levels. If the layers are assumed to contribute material equally to the ground level samples, then the integrated probability of an elevated concentration over multiple pressure layers is given by

$$P[B_{ij}] = \sum_k P[B^k_{ij} | A^k_{ij}] P[A^k_{ij}] \tag{5}$$

Table 1
The composition of each of the factors identified in the mixed 2-way/3-way ME analysis

Factor	Constituents	Possible source origin
1	SO_4^{2-}, NO_3^-, NH_4^+, K^+, Mn(?), V, Zn, Pb, In, As, Sb, Se	Anthropogenic
2	Mn, V(?), Al, Ca, Ti, Si, La, Sm	Soil
3	Cl^-, Na^+, SO_4^{2-}, K^+	Sea salt
4	H^+, SO_4^{2-}	Acidic photochemical
5	I^-, SO_4^{2-}, NO_3^-	Iodine (acidic photochemical)
6	Br^-, SO_4^{2-}, NO_3^-, NH_4^+,	Bromine (acidic photochemical)
7	MSA, SO_4^{2-}, NO_3^-, NH_4^+	Biogenic sulfur

where the index k denotes different pressure level and $k = 1$ and 2, and $P[B_{ij}]$ is the total probability associated with the polluted air masses arriving at these multiple height levels. The total potential source contribution function (TPSCF) can be normalized to give

$$\mathrm{TPSCF}_{ij} = \frac{P[B_{ij}]}{\sum_k P[A_{ij}^k]} = \frac{\sum_k P[B_{ij}^k | A_{ij}^k] P[A_{ij}^k]}{\sum_k P[A_{ij}^k]}. \tag{6}$$

The numeric range of the TPSCF is between 0 and 1. A grid cell having a TPSCF value of 0 is unlikely to be a source region, while a value of 1 is likely to indicate a source region. Since PSCF or TPSCF is computed as a ratio of the counts of selected events (m_{ij}) to the counts of all events (n_{ij}), it is likely that a small but non-zero m_{ij} may result in a high PSCF or TPSCF value in case the corresponding n_{ij} is also small. However, it is obvious that the larger the value of n_{ij}, the higher the probability that material emitted from the grid affects the samples at the receptor site. To minimize the influence of the grid cells with small n_{ij}, an arbitrary weight function was created to reduce the PSCF values for such cells (Cheng et al., 1993; Hopke et al., 1995). The weight function was defined as follows:

$$\begin{aligned} W(n_{ij}) &= 1.00 \quad n_{ij} \geqslant 4 \\ W(n_{ij}) &= 0.85 \quad n_{ij} \geqslant 3 \\ W(n_{ij}) &= 0.65 \quad n_{ij} \geqslant 2 \\ W(n_{ij}) &= 0.50 \quad n_{ij} \geqslant 1. \end{aligned} \tag{7}$$

All of the maps shown in the next section are those of weighed TPSCF results.

The grid size chosen for the TPSCF computation should be sufficiently large to assimilate the uncertainty of a trajectory end point. Kahl et al. (1989) noted the displacement from the median value of a 5-d back trajectory is of the order of 1000 km. Thus for this TPSCF analysis, as the previous studies (Cheng et al., 1993; Hopke et al., 1995), a 5° × 5° grid size corresponding to a distance of about 500 km in the latitudinal direction and about 48–355 km in the longitudinal direction was chosen.

4. Results

Figs. 4–10 show the TPSCF results of all seven 2-way and 3-way factors from ME analysis obtained by using the median values over all 11 yr.

4.1. Anthropogenic sources

As shown in Fig. 1 and Table 1, the first 2-way factor contains more than 30% SO_4^{2-}, about 40% NO_3^-, high percentage of NH_4^+, K^+ and the majority of V, Zn, Pb, In, As, Sb and Se which are considered to be of anthropogenic origin. It can be seen from Fig. 2 that this anthropogenic factor has a concentration maximum starting from December and extending to February, i.e. Arctic winter. This factor represents the main contributor to the

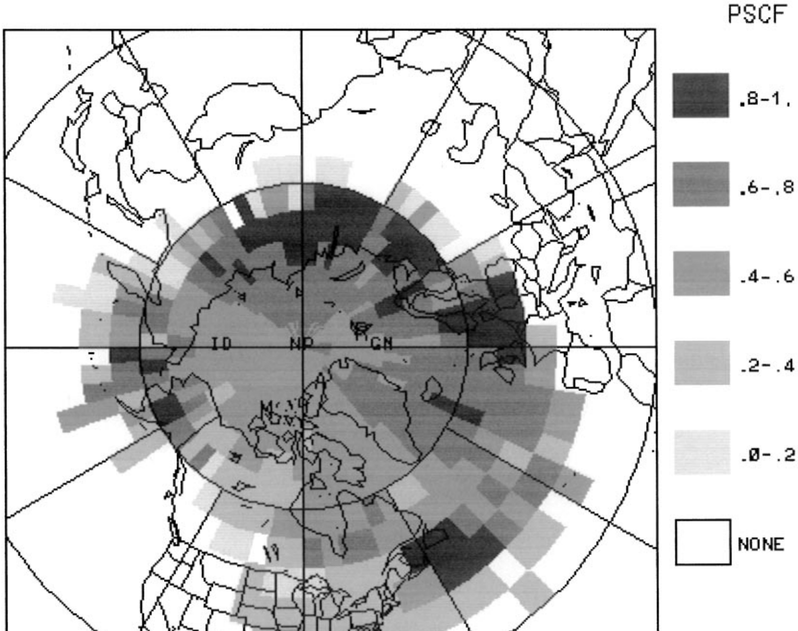

Fig. 4. TPSCF plot for the anthropogenic factor.

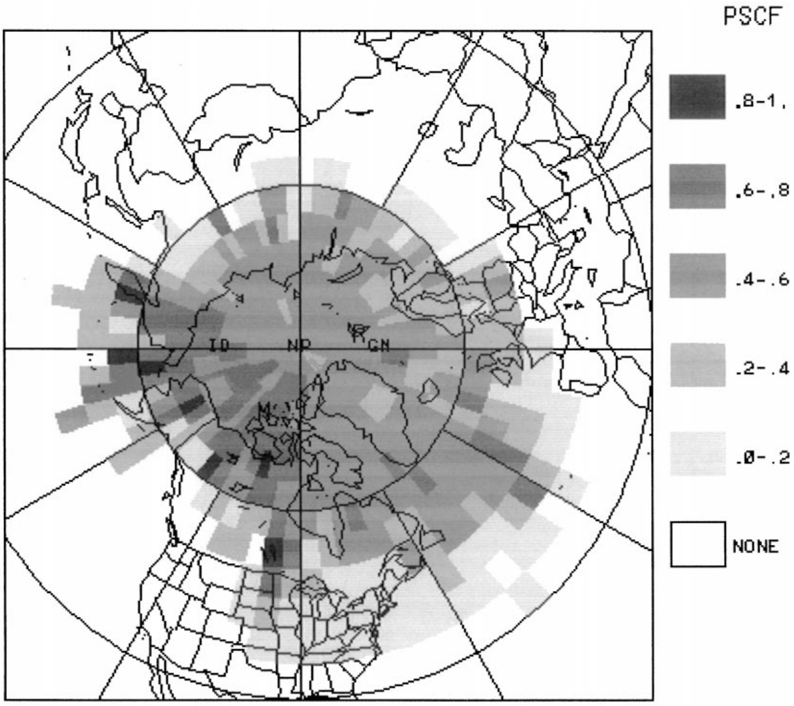

Fig. 5. TPSCF plot for the soil factor.

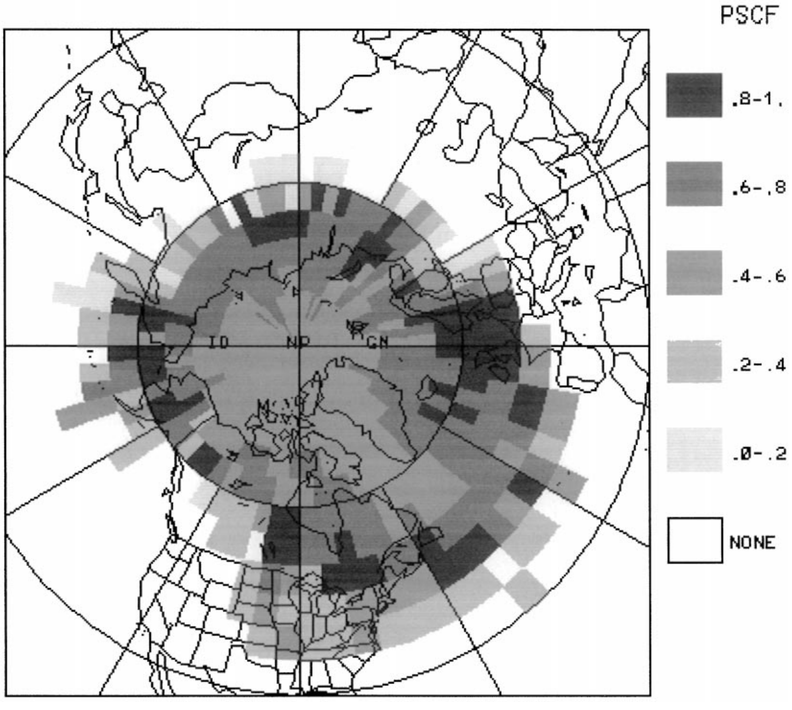

Fig. 6. TPSCF plot for the sea salt factor.

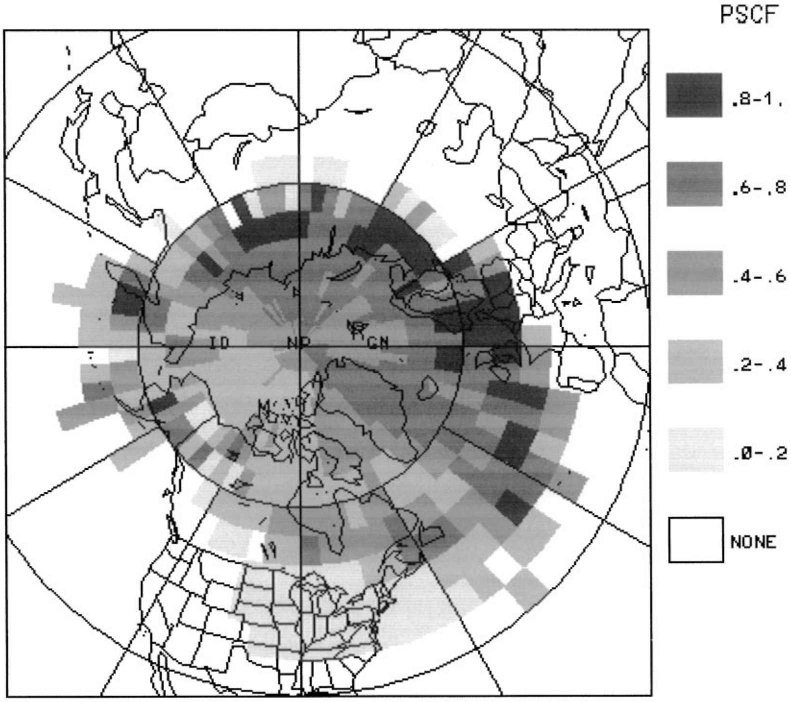

Fig. 7. TPSCF plot for the photochemical factor.

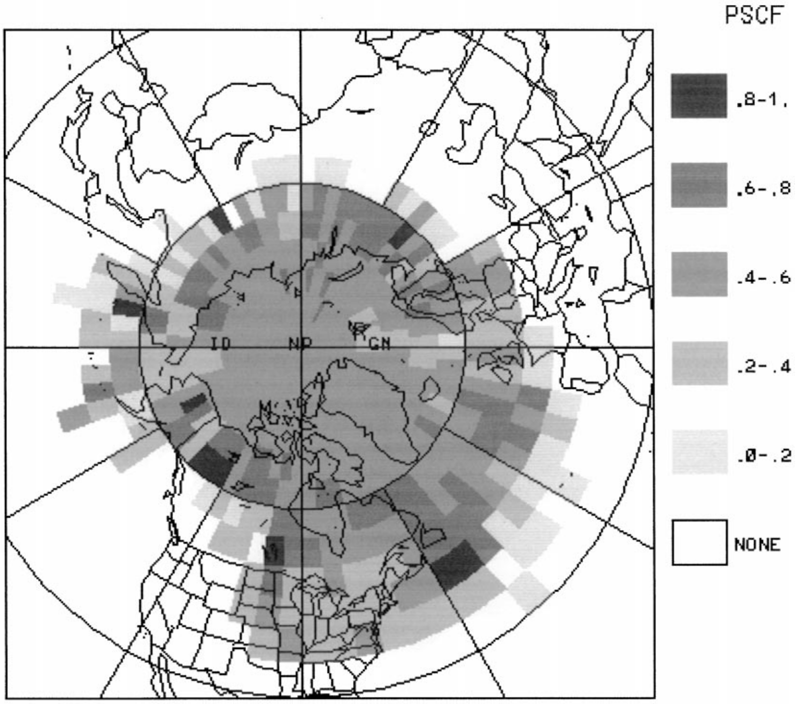

Fig. 8. TPSCF plot for the iodine factor.

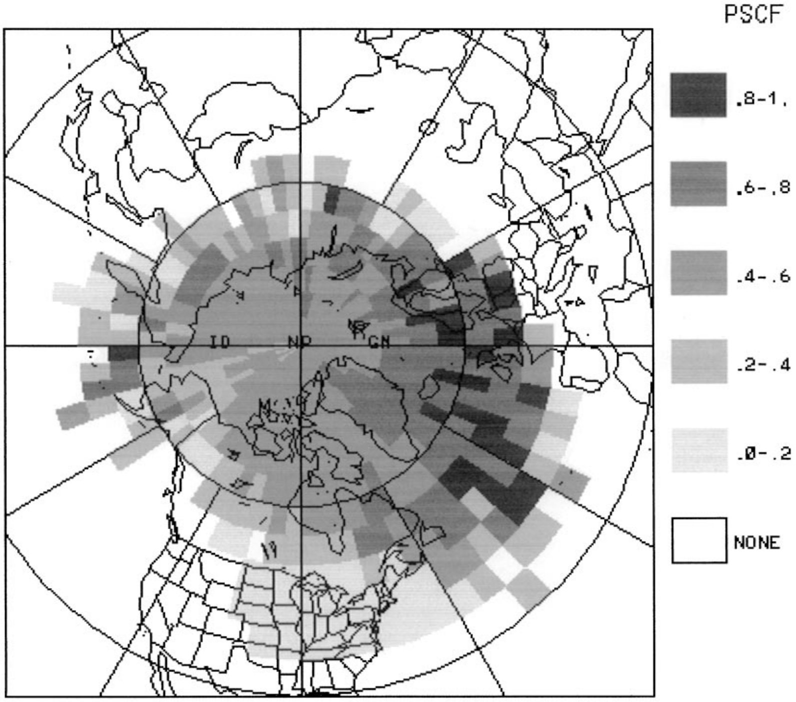

Fig. 9. TPSCF plot for the bromine factor.

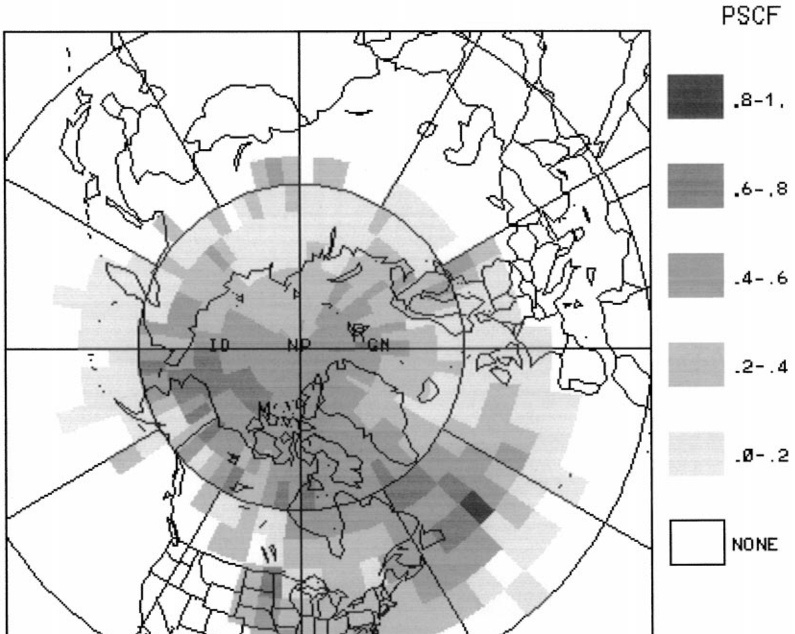

Fig. 10. TPSCF plot for the biogenic factor.

winter Arctic haze. The TPSCF result for this anthropogenic factor is shown in Fig. 4. Grid cells with TPSCF values larger than 0.8 are found mainly in industrialized areas of Europe and Russia as well as several high potential spots along the eastern coast of Canada. In previous studies, Cheng et al. (1993) and Hopke et al. (1995) have investigated the source origin and preferred pathways for non-sea salt sulfur and several metallic species like Pb, V, In, As, Zn, etc. Source areas and transport of individual species were identified. The anthropogenic factor represents the transport of a variety of industrial emissions to Alert when during the winter, Arctic high pressure systems penetrate into the populated mid-latitudes. The TPSCF plot in Fig. 4 was found to integrate the main features of the TPSCF patterns that were found for the individual anthropogenic species (Cheng et al., 1993). The TPSCF plot presented here appears to have fewer spurious areas of high potential that could not be interpreted in the earlier analysis. In general the areas identified as high potential for the winter anthropogenic factor appear to correspond well with the 1980 global anthropogenic SO_2 emission distribution (Dignon, 1992).

4.2. Soil

The second 2-way factor contains most of Mn, Al, Ca, Ti, Si, La and Sm and this factor is dominated by Al, Ca and Si (Fig. 2). All these elements are known to be the major constituents of soil, which suggests there is a wind blown soil dust for the Arctic aerosol. As shown in Fig. 2, there are two maxima in the seasonal variation of soil factor, one in spring and another in the late summer and early fall period. Previous studies suggested that soil dust in the Arctic may come from distance remote areas like Asian desert sources (Rahn et al., 1977; Pacyna and Ottar, 1989). The peak in April/May may be associated with longer-range transported Asian desert dust raised by storms occurring particularly in April and May, while the peak in September/October is probably caused by the onset of transport further south reaching areas of suspendable soil. The source areas in Asia are beyond the range of 5 d trajectories, so they cannot be located in the TSPCF analysis. However, the high potential spots in Fig. 5 suggest that the tundra areas of Alaska and Canada are possible soil sources. Local area can also be an origin of Arctic soil dust during blizzard conditions (Barrie et al., 1989a). It was found that there is a correspondence between blizzard conditions in February 1983 and the sharp peak that appeared then (Xie et al., 1998).

4.3. Sea salt

The third 2-way factor contains most of the Cl^-, a major portion of Na^+ together with part of the NO_3^-, SO_4^{2-} and K^+, which suggests a marine origin. Sea salt has a broad contribution over the period from October to April. Barrie and Barrie (1990) attributed this seasonal feature to a probable combination of longer aerosol residence times and a stronger sea spray source in the winter than in the summer. Fig. 6 shows the TPSCF results for the sea salt factor. Major source areas were found in the Atlantic Ocean and the Bering Sea, which are consistent with the distribution of sea-salt aerosol concentration estimated for the boreal winter (December–January–February) (Kahl et al., 1989). Although sea water is the major contributor of this aerosol component, the Arctic Ocean was not found to be a source area in Fig. 6 presumably because much of it is ice covered during the winter. There were also high TPSCF areas in northeastern and central Canada, in Alaska and in Russia, which may be particles transported from the North Pacific Ocean.

4.4. Photochemical sulfate, and particulate bromine and iodine

As shown in Figs. 1 and 3, the fourth 2-way factor contains most of the H^+ and part of the SO_4^{2-}. The fifth 2-way factor accounts for the majority of iodine and the first 3-way factor provides the source of most of the observed bromine. The seasonal variations of these factors have a common feature, i.e. a maximum in March and April just after polar sunrise (Xie et al., 1998). As the sunlight moves northward in the spring, more photochemical reactions can occur, including photochemical conversion of SO_2 to acidic sulfate, and production of particulate bromine and iodine. Shown in Figs. 7–9 are the TSPCF plots of the acidic iodine and bromine factors. Although these factors are characterized by H^+, I^- and Br, SO_4^{2-} is still the major species in these source compositions. In contrast to the SO_4^{2-} in the anthropogenic factor characterized by a winter maximum, sulfate in these factors peaks in the spring because much more of the SO_2 is oxidized to SO_4^{2-}. Thus SO_4^{2-} in these factors represents SO_x and the high potential areas can be compared with high SO_2 emission regions. Shown in Figs. 7–9, the sources and origins downwind of Europe and Russia are similar in location as the high TPSCF areas seen in Fig. 4.

The production of Br^- from its precursors usually is accompanied by troposphere O_3 depletion and H_2SO_4 transformation from SO_2 (Barrie et al., 1988, 1989b). A weak but significant correlation of daily I and Br concentrations observed in the spring of 1988 (Bottenheim et al., 1990) suggests the possibility of common origins of iodine and bromine. The chemical mechanism involving the production of Br^- and the depletion of ozone in the lower Arctic troposphere at polar sunrise has been investigated (Barrie et al., 1988; Finlayson-Pitts et al., 1990; Bras and Platt, 1995; Vogt et al., 1996; Sander et al., 1997). From Figs. 8 and 9, it seems that the Atlantic

Ocean is the main region of the production of particulate bromine and iodine from the photolysis of marine bromine and iodine compounds.

There is a secondary peak in the iodine factor around September and October which may be caused by photolysis of biogenic iodine emission to the atmosphere during the secondary phytoplankton blooms in northern oceans in late summer (Barrie and Barrie, 1990).

4.5. Biogenic sulfate

As can be seen from Fig. 3, the second 3-way factor is characterized by the presence of methanesulfonate (MSA). MSA is the product of the oxidation of sulfur-containing compounds like dimethylsulfide (DMS) or dimethyldisulfide (DMDS) emitted by the biogenic activity in the surface layer of the ocean. Even though MSA contributes only insignificantly to the mass of Arctic aerosols, it is useful as a tracer for non-sea salt SO_4^{2-} of natural marine origin. Therefore, this factor represents a biogenic component of the Arctic aerosol.

There are two peaks in the seasonal variation of this factor, one around April/May and the other about July/August (Xie et al., 1997b). Li et al. (1993) postulated that the spring peak is caused by transport from oceanic sources in the North Atlantic and North Pacific and possibly by accelerated DMS oxidation in the northern atmosphere as spring solar irradiation increases in intensity. Lavasseur et al. (1994) suggest that DMS released by ice algae during ice breakup may also contribute to this peak. According to Li et al. (1993) the summer peak is due to DMS from more local/regional Arctic sources and possibly from higher-latitude northern oceans, like the Atlantic Ocean off the coast of Norway and the northwestern North Pacific Ocean. From Fig. 10, within the five days trajectories domain there are several high potential grids found in the Atlantic Ocean and in the Northern Arctic Ocean. The high TPSCF grids in Atlantic Ocean probably reflects sources in the low latitudinal Atlantic Ocean in the spring time, while the relative high potential areas in Northern Arctic Ocean revealed the local origins in the summer. The role of more locally emitted DMS in the production of atmospheric aerosols in the Arctic in summer was suggested by Ferek et al. (1995). Their results showed that the Arctic Ocean is potentially a source of DMS, which probably becomes more important as the sea ice melts in the early summer.

5. Discussion and conclusions

In this study, meteorological information in the form of backward air parcel trajectories was used to locate the source origins and preferred transport pathways of Arctic aerosols measured at Alert. The probable aerosol sources used in this study were identified in a previous study by using a multivariate receptor modeling technique, i.e. the ME analysis. The temporal variation information of seven sources obtained by a mixed 2-way/3-way ME modeling together with the back trajectories calculated for the measurement time period were applied in the TPSCF analysis. The resulting TPSCF patterns provided information on the likely source locations or preferred aerosol transport pathways to the high Arctic. There are several constraints with this kind of analysis. First, the sampling was weekly so that the trajectories coming from a number of possible directions could arrive in a single sampling period. Only 5-day back trajectories were calculated because of concern that the cumulative errors in the parcel location beyond 5 d become very large. These restrictions limit the spatial extent of our analysis. The industrialized regions of the Northern Hemisphere can often be greater than 5 d. Also, there is no basis for a differential weighting of the various pressure levels in the analysis, but clearly different height trajectories could have quite different effects on the ground-level concentrations depending on exactly what vertical mixing condition exists. The TPSCF results identify grid cells corresponding to potential source regions and/or preferred pathways. The results are consistent with current knowledge of emissions, transport patterns to the Arctic, and transformation and removal mechanisms.

In summary, anthropogenic aerosol represents by SO_4^{2-} and a number of metallic elements has a TPSCF pattern with high values in the high emission regions of Eurasia. However, there are also high values in the central Atlantic Ocean which may be a pathway for pollutants transported from regions out of the domain of the 5 d trajectories. The tundra areas in Alaska and NWT of Canada are the sources of soil dust observed at Alert. The TPSCF pattern of sea salt suggests that Atlantic Ocean is the main contributor. As for particulate SO_4^{2-} transformed from SO_x, the TPSCF pattern suggests the sources and origins in Europe, and the Asian part of Russia. For particulate Br, I, the Atlantic Ocean seems to be the main source region. The high potential areas in the TPSCF plot of biogenic aerosol source suggest that both low latitudinal Atlantic Ocean and local Arctic Ocean are the contributors of biogenic aerosol.

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