Source Sector and Region Based Analysis of Black Carbon Deposition in the Arctic

1. Introduction

Black Carbon (BC) is a carbonaceous particulate matter produced during incomplete combustion of biomass materials (wood, dung and crop residue burning), and fossil fuels (coal, diesel fuel and natural gas).\(^1\) BC is one of the main absorber of solar radiation in the atmosphere. Furthermore, BC can transport over large ranges and accumulate on mountain tops and the Arctic.\(^1,2\) BC deposition on snow and ice could increase the solar radiation absorption of snow and ice; hence, increase the temperature of snow and accelerate the snow melting process.\(^1,3\) BC has been identified as the second largest contributor to global warming after carbon dioxide (CO\(_2\)). Studies suggest that BC caused 25% of 20\(^{th}\) century warming over the Arctic.\(^1,4\)

Arctic temperature has elevated approximately twice the global average temperature increase rate over the last decade. The temperature elevated up to 4\(^{\circ}\)C during the winter months from 1954 to 2003. Also the Arctic sea ice shrank to the lowest satellite records in September 2007.\(^3,4\) The Arctic glaciers melting will affect the sea level rise, ocean salinity, and extinction of some animal species.\(^4\)

The atmospheric lifetime of BC is estimated to be between 4 to 12 days.\(^1\) BC lifetime is considered very short in comparison with the Greenhouse Gases (GHGs) such as CO\(_2\) with approximate atmospheric lifetime of 100 years, N\(_2\)O with atmospheric lifetime of 114 years, and CFC-115 with atmospheric lifetime of 1700 years.\(^1,5\) Therefore, by starting a policy for reducing
or mitigating the emission of BC, rapid changes in the atmospheric concentration and the associated environmental and health impacts of BC will occur in short terms.¹

In this study the effects of BC emissions from different source sectors (e.g. transportation, power, industry, residential and biomass burning) and source regions (e.g. Europe, North America, China, Russia, Central Asia, South Asia and Middle East) to the Arctic BC concentration are investigated. For this purpose we used a chemical transport model and validated the data using surface observation over North America and aircraft observations over the Arctic during spring and summer 2008.

2. Data and Methods

2.1. Modeling System

2.1.1. Chemical Transport Model

For simulating BC concentration over the Arctic a chemical transport modeling system is used. The simplified research design and modeling system applied to this study is shown in a flowchart in figure 1. This modeling simulation time period was from April 2008 to July 2009 with hourly output. The Sulfur Transport and dEposition Model (STEM) is used to simulate the concentration of pollutants in the atmosphere. The STEM model is a regional scale chemical transport model developed at the University of Iowa and had been utilized in several field campaigns.⁶ This model includes chemicals and particles transport, emissions and deposition. This model is investigating the convective-diffusion equation (Eq. 1) with the Eulerian approach to calculate the concentration of compound i.⁶,⁷

\[
\frac{\partial c_i}{\partial t} + \nabla (\nu c_i) = \nabla . K \nabla c_i + R_i + S_i + G_i \quad (1)
\]
where $c_i$ is the gas phase concentration of compound $i$, $\nu$ is the wind velocity vector, $K$ is for eddy diffusivity tensor, $R_i$ is the total reactions of species $i$, $S_i$ denotes the sources for species $i$, and $G_i$ is the mass transfer between gas and liquid.

The Emissions Database for Global Atmospheric Research (EDGAR) provides emission for different air pollutants by country and spatial resolution of $0.1$ deg. x $0.1$ deg. grid maps. For this study the EDGAR Hemispheric Transport of Air Pollution Version 2 (HTAP_V2) is used as the input emission inventory.

### 2.1.2. Meteorology Model

In this study the Weather Research and Forecasting model (WRF) is used for producing the meteorological factors that are needed for STEM model input. The ice sheet coverage, initial and boundary conditions for the model are provided by the National Center for Environmental Predication (NCEP) Final Analysis (FNL, http://rda.ucar.edu/datasets/ds083.2/). The meteorological factors affecting chemical distribution and concentration were imported into the STEM model every 6 hours.

### 2.1.3. Modeling Domain

In this research, the modeling domain for WRF and STEM models covered most of the Northern hemisphere including significant emission sources such as Asia, Russia, Europe and North America. Also the model extends over the Northern Africa, Middle East and South Asia. The modeling domain of study is shown in figure 2. The models use polar stereographic map projection with 60 km horizontal resolution (249x249 grid cells). This modeling system is described in further details in Kulkarni et al. 2015 and D’Allura et al. 2011. The modeling domain of study is shown in figure 2.
2.2. Observations

For evaluating the modeling system performance the output is compared to aircraft data from the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) field campaign. ARCTAS study was conducted by National Aeronautics and Space Administration (NASA) during spring and summer 2008 to study the factors influencing climate change in the Arctic atmosphere. Furthermore, the surface concentration data is evaluated using annual average data from 150 IMPROVE (Interagency Monitoring of Protected Visual Environment) sites over North America.

2.3. Wet and Dry deposition calculation

Liu et al. 2011 shows that the representation of wet and dry deposition removal in the model has a profound effect on the result of BC modeling over the Arctic. The removal processes of aerosol and gases from the atmosphere happen in two main forms of dry and wet deposition. Dry deposition occurs when an aerosol particle diffuses to the surface or sediments to the ground due to gravitational forces. Wet deposition occurs when atmospheric precipitations scavenge aerosol particles. For this study the dry deposition process of particles was modeled using the “Resistance in Series Parameterization” from Wesley and Hicks 2000. The wet deposition is calculated using precipitation output from WRF model as an hourly rate. Deposition amounts are accumulated by calculating the differences between concentrations at each transport time step (i.e. 15 min) between the original BC concentration and BC concentration output from model when deposition rates are set to zero. The Eq. 2 and Eq. 3 show the method for calculating BC dry and wet deposition amount.

\[
BC_{DD} = BC_{(DD\ rate =0)} - BC_{(original)} \quad (2)
\]
\[ BC_{\text{WD}} = BC_{(\text{WD rate}=0)} - BC_{(\text{original})} \quad (3) \]

In the above equations BC_DD and BC_WD denote BC dry and wet deposition values respectively. BC_{(DD rate=0)} and BC_{(WD rate=0)} are BC concentration when dry and wet deposition rates are set to zero respectively. BC_{(original)} is the original BC concentration output from the model with non-zero BC dry and wet deposition rates. Dry and wet deposition variables are reset to the original values after each transport time step.

### 3. Results and Discussion

#### 3.1. Spatial Distribution

Figure 2 presents the annual average concentration of surface BC over the modeling domain. Figure 2 shows that the modeled BC surface concentration is in the range of \(~0.25\) to \(3 \mu g/ m^3\). The major BC hotspots are over Southeast Asia, Northern India and China. Furthermore, the seasonal and monthly results show that BC concentration peaks during wintertime since there are higher biomass and fossil fuel burning for heating during the winter season. The annual average surface concentration over the U.S. is \(0.16 \mu g/m^3\) with the maximum BC over the Eastern U.S. with average of \(0.75 \mu g/m^3\). The highest annual BC concentration is \(~3 \mu g/m^3\) over China and Northern India. The annual average BC for the Arctic area (altitude higher than \(60^\circ\) N) is between \(~0.025 \mu g/m^3 – 0.075 \mu g/m^3\) with the minimum occurring over Greenland, Alaska and Northern Canada. This value is consistent with the average of \(0.06 \mu g/m^3\) over the Arctic from Sharma et al. 2013.\(^9\)
3.2. Model Evaluations

3.2.1. Meteorological Model Evaluation

We have evaluated the meteorological model results using 22 flights data from NASA ARCTAS field campaign. The results show that the WRF model captures the meteorological model accurately at different altitudes.

Table 1 shows the statistical summary of the major meteorological variables for both ARCTAS observation data and model output. Based on this table and box and whisker plots analysis the model captures the vertical profile and magnitude of meteorological values correctly. However, the model tends to over-predict wind speed by 4% at higher altitudes.

Table 1- Statistical summary of comparison of observed and modeled meteorological parameters for NASA ARCTAS spring and summer flights. Obs and Mdl denote observation and model data

<table>
<thead>
<tr>
<th></th>
<th>Temperature(K)</th>
<th>Pressure(hpa)</th>
<th>Relative Humidity(%)</th>
<th>Wind Speed(m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Obs</td>
<td>Mdl</td>
<td>Obs</td>
<td>Mdl</td>
</tr>
<tr>
<td>Mean</td>
<td>248.4</td>
<td>263.1</td>
<td>610.2</td>
<td>594.6</td>
</tr>
<tr>
<td>Standard Error</td>
<td>0.3</td>
<td>0.3</td>
<td>3.7</td>
<td>3.4</td>
</tr>
<tr>
<td>Median</td>
<td>245.4</td>
<td>265.7</td>
<td>554.9</td>
<td>569.0</td>
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<tr>
<td>Mode</td>
<td>225.0</td>
<td>231.4</td>
<td>1007.0</td>
<td>329.3</td>
</tr>
<tr>
<td>Standard Deviation</td>
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<td>23.1</td>
<td>253.2</td>
<td>232.8</td>
</tr>
<tr>
<td>Range</td>
<td>94.8</td>
<td>93.6</td>
<td>818.9</td>
<td>817.2</td>
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<tr>
<td>Minimum</td>
<td>212.7</td>
<td>212.2</td>
<td>206.7</td>
<td>187.1</td>
</tr>
<tr>
<td>Maximum</td>
<td>307.4</td>
<td>305.8</td>
<td>1025.6</td>
<td>1004.2</td>
</tr>
</tbody>
</table>

3.2.2. Concentration Evaluation

The air pollution concentrations values from STEM model are evaluated using data from 150 IMPROVE sites over the U.S. for the time period of April 2008 to July 2009 (http://vista.cira.colostate.edu/improve/Data/IMPROVE/AsciiData.asp). Figure 3 shows the
annual mean surface BC concentration over the U.S. compared with observations at IMPROVE network sites. Each site represented as a circle in the map. Average model BC over the U.S. is 0.16 µg/m³ while average IMPROVE data is 0.19 µg/m³. Further statistical analysis shows that the root-mean-square deviation (RMSE) between model and observation is 32% and the mean bias error (MBE) is 0.03 µg/m³. The mean simulatedMODELED BC concentration is 1.2 while average global model simulated/ModeLED BC is 1.6 over North America. Comparing with other global models, STEM model showed a better performance in simulating the concentration.

Modeling BC distribution and concentration over the Arctic is considered a challenging task for global chemical transport models. Previous model inter-comparison shows order of magnitude differences between observation and model. Shindell et al. 2008 and Koch et al. 2009 show negative bias between model and observation. Shwartz et al. 2010 shows positive bias comparing global models with observation. This difference between model performances is largely due to the high uncertainty in emission and scavenging efficiency for calculating wet deposition. Regional chemical transport models with focus on the Arctic captured the BC concentration better over the arctic. Koch et al. 2009 and Liu et al. 2011 captured the ARCTAS flights vertical profiles and seasonality well.

3.4. Source Sector Contribution to BC

Due to significant contribution of BC in global warming over the Arctic, it becomes extremely important to understand the influence of specific source region of BC over the Arctic. Stohl et al. 2007 suggests that high altitude boreal forest fire is one of the main contributors to amount of BC over the Arctic. In this study the impact of emission sector categories including residential, transportation, biomass burning, power, and industry is investigated using a series of sensitivity runs. The time series contribution from five emission sectors to BC surface
concentration over the Arctic is shown in figure 4 top panel. In this study the area average surface concentration for altitude 60°N and above is used. The surface concentration range from 0.05 μg/m$^3$ to 0.2 μg/m$^3$ over the Arctic. The maximum BC concentration occurred during wintertime. The contribution from residential sector significantly increases during wintertime since burning biofuels and coal are the main heating resource in South Asia and India. Furthermore, there is high seasonal variability in contribution of biomass burning concentration with the minimum occurring in the winter. There are higher events of European and Siberian forest fires during spring and summer period.

3.3. Source Region Contribution to BC

Koch and Hansen 2005 study shows that regions as far away as Southeast Asia are important contributor to BC concentration over the Arctic.$^{10}$ BC emissions from different source regions (i.e. Europe, China, North America, Central Asia, Middle East, South Asia, Central Asia and Siberia) were studied using sensitivity analysis. Figure 4 bottom panel shows the contributions of different emission regions to BC surface concentrations. Europe and China have the highest impact on the Arctic surface concentration with ~35% and ~25% contribution. Emission from Russia and Continental U.S. (CONUS) are also significant with contributions of ~15% and 7% respectively. Approximately less than 5% percent of emission is transported from each of South Asia and Middle East to the Arctic. In the bottom panel of figure 4 the regions with contributions of less than 0.001 μg/ m$^3$ are not shown. The Russian emission includes both anthropogenic and Siberian wild fires emissions.

The peak BC concentration is occurring during the wintertime; however, the contribution of biomass burning in Siberia (Russia Emission) significantly increases during spring and summer period. There are higher open biomass burning emissions from forest and grassland
burning happening during hot seasons. There is 5-15% difference between the sum of region contributions and sector contributions due to the different averaging processes and the transport between regions outside the domain of study.

### 3.5. BC Wet and Dry Deposition in the Arctic

BC transports from different regions in mid-latitudes to the Arctic and eventually deposits on the Arctic snow. BC transported to the Arctic in low altitudes are more likely to deposit on snow and BC from higher altitudes are generally ventilated out of the Arctic depends on meteorological conditions. BC deposited on the Arctic snow and sea ice reduces radiative forcing of snow causing it to warm and melt more quickly.\(^1\)\(^4\)

Wet and dry deposition flux of BC are investigated separately for different months to study the seasonality. There are higher BC dry deposition occurring in spring time and wintertime. The wet deposition reaches its maximum during spring and summer due to higher precipitation in these seasons. Major hotspots of dry deposition are over China and Southern Asia since there is higher BC concentration over this region. Wet deposition is high in Northern India and Southeastern Asia due to higher precipitation during monsoon months.

Wang et al. 2011 study over the Western Arctic during Spring 2008 shows that the average BC dry deposition flux is 45 µg/m\(^2\).month.\(^3\) Jiao et al. 2014 summarizes the results of 25 different modeling studies and reports the average BC surface concentration over the Arctic (altitudes 60° N and above) is between 250-1670 µg/m\(^2\).month with seasonal variability.\(^14\) Our model value is well in the range of previous studies over the Arctic.
4. Conclusion

We used WRF-STEM chemical transport model for simulation BC concentration and deposition over the Arctic for time period of April 2008-July 2009. NASA ARCTAS flight data and IMPROVE network surface observation data are used to evaluate the model performance. The modeling system captures observational data magnitude and variations correctly.

In this study our focus was to quantify contribution of each source sector and source region to the concentration and deposition of BC over the Arctic. The results show that the major sector contributor to BC concentration is residential and the main source region contribution is from Europe and China annually. There are higher BC dry deposition occurring in winter and spring. The wet deposition maximum happens during spring. Future works include calculating BC content in snow and compare it with observation samples from the Arctic. The effect of BC dry and wet deposition on the Arctic radiative forcing change can be calculated Using BC content in snow. Due to the short lifetime of BC making new policy for mitigating can help reducing BC fast and reduce its environmental and health effect. Therefore, quantifying the source sector and regional contributions to BC concentration in the Arctic is very important for making new international policies.
5. References


6. Figures

Fig 1- Schematic diagram showing WRF-STEM modeling system and design.

Fig 2- WRF- STEM modeling domain setup. The color scheme represents simulated annual mean BC concentration (µg/m³).

Fig 3- Annual average surface BC concentration over the U.S. The circles indicate IMPROVE sites with the color representing the BC concentration in µg/m³.
Fig 5- Temporal variability and contributions of each source sector (top panel) and source region (bottom panel) to average BC concentration over the Arctic. BB denotes biomass burning.

Fig 6- Spatial plots of geographical source contribution (%) to total BC deposition.