Measurements of Wet Deposition of Black Carbon in the Arctic Field Campaign Report

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March 2018
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Measurements of Wet Deposition of Black Carbon in the Arctic Field Campaign Report

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### Acronyms and Abbreviations

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<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>AMAP</td>
<td>Arctic Monitoring and Assessment Programme</td>
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<tr>
<td>ARM</td>
<td>Atmospheric Radiation Measurement</td>
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<tr>
<td>BC</td>
<td>black carbon</td>
</tr>
<tr>
<td>CMBC</td>
<td>black carbon mass concentration</td>
</tr>
<tr>
<td>CNBC</td>
<td>black carbon number concentration</td>
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<tr>
<td>DOE</td>
<td>U.S. Department of Energy</td>
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<tr>
<td>ESM</td>
<td>Earth System Model</td>
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<tr>
<td>NILU</td>
<td>Norwegian Institute for Air Research</td>
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<tr>
<td>NOAA</td>
<td>National Oceanic and Atmospheric Administration</td>
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<td>SP2</td>
<td>single-particle soot photometer</td>
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1.0 Introduction

Black carbon (BC) aerosols are light-absorbing carbonaceous particles emitted globally by incomplete combustion, open biomass burning, and biofuel burning (Bond et al., 2013; Kondo 2015). Climate model calculations indicate that the global emissions of BC contribute to arctic warming through heating in the atmosphere and by lowering the albedo of snow and ice after deposition (e.g., Hansen and Nazarenko, 2004; Jacobson 2004, 2010; Flanner et al., 2007, 2009; Koch et al., 2009; Shindell et al., 2012; Bond et al., 2013). BC particles distributed in the Arctic have been shown to originate dominantly from lower latitudes (AMAP 2015). Once emitted, they can be transported in the atmosphere towards and within the Arctic and removed by wet and dry deposition along their transport pathways; these removal processes influence the distributions of BC in the atmosphere and snow in the Arctic (Liu et al., 2011; Mahmood et al., 2016).

Model calculations indicate that the importance of the removal of BC outside the Arctic relative to that in the Arctic depends on various factors, including the seasons and locations (Shindell et al., 2008; Browse et al., 2012; Jiao and Flanner 2016). Attempts have been made to validate schemes expressing the deposition processes in global models by comparison of calculated BC mass concentrations in surface snow with measured concentrations in the Arctic (e.g., Wang et al., 2011; Qi et al., 2017).

The uncertainties of the techniques widely used for measurements of BC in snow in the Arctic in previous studies (e.g., Forsström et al., 2009; Doherty et al., 2010, 2016) are large or have not been fully evaluated. Few accurate measurements have been made of the concentrations and size distributions of BC in snow that can be compared with model calculations reliably. Macdonald et al. (2017) and Sinha et al. (2018) measured the concentration of BC in freshly fallen snow at a high temporal frequency throughout the entire cold season of 2014–2015 at Alert (82°N) in Canada and at Ny-Ålesund (79°N), Spitsbergen, using a technique similar to that used in the present study. Still, very few BC measurements have provided time-resolved quantitative BC flux data throughout the seasons. Additional measurements of this type are needed to characterize wet and dry deposition of BC during the season.

2.0 Methods

2.1 Measurements of BC in Water

We measured BC number and mass concentrations (CNBC and CMBC) in snow and rain samples using a single-particle soot photometer (SP2) combined with a nebulizer (Nebulizer-SP2). The effect of the coagulation of BC particles suspended in water on the measurement of BC size distribution has been shown to be negligible under typical BC number concentrations in water based on theoretical calculations and laboratory experiments (Moteki and Mori 2015; Mori et al., 2016). The stability of the CMBC values during the storage of water samples was evaluated using rainwater samples (Ohata et al., 2013; Mori et al., 2016). The diameter range of BC measured by SP2 used to be about 70-4 170 nm (Mori et al., 2016).

The size distributions of BC are very useful for detailed studies of the microphysical processes of wet removal of BC particles (Moteki et al., 2012; Kondo et al., 2016; Ohata et al., 2016) and for understanding the microphysical properties of BC in snow and rain. Size distributions are also needed to
calculate the radiative effects of airborne BC particles and of those deposited on snow (Schwarz et al., 2013; Moteki et al., 2017). We calculated CNBC and CMBC by integrating the number and mass size distributions.

### 2.2 Collection of Snow and Rain Samples

In this study, we measured BC concentrations in falling snow and rain collected in situ during each precipitation event. For real-time sampling of falling snow and rain, the timing of BC deposition is determined with an uncertainty of several hours. We collected falling snow and rain at the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facility observatory at Barrow, Alaska. We let snow and rain collect in a plastic container and rain collector. The principle snow-sampling system is similar to that using “Box” described in Sinha et al. (2018), although somewhat different in details. We collected rain samples using a manual rain sampler positioned on the top of the building. The manual rain sampler used a funnel and a glass bottle to minimize possible loss of BC on the inner wall. Rainwater was collected daily by the rain collector. The samplings were made in 2013-2017.

### 3.0 Temperature and Precipitation

We used the meteorological data of precipitation and temperature obtained near the Barrow airport. The data of daily temperature and precipitation are available from the Alaska climate research center (http://climate.gi.alaska.edu/acis_data). Figure 1 (upper panel) shows the daily mean temperature at Barrow between 2013 and 2017. The monthly mean temperature for the same period is also shown (lower panel). The temperatures were below 0°C between October and May on average.

![Figure 1](image1.png)

**Figure 1.** Daily mean temperature (upper panel) and monthly mean temperature (lower panel) for the period of 2013-2017 in Barrow.
Figure 2 shows the daily precipitation for snow and rain 2013 and 2017. The monthly mean precipitation for the same period is also shown (lower panel). Most of the snowfall occurred between September and May and rainfall occurred between May and October on average, corresponding to the seasonal variation of the temperature.

![Figure 2. Daily mean precipitation (upper panel) and monthly mean precipitation (lower panel) for the period of 2013-2017 in Barrow.](image)

4.0 BC in Water

4.1 BC Size Distributions

Figure 3 shows the average number size distributions in snow and rain for each season. The number size distributions did not show significant seasonal variations.
Figure 3. Normalized number size distribution of BC in snow and rain averaged for each season.

Figure 4 shows the average mass size distributions in snow and rain for each season. The mass size distribution was similar for snow and rain. The size distributions significantly shifted to larger diameters in fall.

Figure 4. Normalized mass size distribution of BC in snow and rain averaged for each season.

We used BC mass per particle (m_{BC}), derived as the C_{MBC}/C_{NBC} ratio and the fraction of BC mass in the diameter range larger than 600 nm, denoted as f_{600}, as measures of the size distributions. These values are shown in Figure 5. The average and median m_{BC} and f_{600} values are given in Table 1. These values are
consistent with the mass size distributions (Figure 4). Namely, the $m_{BC}$ and $f_{600}$ values are significantly larger in fall than those in the other seasons. The size distributions of BC in rain and snow should reflect those in air where clouds form. It is likely that BC size distribution in ambient air aloft shifted to larger sizes in fall. There is a need to investigate the origins of air parcels in different seasons.

Figure 5. Time series plot of $f_{600}$ and $m_{BC}$.

Table 1 Median (mean ± standard deviation) values of $f_{600}$ and $m_{BC}$ in falling snow and rain at Barrow (this study).

<table>
<thead>
<tr>
<th></th>
<th>Rain</th>
<th>Snow</th>
<th>Rain</th>
<th>Snow</th>
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<tbody>
<tr>
<td>$f_{600}$</td>
<td>0.22 (0.22±0.15)</td>
<td>0.25 (0.25±0.12)</td>
<td>5.6 (5.6±2.2)</td>
<td>5.6 (7.8±4.5)</td>
</tr>
<tr>
<td>$m_{BC}$ (fg)</td>
<td>5.6 (5.6±2.2)</td>
<td>5.9 (6.0±1.6)</td>
<td>9.8 (15±13)</td>
<td>10.4 (9.7±4.3)</td>
</tr>
</tbody>
</table>

4.2 BC Number and Mass Concentrations

The time series of the daily mean number and mass concentration of BC in snow and rain ($C_{NBC}$ and $C_{MBC}$) are shown in Figures 6 and 7 (upper panels). In these figures, the mass concentration of BC in ambient air ($M_{BC}$) measured at the National Oceanic and Atmospheric Administration (NOAA) facility at Barrow are also shown. The measurements of $M_{BC}$ are described in detail in Sinha et al., 2017. The $C_{MBC}$ (and $C_{NBC}$) in snow and $M_{BC}$ seems to be correlated to some extent. However, more detailed investigations are needed to quantify this point. $C_{MBC}$ and $C_{NBC}$ in rain showed a large variability, uncorrelated with $M_{BC}$, suggesting that ambient concentration near the surface and aloft may be significantly different. There is a need to identify the altitudes influenced by precipitation. The measurements of thick clouds by cloud radar at the ARM site will be useful for this analysis.
Figure 6. Upper panel: daily mean values of $M_{BC}$ and $C_{NBC}$. Lower panel: daily values of precipitation and $F_{NBC}$.

Figure 7. Upper panel: daily mean values of $M_{BC}$ and $C_{MBC}$. Lower panel: daily values of precipitation and $F_{MBC}$.

4.3 BC Deposition Flux

We derived the wet deposition flux of BC from the measurements of CNBC and CMBC in snow and rain. For the $i$-th precipitation event, BC number and mass deposition fluxes, denoted as $F_{NBCi}$ (\# m$^{-2}$ d$^{-1}$) and $F_{MBCi}$ (\µg m$^{-2}$ d$^{-1}$), respectively, are given by the following equations:

$$F_{NBCi} = C_{NBCi} \times P_i$$  \hspace{1cm} (1)

$$F_{MBCi} = C_{MBCi} \times P_i$$  \hspace{1cm} (2)
where \( C_{\text{NBC}} \) and \( C_{\text{MBC}} \) are the number and mass concentrations of BC in the \( i \)-th snow sample, respectively, and \( P_i \) (mm d\(^{-1}\)) is the amount of precipitation during the corresponding precipitation event.

To derive BC wet deposition fluxes per month, we took into account all of the precipitation events recorded by the meteorological observations, even for days when falling snow or rain was not sampled. Considering this factor, monthly BC number and mass deposition fluxes were defined as

\[
(F_{\text{NBC}})_j = \frac{\sum F_{\text{NBC}}}{R_j} \\
(F_{\text{MBC}})_j = \frac{\sum F_{\text{MBC}}}{R_j},
\]

where \( R_j \) is the ratio of precipitation summed over all snow sampling events in the \( j \)-th month to the total monthly precipitation amount \( P_j \). \( R_j \) is given as

\[
R_j = \frac{1}{P_j} \sum P_i.
\]

The derived \( F_{\text{NBC}} \) and \( F_{\text{MBC}} \) are shown in Figures 6 and 7 (lower panels). The monthly BC number and mass deposition fluxes are shown in Figures 8 and 9, respectively. The BC fluxes did not show a marked seasonal variation. This is partly occasional high BC fluxes in summer time. A more detailed analysis is made to interpret the observed BC flux in depth.

**Figure 8.** Upper panel: monthly mean values of \( M_{\text{BC}} \) and \( C_{\text{NBC}} \). Lower panel: monthly values of precipitation and \( F_{\text{NBC}} \).
Figure 9. Upper panel: monthly mean values of $M_{BC}$ and $C_{MBC}$. Lower panel: monthly values of precipitation and $F_{MBC}$.

5.0 Comparison with Other Data Sets

We made sampling of snowpack in Alaska, including Barrow, in February and March in 2012-2015. The locations of the sampling are shown in Figure 10. Relevant parameters measured north of about 66°N are summarized in Table 2 (unpublished data). The values of the two data sets are generally comparable, considering possible year-to-year variability.

Figure 10. Locations of sampling of snowpack in Alaska.
Table 2. Median values of $C_{\text{MBC}}$, $C_{\text{NBC}}$, and $m_{\text{BC}}$ in snowpack sampled between February and March in 2012-2015.

<table>
<thead>
<tr>
<th></th>
<th>66-69°N</th>
<th>Barrow</th>
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<tbody>
<tr>
<td>$C_{\text{MBC}}$ (μg L$^{-1}$)</td>
<td>2.4±1.9</td>
<td>1.3</td>
</tr>
<tr>
<td>$C_{\text{NBC}}$ (10$^8$ L$^{-1}$)</td>
<td>3.4±2.1</td>
<td>1.2</td>
</tr>
<tr>
<td>$m_{\text{BC}}$ (fg)</td>
<td>6.7</td>
<td>11.3</td>
</tr>
</tbody>
</table>

6.0 Plans for Further Analysis

We need to identify the altitude regions where BC particles act as condensation nuclei, are included in cloud particles, and are precipitated out. A more direct way is to use the cloud data obtained by a cloud radar and a lidar at the ARM site. Thick cloud layers should potentially contribute to precipitation. Meteorological conditions will be related to observed precipitation.

We plan to make 3-dimensional model calculations with the Earth System Model (ESM) developed at the metrological institute, Japan. The model will provide BC concentrations in snow and rain and wet deposition flux of BC. By this comparison, we will be able to interpret the seasonal variations of BC in water. The comparison will also be useful in validating the schemes of BC wet deposition used in the model. We also plan to cooperate with NILU (Norwegian Institute for Air Research) in estimating source regions of BC removed by precipitation. The FLEXPART model, which is a sophisticated trajectory model, is to be used for this purpose. The comparisons with the models will improve our understanding of the effect of BC on climate in the Arctic.

7.0 Summary

We made the measurements of BC in falling snow and rain, together with the ambient BC concentrations at Barrow, for about three years for the first time. We derived seasonal variations of BC size distributions and concentrations in water, and wet deposition flux with high accuracy. These data sets are useful in validating BC deposition schemes of climate models, which are used to assess the effect of BC on the climate in the Arctic.

8.0 References

AMAP. 2015. AMAP Assessment 2015: Black carbon and ozone as Arctic climate forcers. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. vii + 116 pp.,


