Master’s thesis

Master’s Degree Programme in Atmospheric Sciences

DISTRIBUTION OF BLACK CABRON IN NORTHERN EUROPE AND THE ARCTIC, AND RELATIONS WITH CLIMATE FORCING AND AIR QUALITY

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In loving memory of my Mother

Sita Gaire

(1967-2008)
Abstract

The objective of this master’s thesis was to better understand the impact of black carbon and its distribution in Northern Europe and the Arctic. To achieve the goal of the project, information on the observations relevant to black carbon (BC) pollution in Arctic dataset was collected. For the observational data all main BC measurement campaigns along with active satellite operations were collected.

In this study, the BC concentration and deposition was estimated by the System Integrated Modelling of Atmospheric composition - a chemical transport model (CTM) SILAM. The model was driven with monitoring atmospheric composition and climate (MACCity), emission database for global atmospheric research hemispheric transport of air pollution (EDGAR-HTAP), and evaluating the climate and air quality impacts of short lived pollutants (ECLIPSE) emission inventories. For the computations, the year 2010 was chosen because of a better availability of data during that year. In the literature section, black carbon process in the atmosphere is explained along with its properties and characteristics. Furthermore, data description and data analysis is included which is followed by interpretation of model output on the seasonal deposition and concentration.

As shown by the model-measurement comparison, the model basically captured the measured BC and organic carbon (OC) quite well for all emission inventories. However, the correlation coefficient for OC was still weak for most of the stations in Europe. The overall performance of BC for European stations is substantially better than in the Arctic areas. Deposition for BC and OC shows that the seasonal transport of BC from source regions is evident in the Arctic and near Arctic areas. Patterns of dry deposition is higher in winter period than in summer period. The SILAM model suggests winter period concentration of BC by MACCity and ECLIPSE inventory of 0.23 μg/m³ and 0.26 μg/m³ respectively for year 2010.

This study provides a best performing setup for BC modeling, transport and deposition of BC in the Northern Europe and the Arctic despite the absence of ageing process. More
observational data from Arctic stations would provide better result and model performance. Finally, the study gives an insight of the quality of existing emission inventories and the capabilities of reproducing seasonal deposition and concentration of BC to the Arctic.
# Abbreviation

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tr>
<td>ACCMIP</td>
<td>Atmospheric chemistry and climate model intercomparison project</td>
</tr>
<tr>
<td>AMAP</td>
<td>Atmospheric monitoring and assessment programme</td>
</tr>
<tr>
<td>BC_d</td>
<td>Black carbon daily measurement</td>
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<td>BC_h</td>
<td>Black carbon hourly measurement</td>
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<tr>
<td>BC</td>
<td>Black carbon</td>
</tr>
<tr>
<td>CCN</td>
<td>Cloud condensation nuclei</td>
</tr>
<tr>
<td>CORR</td>
<td>Correlation coefficient</td>
</tr>
<tr>
<td>eBC</td>
<td>Equivalent black carbon</td>
</tr>
<tr>
<td>EC</td>
<td>Elemental carbon</td>
</tr>
<tr>
<td>ECCAD</td>
<td>Emissions of atmospheric compounds and compilation of ancillary data</td>
</tr>
<tr>
<td>ECLIPSE</td>
<td>Evaluating the climate and air quality impacts of short lived pollutants</td>
</tr>
<tr>
<td>EDGAR-HTAP</td>
<td>Emission database for global atmospheric research Hemispheric transport of air pollution</td>
</tr>
<tr>
<td>EMEP</td>
<td>European monitoring and evaluation programme</td>
</tr>
<tr>
<td>FMI</td>
<td>Finnish Meteorological institute</td>
</tr>
<tr>
<td>GCS</td>
<td>Geographical coordinate system</td>
</tr>
<tr>
<td>GRADS</td>
<td>Gridded analysis and display system</td>
</tr>
<tr>
<td>IASOA</td>
<td>International Arctic system for observing the atmosphere</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Description</td>
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<tr>
<td>IPCC</td>
<td>Intergovernmental panel on climate change</td>
</tr>
<tr>
<td>NIES</td>
<td>National institute of environmental studies</td>
</tr>
<tr>
<td>OC</td>
<td>Organic carbon</td>
</tr>
<tr>
<td>RCP</td>
<td>Representative concentration pathways</td>
</tr>
<tr>
<td>RMSE</td>
<td>Root mean square error</td>
</tr>
<tr>
<td>SILAM</td>
<td>System for integrated modeling of atmospheric composition</td>
</tr>
<tr>
<td>SLCF</td>
<td>Short lived climate forcers</td>
</tr>
<tr>
<td>SSF</td>
<td>Surface scattering albedo</td>
</tr>
<tr>
<td>SOA</td>
<td>Secondary organic aerosol</td>
</tr>
<tr>
<td>STP</td>
<td>Standard temperature pressure</td>
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Acknowledgement

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1. Introduction

Due to annual temperature rise and global climate change, several pollutants have recently received much more attention (e.g. Hirdman et al., 2010; Quinn et al., 2008). Climate change is a complex phenomenon comprising several atmospheric processes and their feedbacks (e.g. Lund, 2008). Several global models such as modelE and GATOR-GCMM and observations suggest that the Arctic is vulnerable to climate change where melting of sea ice and snow cover increases dramatically. (Koch et al., 2005; Manabe et al., 1992). The Arctic temperature is increased by twice the global rate over last 100 years (IPCC 2007; Quinn et al., 2007). Fourth assessment report of the Intergovernmental Panel for climate change (IPCC) indicates that aerosol particles affect the climate system on radiation budget, meteorological system and interactions with the hydrological cycle (IPCC, 2015).

Atmospheric aerosol is the suspension of solid or liquid particles in the atmosphere. Aerosols are important climate forcers, but the magnitude of forcing are highly uncertain and depends on altitude, cloud distribution, surface albedo and optical properties of aerosols (Eckhardt et al., 2015). It is generally accepted that absorbing aerosols particles, (mainly black carbon) are the key atmospheric particulate components affecting climate change and air quality by absorbing incoming short and long wave radiation (Eckhardt et al., 2015; Petzold et al., 2013). In addition to surface radiative forcing, deposition of absorbing aerosols on snow and ice leads to faster melting and plays a role in the surface warming (Eckhardt et al., 2015). Due to its direct and indirect effects on the global energy budget it is of great interest to understand past and present effects on Arctic climate.

Black carbon is primarily formed by incomplete combustion of fossil fuels, from off and on-road vehicles, biomass and biofuels burning, industrial emissions and domestic combustion. (Bond et al., 2012; Quinn et al., 2011). In the context of climate change, the term forcing refers to changes in the radiation balance of the surface-troposphere system imposed by external factors, with no changes in stratospheric dynamics, without any surface and tropospheric feedbacks in operation. It has been demonstrated that BC aerosols can decrease
the albedo of snow/ice, resulting in a global radiative forcing of \(+0.10 \pm 0.10\) Wm\(^{-2}\) (Eleftheriadis et al., 2009).

Pollutant levels in the Arctic are especially important for the climate change. Black carbon (BC), has a strong local climate impact when it is deposited onto snow and ice surfaces, reducing their albedo and enhance heating of snow leading to rapid melting, in turn leading to larger albedo change (Vignati et al., 2010; Schindell et al., 2008).

Pollution sources in the Arctic areas are scarce, hence the transportation of pollutants from emitting source regions in mid- and low-latitudes are the main driving force for pollution. Due to a complex meteorological system in the Arctic, such as the formation of the polar vortex during winter leading to stably stratified atmosphere, there is a strong seasonal cycle of the transport patterns. During spring time, polar vortex weakens which allows greater exchange of particles and gases in the upper level. (Koch and Hansen 2005) (Koch et al., 2005) (Eleftheriadis et al., 2009). Transport pathways vary with the seasons and the highest concentrations in the Arctic are observed during winter and early spring and lowest in summer. The polar front during winter lies at about 50°N hence allowing long range transport from major industrial regions in Europe, Russia and North America into the Arctic. During summer the front lies in 70° N which prevents the pollution and air masses from effectively reaching the Arctic. (Eleftheriadis et al., 2009). Seasonal variability of BC concentration is caused mainly by transport pathways to the Arctic and due to wet deposition on the way towards deposition (Ruppel et al., 2013).

It is shown that several aged particles from long range transport are more abundant during the winter and spring period than in summer. (Huang, 2010). Only long range transport mechanism cannot explain the observed seasonality and uncertainty in the modeling of Arctic BC. Therefore, reproducing seasonality of atmospheric removal process is investigated furthermore to close the knowledge gap from this research (Huang, 2010).

Several models have been used to capture the seasonal distribution of aerosols in the Arctic areas (Shindell et al., 2008; Koch et al., 2009; Eckhardt et al., 2015), including source areas
and remote areas. Especially during the haze season, most of the models underpredict BC near surface concentrations compared to observations near the Arctic regions (Sharma et al., 2013). The factors responsible for the underprediction could be a poorly modeled chemical transformation, transport, parameterization of BC ageing and scavenging processes. (Sharma et al., 2013). Large uncertainties of model prediction originate in black carbon emission (Koch et al., 2009; Vignati et al., 2010). For the better model performance, the atmospheric carbon cycle should be evaluated (Vigantti et al., 2010).

This research approach is to explore the capabilities of system for integrated modelling of atmospheric composition chemical transport model (SILAM-CTM) to simulate BC and OC in the Northern Hemisphere for year 2010. The model simulates concentration and deposition using MACCity, EDGAR and ECLIPSE emission inventories. The model includes modern original parameterization for dry and wet deposition.

Seasonal distribution of BC is accomplished with deposition and concentration comparison with available observations. Finally, model results for different emission inventories are compared with each other and the typical features of BC transport to Northern Europe and the Arctic are revealed.

This research is structured as follows: first sections gives the brief introduction and objective of the study. Subsequently, section 2 provides the theoretical background of black carbon features and characteristics of Arctic areas. The third section explains the study methodology; and the fourth section interprets the results and comparisons. Conclusion and discussion will be drawn in the final section 5.

1.1 Objectives of research
The overall goal of the study was to understand and quantify the impact of black carbon deposition on snow-covered Arctic areas and Northern Europe. A specific technical question was “What could be the best performing model setup for BC modeling, transport and
deposition in the Arctic?" Further, the research also provides insight into “Quality of existing emission inventories and the model capability of reproducing seasonal deposition and concentration in the Arctic.”

The current assessment is limited to the basic transport and deposition processes, thus excluding dynamics of the ageing process (moderate BC hydrosopicity is assumed immediately at the emission moment) and cloud condensation nuclei formation.

1.2 Methods and materials

To support the study, an inventory of observation information is gathered, relevant to BC pollution of the Arctic. Identification of available emission inventories from the Northern Hemisphere and their features with regard to anthropogenic and natural BC emission is performed. This was followed with the model simulation of BC dispersion in the Northern Hemisphere and its transport to the Arctic, with selected emission inventories. The obtained model results are compared with the observations. The best performing set up and processes relevant for modeling of BC transport and deposition are identified. From the model output, the main feature of BC deposition in the Arctic is quantified.

This thesis focuses on absorbing aerosol i.e. BC, EC and OC. In addition, the average seasonal deposition and concentration in the Arctic and near Arctic areas is calculated and compared between two emission inventories. Also, carbon budget of BC deposition and concentration for whole domain is calculated and evaluated.

The datasets used in the research includes EMEP observational dataset of the year 2010, including MACCity, EDGAR, and ECLIPSE emission inventories. Also, for fire emission datasets from IS4FIRES is extracted (IS4FIRES, 2016). Data for Arctic stations is extracted from IASOA observational dataset.
The following software has been used in this thesis:

a) Grads version 2.0.2.oga.2 for analyzing result output of model run

b) Python 2.0 for model measurement comparison,

c) Matlab R2014a,

d) Arc GIS 10.3.1 Desktop
2. Theoretical background

Concepts related to BC and its transformations in the atmosphere are presented throughout this chapter. It covers the basics of black carbon, sources, transport, ageing phenomena and its deposition mechanism.

2.1 Basics of black carbon

Black carbon is defined as an ideal light absorbing substance composed of carbon based fuels (Petzold et al., 2013). It is distinguished from other forms of carbon on the basis of its properties such as refractory, insolubility in water, exists as an aggregate of small carbon spherules and its strong absorption of light at all wavelengths (Bond et al, 2013). Along with soot, other particles such as sulfur dioxide, nitrogen oxides and organic compounds are also emitted from the combustion.

Elemental carbon (EC) is a subset of BC that includes only carbon not bound to other elements, but possibly present in one or more of multiple allotropic forms (Petzold et al., 2013; Schwartz and Lewis, 2012). Measurements based on thermooptical method are referred to EC, whereas measurement based on light absorption methods is referred to BC. Organic carbon (OC) refers to the carbon mass with organic aerosol from organic precursor gases emitted from anthropogenic and natural sources.

Black carbon influences climate through multiple mechanisms like direct effect on radiation, snow/ice albedo effect and other effects (EPA-450/R-12-001 March 2012). The main effect is direct solar light absorption, which is influenced by processes in liquid, mixed phase and ice clouds and by deposition on snow and ice (Bond et al., 2013).

In the atmosphere, BC undergoes different changes after being emitted from the sources. The main mechanisms are described in figure (1)
BC comes from different sources like residential combustion, off and on road vehicles, industries and forest fires. It has all the varieties of composition and highly absorptive property so that it can absorb gases and coagulate with other particles. Some of the particles reach the clouds and serve as cloud condensation nuclei in favorable conditions. Along with transport, it undergoes chemical and physical transomation referred to as ageing. Ageing process results in the overall shift of particles from hydrophobic to hydrophilic state (Petzold et al., 2010). Processes contributing to ageing are condensation, coagulation, heterogenous nucleation and oxidation of organic material that coats the particles (Petzold et al., 2010). Ageing changes the life time and enables more efficient removal mechanism. Dry and wet deposition mechanism are the two removal processes from the atmosphere. Dry deposition is controlled by gravitational sedimentation, impaction and an interception. In addition particles are deposited because of force caused by different types of gradients and their
corresponding mechanisms include thermophoresis, diffusiophoresis and electrophoresis. Whereas, wet deposition is influenced by scavenging with snow flakes, fog and rain.

The current assessment is limited to the basic transport and deposition processes, thus excluding dynamics of the ageing process (moderate BC hydroscopticity is assumed immediately at the emission moment) and cloud condensation nuclei formation.

### 2.1.1 Sources of black carbon

The main part of the anthropogenic BC emission originates from the northern mid-latitudes with peaks around 35°N (Kristensen, 2013). BC is produced in a variety of combustion sources either natural or anthropogenic. At a global scale, major sources are diesel-powered off and on road vehicles, open burning of forest and savannas and solid fuels used for cooking and house heating. Industrial activities also account for a significant amount of emission whereas aviation and shipping emissions are minor (Bond et al., 2013).

The contribution of BC from North America and Europe each accounts ~40% of total BC deposition in the Arctic, while ~20% is from East Asia. (Schindell et al., 2008). From mid-latitudes, the developed nations have reduced BC emissions while there is an increasing trend in lower latitudes from the developing nations. (Schindell et al., 2008). From preceding findings, the dominant sources of BC for Arctic were found to contribute strongly in winter and early spring – more than during summer and autumn. Also, several case studies found that source regions such as agricultural and boreal fires in central and western Eurasia are contributing to the BC pollution in Arctic areas. (Hirdman et al., 2010)

### 2.1.2 Formation and properties of black carbon

Black carbon properties make it relevant to climate change, air chemistry, ambient air quality, biogeochemistry and paleochemistry. It is a distinct type of carbonaceous material formed primarily in flames during combustion of carbon based fuels when oxygen is insufficient for complete combustion. Even during the sufficient supply of oxygen, certain factors like operational condition, moisture content in fossil fuel, oxygen poor zones are not mixed well (Bond et al., 2013). The atmospheric lifetime of black carbon, its impact on
clouds, and its optical properties depend on interactions with other aerosol components in the atmosphere (Bond et al., 2006). The major properties and its consequences on atmosphere are described further in Table 1.

Table 1. Major properties, their characteristics and consequences in atmosphere (Petzold et al., 2013)

<table>
<thead>
<tr>
<th>Property</th>
<th>Characteristics</th>
<th>Consequences</th>
</tr>
</thead>
<tbody>
<tr>
<td>Structural appearance</td>
<td>Graphite like structure</td>
<td>Low chemical reactivity, slow removal</td>
</tr>
<tr>
<td>Morphology</td>
<td>Chain like aggregates</td>
<td>High absorption capacity</td>
</tr>
<tr>
<td>Temperature resistancy</td>
<td>Highly refractory material with volatilization temperature near 4000K</td>
<td>Low chemical reactivity, slow removal</td>
</tr>
<tr>
<td>Soluble character</td>
<td>Insoluble in most common solvents</td>
<td>Slow removal before coating and longer residence time</td>
</tr>
<tr>
<td>Absorption efficiency</td>
<td>Strong light absorption</td>
<td>Reduces albedo of snow, ice and darkens clouds</td>
</tr>
</tbody>
</table>

During the combustion process, small graphitic particles are formed which are in the order of tens of nanometers (Slowik et al., 2004). These graphitic particles change rapidly after the emission as they collapse into densely packed clusters (Martins et al., 1998) and take up water and other gas species also emitted during combustion. BC is hydrophobic shortly after emissions, but develops into increasingly hydrophilic or hydroscopic as it receipts gases, coagulates among neighbouring particles and undergoes several processes with species circumambient atmosphere.

A recent review of the observations of aerosol hygroscopicity from remote and urban regions reveals that hydrophobic particles are found only near emission sources. The above mentioned results endorse that the lifespan of hydrophobic particles is (in the order of)
hours. As BC-containing aerosol becomes more hydrophilic, intensity of removal from the atmosphere through in-cloud scavenging and precipitation increases (AMAP, 2011). On that account, the conversion of BC-containing particles from hydrophobic to hydrophilic changes the lifetime of the BC. Subsequently, after external mixing BC undergoes internal mixing and its light absorption is further intensified when coated with scattering material (Fuller et al., 1999).

### 2.1.3 Transport of black carbon

Pollution in the remote Arctic areas is the result of long range transport from source regions outside the Arctic areas. Transport to the Arctic troposphere is hindered by the Arctic front which is formed by surfaces of constant low potential temperature that form closed domes over the Arctic (Stohl et al, 2006).

![Transport process of black carbon into the Arctic](https://example.com/black-carbon-transport.png)

**Figure 2. Transport process of black carbon into the Arctic (Quinn et al., 2011)**

Stohl et al (2013) characterized three different paths for transport of pollution into the Arctic:
1. Low level transport followed by a rise in the Arctic when air is cold and dry

2. Low level transport alone, which involves transport over snow-covered surfaces

3. Uplift outside the Arctic, close to the sources by high altitude transport and descent in the Arctic

Pollution from Europe can pursue all three passages during the winter period and the first and third in summer period. But transport from Asia and North America can follow only the last path. The reason behind the high possibility of pollution transport in winter time is due to low temperature while minimum during summer is due to slow rate, higher temperature and efficient removal mechanisms. (Lund et al., 2008). The atmospheric lifetime of BC varies from several days to weeks. Due to its shorter lifetime, the effect is mostly regional. Maximal BC is observed in the Arctic during winter and early spring season, which is in connection with Arctic haze phenomenon. (Ruppel et al., 2015)

It is suggested that abundance in the Canadian Arctic was dominated by Russian emissions (Huang, 2010), who also suggested that contribution from South Asia was less than 10% of European contribution.

2.1.4 Ageing

Fresh BC released from emission sources gets mixed with other aerosols through condensation, coagulation and oxidation in the atmosphere. Afterwards, it mixes with water soluble compounds like sulfates and organic acids, which change its property from hydrophobic to hydrophilic. Under suitable conditions, the hydrophilic BC serves as a cloud condensation nuclei (CCN). (Oshima et al., 2009).

When BC is coated with non-absorbing particle the absorption efficiency increases (Bond et al, 2006). This depends on the BC core, the diameter of the shell and the coating materials. (Oshima et al., 2009). Schulz et al. (2006) had compared the lifetime of BC with different models and found out the lifetime ranges from 4.9 to 11.4 days with an average of 7.3 days. (Lund et al, 2008). The coating of BC increases their wet scavenging efficiency (Oshima et
al., 2009). Ageing of soot particles causes uncertainties for the estimation of both direct and indirect effects of BC in climate and air quality (Reimer et al., 2004).

2.1.5 Deposition mechanism on snow/ice surface

The ultimate ways of deposition of particles are wet and dry deposition.

2.1.5.1. Dry deposition mechanism

Dry deposition is the removal mechanism of particles from the atmosphere onto surface in the absence of precipitation. It depends on the particle size, density, terrain, vegetation features, etc. Dry deposition is commonly affected by atmospheric turbulence.

Particle deposition is caused by impaction, interception, diffusion, and gravitational sedimentation which are described as follows:

a. Gravitational sedimentation: Deposition velocity is about-equal to the particle settling velocity. It occurs for large particles whose diameter are greater than >3-4 μm.
b. Impaction: When the aerosol particles are not able to follow the curved streamlines then the larger particles tend to leave the streamlines and fall on the surface. These types of particles are relatively large, about few hundred nanometers.

c. Interception: When the particle passes near the surface it follows the stream lines, but can touch the surface leading to interception of particles.

d. Brownian diffusion: When the particles hit the surface due to the random thermal motion then Brownian diffusion happens. It occurs for the smallest aerosol particles which are <50-100 nm.

2.1.5.2. Wet deposition
Wet deposition is the most efficient aerosol removal mechanism with two commonly observed mechanisms. The removal process is either by in-cloud scavenging (rainout) or below cloud scavenging (washout). In-cloud scavenging occurs during the activation of particles to form a liquid drop due to nucleation, which falls eventually to the surface. Washout occurs when precipitation particles coagulate with aerosol particles on their way of falling. (Huang et al., 2010). Particles are also removed from the atmosphere by snow and fog.

Wet deposition is an episodic event, whereas dry deposition takes longer time. (Huang et al., 2010). Within a cloud, removal by rainout is >50% and, depending on the vertical profile of pollution, washout can be as low counts <0.1% removal of aerosol mass (Huang et al., 2010; Jacobson, 2003).

2.2 The Arctic areas and its characteristics
The Arctic area is characterized as the northernmost area of the world, covering approximately 14.5 million square kilometers with the latitude above 66°33”44”. The Arctic area encloses countries of United States (Alaska), Canada, Greenland, Iceland, Norway, Sweden, Finland and Russia.
Solar radiation at North Pole fluctuates with the season, receiving the highest level of radiation in summer and lowest in winter. The large amount of received solar radiation is reflected back into the atmosphere by extensive cloud, snow and ice cover.

A climatic condition in Arctic is divided into maritime and subcontinental subtypes. Maritime climate is found in Iceland, Norwegian coast, adjoining parts of Russia and along the Alaskan coast. While continental climate in the Arctic is from Northern Scandinavia towards Siberia and also in Eastern Alaska towards Canadian Arctic. Total annual precipitation is less than 500 mm in which Arctic coast experiences higher and Central Polar Basin is lower. Two dominant air current flows into the Arctic where cold air is flowing in winter from the high pressure zone over northern Siberia to the Pacific and air flowing
northwest from high pressure area over the Canadian Arctic towards the low pressure over the Atlantic (AMAP, 2015).

The current scenario of arctic-temperature rise is vigorous with decline of ice thickness and a permafrost area is thawing in Northern Alaska. Lenton (2012) has suggested that Arctic warming is due to BC together with a reduced amount of reflective sulphate aerosols.

### 2.3 Black carbon deposition on snow and ice surface

Single scattering albedo (SSA) of snow at visible wavelength is the highest approaching 1 in clean areas, but the light absorbing particles like BC can significantly reduce snow albedo. Even a small reduction in snow albedo may have notable forcing because of the resulting warming effects the snow morphology, melting rate, thus leading to amplification of radiative forcing (Bond et al., 2013). Chemical analysis of BC during the spring period of surface snow in North America, Russian Arctic and Greenland suggested that BC there is mainly associated with biomass burning (Bond et al., 2013).

From the initial change of snow albedo the larger feedback is not easy to evaluate. But for a longer period of time, lower snow albedo affects the snow ageing process (Hansen and Nazarenko, 2004; Hansen et al., 2005; Wang et al., 2011).

#### 2.3.1 Climate forcing by black carbon

BC influences the Arctic climate through several mechanisms. The foremost effect is to warm the Arctic atmosphere by absorbing solar radiation. Deposition of BC on snow and ice surface substantially reduces albedo, which enhances absorption and early melting of snow and ice. Furthermore, cloud distribution, lifetime and microphysical properties are influenced by aerosols through indirect and semi-direct effects which alter both shortwave and longwave energy budgets. (Quinn et al., 2011).

#### 2.3.2 Direct forcing

BC strongly absorbs incoming solar radiation, which leads to significant warming in the atmosphere. So, IPCC has estimated the direct radiative forcing of BC is in the magnitude ranging from +0.34 W/m² to 0.9 W/m² (Huang et al., 2010). Forcing by biomass burning
was estimated by Flanner et al. (2007) to vary from 0.49 in 2001. But it is important to note that there is a strong interannual variability in the Arctic associated with biomass burning.

### 2.3.3 Indirect forcing

About 60% of global surface is covered by clouds which affect the global atmospheric energy budget (Huang et al., 2010). Especially, the water soluble compounds tend to involve in cloud process and act as CCN. BC can strongly reduce low level cloud cover by heating the air and facilitating the evaporation of cloud droplets depending on the location of absorbing aerosols with respect to the cloud (Huang, 2010; Hansen et al., 1997).

In a pristine environment of the Arctic, where aerosol number concentration is low, the chances of particles to be activated as cloud droplets is higher (Komppula et al., 2005). Low level clouds in the Arctic are sensitive to aerosol particle concentration. An Arctic haze phenomena can increase the cloud long wave emissivity resulting in estimated surface warming between 1 and 1.6°C (Garret and Zhao, 2006; Huang et al., 2010). The average radiative forcing from BC by altering the surface albedo was estimated at +0.1 W/m² on a global scale and +0.3 W/m² on the Northern Hemisphere. (Hansel and Nazarenko, 2004; Solomon et al., 2007; Huang et al., 2010)

### 2.3.4 Snow ice forcing

Already a limited amount of BC concentration when deposited into snow and ice surface has high capability to reduce surface albedo by several orders of magnitude (Quinn et al., 2011). Furthermore, changes in snow albedo can exert a notable influence on climate by altering the timing of snow and ice melting and triggering snow/ice feedback (Quinn et al., 2011; Hansen and Nazarenko, 2004; Flanner et al., 2007). Global models estimate that about 0.03 W/m² originate from BC forcing of snow. (Jacobson 2004; Quinn et al., 2011; Koch et al., 2009b).
3. Methodology

The methods used throughout the research are presented in this chapter. It covers SILAM-CTM model setup for the model runs, input emission inventories. The chapter also provides insight to methods and tools that are used in the analysis process, and facilitates interpretation of processes and results, contributing to a better understanding of the results.

3.1 Data collection

Observational data for model measurement comparison in Europe were taken from European monitoring and evaluation programme (EMEP) database (Ebas, 2016) website for the year 2010. Observational data for Alert station is from Canadian aerosol baseline measurement program (Sharma et al., 2013; Stone et al., 2014). Mainly, filter absorption photometer and high and low volume sampler is used for the measurement of eBC in stations (figure 5).

Figure 5. Location of observational stations in the Arctic areas with sea ice extent map of March 2010
Location of all observational stations is pinned in map of sea ice extend of March, 2010 in Figure 5. (Fetterer et al., 2009). From year 2010, March was the month with maximum amount of sea ice extent in the Northern Hemisphere. Further details about the stations are provided in Table 2 and 3.

3.2 SILAM model

The modeling tool used in this study is the System for integrated modeling of Atmospheric composition. SILAM is a universal framework built for a wide range of meso-to-global scale dispersion tasks, such as inclusive of emergency response, air quality assessment, scenario analysis, data assimilation and analysis etc. Its transport core includes both Eularian and Lagrangian advection-diffusion formulations (Sofiev et al., 2011).

Dry deposition depends on the particle size and mechanism of dry deposition varies from the primarily turbulent diffusion driven removal of fine aerosols to the primarily gravitational setting of coarse particles. The wet deposition parameterization in SILAM is based on the observations performed for moderately hydrophobic aerosols with no growth with humidity. It distinguishes between sub and in-cloud scavenging by snow and ice (Sofiev et al., 2011).

The model requires gridded basic meteorological data and appropriate information on emission, as well as the physiographical data. Vertical resolution allows free selection of output vertical layers in several vertical types.

For the model run, all the meteorological information and necessary geophysical and land cover maps are taken from ERA interim. Fire emission was obtained from IS4FIRES system of FMI.

The outline of the model features is given in Figure 6.
Anthropogenic emissions are obtained from different sources depending on the spatial resolution. The horizontal model grid was configured with resolution of 0.72 degree of latitude and longitude. Point source information are added where available for all emission fields. Transformation scheme is acid basic which sets chemical and physical processes undergoing during the computation. Optical coefficients depend on relative humidity and temperature (Silam, 2016). The output from SILAM model run is further analysed using gridded analysis and display system (GRADS).
3.3 Emission inventories

In the current set of runs, all emissions are treated as primary emissions (Huang et al., 2010). The simulations are performed with two emission inventories. Comparison between observations and model simulations using multiple inventories is an appropriate method that could potentially implied to identify errors in inventory data (Granier et al., 2011). This research is focused mainly on publicly available inventories of anthropogenic and biomass burning emissions for the year 2010. Year 2010 is selected because of comparatively good availability of both emission and observational data. In this research, SILAM model simulation is performed with MACCity and ECLIPSE-GAINS4a emission inventories for the year 2010.

3.3.1 MACCity

MACC/CityZEN EU projects is a global emission dataset covering the period of year 1960 to 2020. The anthropogenic emissions are derived from atmospheric chemistry and climate model intercomparison project (ACCMIP) and representative concentration pathways (RCP) 8.5 datasets. In MACCity, biomass burning emissions comprise all emissions resulting from natural and man-made vegetation fires, including emissions from combustion of soil organic matter. Emissions from combustion of agricultural wastes or biofuels are excluded. The dataset has monthly temporal and 0.5×0.5 degrees of spatial step (Eccad, 2016).

3.3.2 EDGAR-HTAP 4.2

The EDGAR inventory is distributed on 0.1×0.1 degree grid over the globe. Emission is calculated for each compound as product of country specific data, the mix of technologies, abatement percentage and country specific emission factors. Point sources are also considered using maps with the locations of individual plants and industrial activity facilities (Granier et al., 2012). EDGAR database is for global atmospheric research which has data from 1970 to 2008. It provides global past and present day anthropogenic emissions of greenhouse gases and air pollutants on spatial grid.
### 3.3.3 ECLIPSE_GAINS_4a

ECLIPSE_GAINS_4a is a global emission dataset developed with the GAINS model for the period 2005 to 2050. The dataset provides gridded global emissions at 0.5° spatial resolution from all anthropogenic sources excluding international shipping and aviation. ECLIPSE inventory was created using GAINS which provides emissions of long-lived species in a consistent framework. Product has temporal resolution and spatial reference of 0.5×0.5 degrees (Eckhardt et al., 2015; Eccad, 2016).

During this research, shipping emission is added from AU-RCP6.0 and FMI. The combined ship emissions are for North of 60°N and RCP 6.0 emissions for rest of the world (Tntcat, 2016).

All above inventories are lumped into various sectors and their information is described detail in Table 2 below. The emission inventories for BC have two parts – for anthropogenic emissions and biomass burning. For SILAM model fires data were picked from the IS4FIRES dataset. Emissions from oil and gas flaring are present only in ECLIPSE inventory.

Table 2. Sectors with their availability of BC and OC data for MACCity, ECLIPSE and EDGAR-HTAP emission inventories

<table>
<thead>
<tr>
<th>Category</th>
<th>Sectors</th>
<th>MACCity</th>
<th>ECLIPSE</th>
<th>EDGAR</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>BC</td>
<td>OC</td>
<td>BC</td>
</tr>
<tr>
<td>Anthropogenic</td>
<td>Aviation</td>
<td>●</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Transportation</td>
<td>● ●</td>
<td>● ●</td>
<td>●</td>
</tr>
<tr>
<td></td>
<td>Energy</td>
<td>● ●</td>
<td>● ●</td>
<td>●</td>
</tr>
<tr>
<td></td>
<td>Solvents</td>
<td>● ●</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Waste</td>
<td>● ●</td>
<td>● ●</td>
<td>●</td>
</tr>
<tr>
<td></td>
<td>Industries</td>
<td>● ●</td>
<td>● ●</td>
<td>●</td>
</tr>
<tr>
<td></td>
<td>Residential</td>
<td>● ●</td>
<td>● ●</td>
<td>●</td>
</tr>
<tr>
<td></td>
<td>Agricultural waste</td>
<td>● ●</td>
<td>● ●</td>
<td>●</td>
</tr>
</tbody>
</table>
These three emission inventories have developed their own emission estimates where most of them have common sectors. Most anthropogenic BC emissions have occurred in northern mid-latitudes with peak emissions around 35°N. The model calculates emission for hemispheric regions by all major economic sectors, including energy and industrial production, transport, residential, agricultural waste, agriculture, ships and burning including savanna and forest fires. In ECLIPSE, model considers nearly 2000 sector fuel technology combinations for which emissions are calculated (Stohl et al., 2013).

### 3.3.4 Model measurement comparison

The overall process was initiated by simulating MACCity and ECLIPSE emission inventories. Along with it, observational data are extracted from EMEP, IASOA and other different sources. Simulated result output from both inventories are intercompared with observational result with the help of Python. The procedure of model measurement comparison process is described further in Figure 7.
The model measurement comparison is performed by time and space integrated paired data and statistical result of BC, EC and OC for all stations are analysed and interpreted. Statistical analysis includes root mean square error (RMSE), correlation coefficient (CORR) and BIAS.

3.3.5 Emission, deposition and concentration comparison
Source of emission differ with the location of sectors. Thus, sectoral emissions along with the summation of source are intercompared between MACCity, EDGAR-HTAP and ECLIPSE inventories on 2\textsuperscript{nd} of January 2010.
Emission, seasonal concentration and deposition comparison

Figure 8. Schematic diagram for emission, seasonal concentration and deposition comparison

Result output of simulated inventories are evaluated using GRADS. Also, simulated deposition and concentration result output are evaluated for MACCity and ECLIPSE driven runs (figure 8). Result output is intercompared for summer and winter period. These seasonal values for both inventories gives an estimation of model performance in the Arctic. Yearly values are calculated in mg/m²/year, whereas daily values were evaluated in kg/day.
4. Results

In this section, the results are presented for two seasons: winter (November to April) and summer (May to October).

4.1.1 BC emission comparison

Determination of emission source sectors and geographical regions that causes an impact on the Arctic climate is the main assessment of this chapter. Recent research indicates that the emission from parts of Europe, East and Northern Asia and North America contribute to Arctic surface and mid-tropospheric BC concentration to varying degrees (Stohl et al, 2006; Shindell et al., 2008, AMAP 2011). The relative contribution of individual emission source to BC in the Arctic depends not only on emission strength, but also on the relative efficiency of transport from source to receptor regions. Transport phenomenon from Eurasia to the Arctic is more efficient during winter than during summer thus emission occurring in winter will have a heavier impact on Arctic BC than summer period (AMAP 2011). Black carbon emission sources changes rapidly as a result of greater energy consumption. (Bond et al., 2013)

Throughout the assessment, daily emission comparison of BC from MACCity, EDGAR-H TAP and ECLIPSE emission inventories were employed. The spatial distribution of emission fluxes around the Arctic and near Arctic areas. Figure 9 presents that countries contributing to BC emissions are mainly Russia, Canada, Northern European countries and Siberia. BC emission in South and East Asia is growing rapidly in these past decades, which is is also evident in Figure 9. Furthermore, it is much stronger there than in Europe, North America and Russia combined (Stohl et al., 2006). Pollution plumes from boreal forest fires originating from Siberian Arctic also influence the atmospheric composition on a hemispheric scale.
During summer 2004, severe forest fires burning in Alaska and Canada led to a strong increase of BC concentrations at four different stations located in the Arctic areas (Stohl et al., 2006). Agricultural burning and boreal forest fires in central and Western Eurasia dominates the emission sources of aerosol concentrations in large part of the Arctic troposphere (Stohl et al., 2007; Hirdman et al., 2010). Conjointly, Barrow station in summer was influenced by emissions from anthropogenic sources likewise forest fires originating in the central and eastern parts of Russia.

Furthermore, from emissions of atmospheric compounds and compilation of ancillary data (ECCAD) site (Eccad, 2016) yearly values of emissions per year with their sectoral emissions are summarised in Table 3. Shipping emissions for ECLIPSE is later added from FMI, so it is excluded from the Table 3.
Table 3. Yearly emission value for ECLIPSE and MACCity inventories with sectoral differences

<table>
<thead>
<tr>
<th>Inventories</th>
<th>Values (mg/m²/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MACCity</td>
<td>7.74</td>
</tr>
<tr>
<td>ECLIPSE</td>
<td>6.62</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sectors for BC</th>
<th>MACCity result (mg/m²/year)</th>
<th>ECLIPSE result (mg/m²/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ships</td>
<td>0.23</td>
<td>0</td>
</tr>
<tr>
<td>Agricultural waste</td>
<td>0.22</td>
<td>0.51</td>
</tr>
<tr>
<td>Residential</td>
<td>2.9</td>
<td>3.47</td>
</tr>
<tr>
<td>Industries</td>
<td>2.53</td>
<td>0.73</td>
</tr>
<tr>
<td>Energy</td>
<td>0.07</td>
<td>0.98</td>
</tr>
<tr>
<td>Transportation</td>
<td>1.61</td>
<td>2.06</td>
</tr>
<tr>
<td>Waste</td>
<td>0.05</td>
<td>0.13</td>
</tr>
</tbody>
</table>

Both MACCity and ECLIPSE inventories have figured out energy, transportation and residential source as the major polluting sectors which are highlighted in Table 3. All the biomass burning sources like forest fires and savanna burning are included in MACCity whereas these are missing from other two emission inventories. Shipping, agricultural waste and agricultural emission are not included in ECLIPSE.

4.2 Analysis of model measurement comparison for European stations

Table 4 displays the description of station information and compound along with their statistical result from model measurement comparison for MACCity and ECLIPSE emission inventories. It provides a summary of statistical results of model performance for BC, EC and OC with observations at EMEP sites. Due to missing data from some station, only those stations with all information about EC, BC and OC are included in table 4. Correlation coefficient (CORR) shown in Table 4 for EC, BC and OC is moderate for almost all stations.
Table 4. Station information with their available compounds and their statistical result (BIAS, Correction coefficient and RMSE)

<table>
<thead>
<tr>
<th>Station code</th>
<th>Station code</th>
<th>latitude</th>
<th>longitude</th>
<th>Mean measured value (µg/m³)</th>
<th>BIAS (µg/m³)</th>
<th>CORR</th>
<th>RMSE (µg/m³)</th>
<th>BIAS (µg/m³)</th>
<th>CORR</th>
<th>RMSE (µg/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH0002R</td>
<td>Payerne</td>
<td>46.8</td>
<td>12.9</td>
<td>0.58</td>
<td>BC&lt;sub&gt;d&lt;/sub&gt; 0.13</td>
<td>0.36</td>
<td>0.48</td>
<td>0.23</td>
<td>0.41</td>
<td>0.57</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.25</td>
<td>EC           0.34</td>
<td>0.60</td>
<td>0.45</td>
<td>0.40</td>
<td>0.39</td>
<td>0.56</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.28</td>
<td>OC           -1.88 0.41</td>
<td>2.23</td>
<td>-1.81</td>
<td>0.52</td>
<td>2.23</td>
<td></td>
</tr>
<tr>
<td>CZ0003R</td>
<td>Kosetice</td>
<td>49.6</td>
<td>-1.3</td>
<td>0.49</td>
<td>EC           0.39</td>
<td>0.39</td>
<td>0.69</td>
<td>0.96</td>
<td>0.59</td>
<td>1.51</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.30</td>
<td>OC           -2.54 0.26</td>
<td>3.78</td>
<td>-1.83</td>
<td>0.67</td>
<td>2.82</td>
<td></td>
</tr>
<tr>
<td>DE0003R</td>
<td>Schauinsland</td>
<td>47.9</td>
<td>7.9</td>
<td>0.16</td>
<td>EC           0.86  0.44</td>
<td>0.97</td>
<td></td>
<td>0.87</td>
<td>0.20</td>
<td>1.01</td>
</tr>
<tr>
<td>DE0044R</td>
<td>Melpitz</td>
<td>51.5</td>
<td>12.9</td>
<td>1.60</td>
<td>EC           -0.65 0.47</td>
<td>2.47</td>
<td>-0.29</td>
<td>0.75</td>
<td>1.84</td>
<td></td>
</tr>
<tr>
<td>GB0036R</td>
<td></td>
<td>51.6</td>
<td>-1.3</td>
<td>0.52</td>
<td>BC&lt;sub&gt;h&lt;/sub&gt; 0.11</td>
<td>0.66</td>
<td>0.49</td>
<td>0.33</td>
<td>0.57</td>
<td>0.72</td>
</tr>
</tbody>
</table>
Values highlighted with green color are good statistical results whereas values highlighted with yellow color are inferior values. For almost all stations, OC was under-estimated by simulations with both inventories whereas for BC the comparison outcome varies for different sites. For the station Ispra, in Italy the model is underestimating OC and overestimating BC. Ispra station is located in a semi-rural area with which is >20 km far from pollution source. RMSE is lowest for Payerne, Kosetice and Ispra stations. For the station Harwell, all the results are comparatively better with RMSE, correction coefficient and BIAS. The correlation coefficient varies widely from 0.3 up to 0.75. The lower correlation found in several sites is probably by the virtue of the unaccounted effect of local emission sources (Tsyro et al, 2007).

Model measurement comparison of few stations for BC and OC are described further in chapter 4. Few more important results of model measurement comparison are attached in appendix A and B for BC, EC and OC.
4.2.1 Payerne station timeseries plot of BC

Figure 10 shows the model measurement comparison at Payerne station, hourly model time series plotted against the daily observation. As seen in Table 4, the temporal correlation coefficient is comparatively high reaching 0.6 for MACCity-driven run.

![Model measurement comparison of BC for the year 2010 from station (CH0002R, Payerne, Switzerland) with modeled result output from MACCity and ECLIPSE emission inventories](image)

Figure 10. Model measurement comparison of BC for the year 2010 from station (CH0002R, Payerne, Switzerland) with modeled result output from MACCity and ECLIPSE emission inventories

On average, the model overestimates observed BC concentration, more with ECLIPSE than with MACCity emission. The correlation between observation and model results is on average for the MACCity driven run than for ECLIPSE based simulations.

The station is located in rural background which is surrounded by agricultural land, forests and small villages. Seasonality is driven by emission variation during the year. Thus, emissions from mobile sources are distributed relatively equally over the year, while commercial combustion is lower during the warm period. The modeled result and observational result are in a general agreement on BC seasonal variation, with the highest concentration in winter period. MACCity emission variation is somewhat too low, possibly due to underestimated winter-time domestic heating emissions. (Tsyro et al., 2007)
4.2.2 Harwell station timeseries plot of BC

Figure 11 shows the model measurement comparison at Harwell station, hourly model time series plotted against hourly observation. As seen in Table 5, the temporal correlation coefficient is comparatively high reaching to 0.66 for the MACCity-driven run.

![Harwell station timeseries plot of BC](image)

Figure 11. Model measurement comparison of BC for year 2010 from station (GB0036R Harwell, Great Britain) with modeled output for MACCity and ECLIPSE emission inventories

On average, the model overestimates most of the time of observed BC concentration more with ECLIPSE than with MACCity emissions. Correlation between observation and model results is higher for the MACCity driven run than for the ECLIPSE based simulations.

The station is located in Southern England, is affected by clean maritime air masses and polluted air masses from local and more remote industrial activities. The model result and observational result are in general agreement on BC seasonal variation with higher concentrations in winter period. Some peaks can be observed also in the summer period.
4.2.3 Ispra station timeseries plot for OC

Figure 12 shows the model measurement comparison at Ispra station, hourly model time series plotted against the daily observation. As seen in Table 5, the temporal correlation coefficient is comparatively high reaching 0.5 for ECLIPSE driven run.

![Organic carbon at IT0004R, 2010, MACCity, ECLIPSE vs EMEP](image)

Figure 12. Model measurement comparison of OC for the year 2010 from the Station (IT0004R, Ispra, Italy) with modeled result output from MACCity and ECLIPSE emission inventories

Results from both inventories underestimate OC at all times. The correlation between observation and model results is higher for the ECLIPSE run than for the MACCity run. Ispra station is classified as a rural background site where the concentration of carbonaceous aerosol is comparable to those of urban European background sites, which is likely due to the impact of regional air pollution in Po valley. The main source of carbonaceous aerosols is due to central Europe continental pollution, local traffic and residential heating (Gilardoni et al., 2011).
4.3 Analysis of model measurement comparison for Arctic stations

Table 5 displays the statistics for Arctic stations. For the comparison, the equivalent black carbon (eBC) is computed from the modeled BC results.

eBC at Alert station is measured using Aethalometer. The obtained data is light absorption derived with a mass absorption cross-section of 16.6 m²/g at standard temperature and pressure (STP). Data for Zeppelin and Vavihill station is calculated using multi-angle absorption photometry equation (Petzold et al., 2003). Station information with their available compounds and their statistical result (BIAS, Correction coefficient and RMSE)

Table 5. Station information from Arctic areas with BC and the their statistical result (BIAS, correlation coefficient and RMSE)

<table>
<thead>
<tr>
<th>Stations</th>
<th>Latitude</th>
<th>Longitude</th>
<th>Mean measured value (µg/m³)</th>
<th>BIAS (µg/m³)</th>
<th>RMSE (µg/m³)</th>
<th>CORR</th>
<th>BIAS (µg/m³)</th>
<th>RMSE (µg/m³)</th>
<th>CORR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alert</td>
<td>82.5</td>
<td>-62.5</td>
<td>0.027</td>
<td>-0.020</td>
<td>0.035</td>
<td>0.60</td>
<td>-0.022</td>
<td>0.04</td>
<td>0.61</td>
</tr>
<tr>
<td>Tiksi</td>
<td>71.6</td>
<td>128.8</td>
<td>0.06</td>
<td>-0.046</td>
<td>0.77</td>
<td>0.23</td>
<td>-0.047</td>
<td>0.78</td>
<td>0.29</td>
</tr>
<tr>
<td>Vavihill</td>
<td>56.0</td>
<td>13.1</td>
<td>0.71</td>
<td>-0.156</td>
<td>0.67</td>
<td>0.55</td>
<td>0.106</td>
<td>0.697</td>
<td>0.55</td>
</tr>
<tr>
<td>Zeppelin</td>
<td>78.9</td>
<td>11.8</td>
<td>0.013</td>
<td>-0.023</td>
<td>0.058</td>
<td>0.46</td>
<td>-0.027</td>
<td>0.062</td>
<td>0.39</td>
</tr>
</tbody>
</table>

Values highlighted with green color are good statistical results. The eBC is underestimated at all stations with both emission inventories. It suggests that various local sources in the Arctic and near Arctic areas are missing in both inventories. Table 4 shows a good correlation coefficient is found in Alert and Vavihill station with 0.6 and 0.55 for both inventories.
4.3.1 Alert station

Figure 13 shows the model measurement comparison of Alert station, hourly model time series against observation. As seen in Table 4, temporal correlation is comparatively high, reaching 0.6 for both inventories but the model underestimates BC concentration.

Figure 13. Model measurement comparison of BC for the year 2010 from Station Alert with modeled output with MACCity and ECLIPSE emission inventories

The modeled and observational results are in a general agreement on BC seasonal variation, with higher concentration during winter. The Arctic station is located on the furthest north of all the Arctic stations on the north-eastern tip of Ellesmere Island. For about 10 month land and ocean is covered with ice or snow. (Hirdman et al., 2010). The seasonal cycle of BC is characterized by higher concentration values in winter than in summer. The strong decrease begins at the end of May.

4.3.2 Zeppelin station

Figure 14 shows the model measurement comparison at Zeppelin station hourly time series plotted against observational data. Due to unavailability of observational data from April to July intercomparison during summer period is unapplicable.
As seen in Table 4, the temporal correlation coefficient is on average about 0.46 for MACCity driven run, whereas it is unexpectedly lower in ECLIPSE driven run of about 0.39. Differences in correlation between the two inventories might be due to the absence of local sources in ECLIPSE which has been clearly explained in MACCity.

Zeppelin is situated on a mountain ridge on the west coast of Spitsbergen. Air masses are either arriving from the ice-free North Atlantic Ocean or from the general ice covered Arctic Ocean (Hirdman et al., 2010). As expected, the seasonal variation seems evident between summer and winter period. From this station, concentration is unevenly higher during spring season. The Zeppelin station is not strongly influenced by local emissions; however summer values are enhanced by 11% due to local ship emissions (Eckhardt et al., 2013; Stohl et al., 2013)
4.3.3 Tiksi station

Figure 15 shows the model measurement comparison at Tiksi station, hourly model time series plotted against the observation. As seen in Table 3, the temporal correlation coefficient is very low for both inventories it is about 0.25.

![Graph of Black carbon at Tiksi, 2010, obs vs MACCity, ECLIPSE](image)

Figure 15. Model measurement comparison of BC for the year 2010 at the Tiksi station with modeled result output from MACCity and ECLIPSe emission inventories

During the end of January and mid of June the peaks are quite high, about 2 times higher than other time period. The reason behind it could be that it is close to the local sources of pollution. As a result, the annual mean is 2.5 times higher than the other stations. (Eckhardt et al., 2015)

Except for these two outliers in January, seasonality is not distinct for this station. The MACCity and ECLIPSE performance for this run is quiet inferior. The probable reason might be missing local sources of pollution. In this context, the model underestimates observed BC concentration for both simulations.
4.3.3.1. Vavihill station

Figure 16 shows the model measurement comparison at Vavihill station, hourly model time series plotted against the daily observation. As seen in Table 3, temporal correlation coefficient is comparatively high reaching 0.55 for both runs.

Figure 16. Model measurement comparison of BC for the year 2010 from station Vavihill, Sweden with modeled result output from MACCity and ECLIPSE emission inventories.

Due to a lack of results from July to December, the comparison is limited with the first half of the year. On average, MACCity driven run is likely to underestimate the result, whereas ECLIPSE has slightly overestimated during winter and spring period.

Vavihill station is a continental background site with no local source of pollution. It is situated in the southernmost part of Sweden. Peaks during the spring and summer period might be caused by the transport from remote industrial areas.

4.4 Comparison of concentration and deposition maps

This section explores the differences and similarities between the modeled concentration and deposition maps. Deposition and concentration results are characterized for two seasons: summer (May-October) and winter (November-April). The effect of dry deposition is not
well visible on seasonal patterns of BC in the Arctic lower troposphere, but results in significant changes in seasonal concentration (Huang et al., 2010). As shown in previous sections, SILAM is able to simulate the observed seasonality of BC over the Arctic and Northern hemisphere. (Heinola et al., 2013; Jurado et al., 2008).

Local source of BC is currently small and limited in Arctic areas. Some anthropogenic emissions contribute to it from the northern parts of Russia, and emissions from oil and shipping. Huang (2010) study suggests that in most of the Arctic stations, i.e. Zeppelin, Svalbard, Barrow and Alaska most of the aged particles are transferred to Arctic in winter period. On the other side, backward trajectories suggested that during the winter and spring, industrial regions in Eurasia and North America are the major source of Arctic aerosols. Suggested results from atmospheric chemistry and climate model intercomparison project (ACCMIP) models against observations it was found major contributor for BC deposition in Greenland is North America whereas else in Arctic and near Arctic areas European emission counts to the greatest extent (Lee et al., 2013b; Jiao et al., 2014). But these studies were insufficient to explore the seasonality of Arctic and near Arctic areas. Hence, this study focuses on atmospheric removal processes, such as dry and wet deposition to better understand the observed seasonal cycle. (Huang et al., 2010)

The Arctic region (North of 70°N) is very dry during winter with daily average precipitation rate between 0 and 1 mm/day. During summer time it experiences more than 2-3 times higher precipitation than winter. (Huang et al., 2010).

4.4.1 Seasonal deposition comparison
The simulations driven by two emission inventories with their dry and wet deposition results are analyzed for two seasons.

4.4.1.1. BC deposition from November to April
Comparison of the SILAM results with MACCity and ECLIPSE for BC emission inventories for winter period are shown in figure 17. For all particle size ranges, the dry deposition in the Arctic is higher in winter months due to high surface wind speed and more intense vertical
mixing (Huang et al., 2010). But the wet deposition losses during summer are greater because of more precipitation in the Arctic than in the winter.

![Figure 17. BC deposition from November to April](a) Dry deposition of SILAM-ECLIPSE, (b) Dry deposition of SILAM-MACCity, (c) Wet deposition of SILAM-ECLIPSE and (d) Wet deposition of SILAM-MACCity
National institute of environmental studies (NIES) model estimates the deposition of BC in Arctic is about 0.065 Tg/year (Sharma et al., 2013). During summer period transport of BC from Asia are experienced with wet scavenging so transport mechanism is not so competent to reach Arctic areas. Shipping in Arctic and sub-Arctic areas can be clearly viewed in figure 17. It seems that shipping in North Sea route and North West passage emission of BC can be viewed clearly in Figure 18. According to Browse (2013) Arctic shipping contributes 0.3% of BC deposited on North.

### 4.4.1.2. BC deposition from May to October

Comparison of the SILAM result output with MACCity and ECLIPSE for BC emission inventories for summer period are shown in figure 18. Removal mechanism is more effective during summer period than in the winter period, receiving more frequent rainfall. However, it is also patchier during summer, as seen from the results for both emission inventories.
Figure 18. BC deposition from May to October (a) Dry deposition of SILAM-ECLIPSE, (b) Dry deposition of SILAM-MACCity, (c) Wet deposition of SILAM-ECLIPSE and (d) Wet deposition of SILAM-MACCity

Transport mechanism is not effective during summer period than during winter period. Summer season has monsoon in Asia so the wet deposition mechanism is efficient and faster than in winter period. As a result, more frequent rainfall in summer leads to faster particle removal by wet deposition and shorter particle transport distance.

### 4.4.1.3. OC deposition from November to April

Deposition of OC during the winter (November-April) is shown in Figures 19. The emission of OC is higher than BC, as can be seen from the comparison with the previous section. Part of OC is released as the aerosol precursors, which undergo processing in the atmosphere to form secondary organic aerosols. The main sources of organic carbon are biomass burning and fossil fuel combustion and also biogenic secondary organic aerosols (SOA).
Figure 19. OC deposition from November to April (a) Dry deposition of SILAM-ECLIPSe, (b) Dry deposition of SILAM-MACCity, (c) Wet deposition of SILAM-ECLIPSE and (d) Wet deposition of SILAM-ECLIPSE
SILAM-ECLIPSE seems to have higher levels in parts of China than the other run. In another case, SILAM-MACCity has deposition wet deposition in Siberian Arctic than SILAM-ECLIPSE.

Wet deposition is more efficient over the sea and oceans. Deposition is high in China which is the dominant source in the Northern Hemisphere. Runs with both emission inventories suggest substantial contribution from the hemispheric sources into the wet deposition over the Arctic areas.

4.4.1.4. OC deposition from May to October
Deposition of OC during the summer (May-October) is shown in Figures 20. During summer season dry deposition is ineffective than during winter. But due to removal mainly by rain wet removal is dominant over wet season than during dry season. These patterns can be observed from Figure 20.
Figure 20. OC deposition from May to October (a) Dry deposition of SILAM-ECLIPSE, (b) Dry deposition of SILAM-MACCity, (c) Wet deposition of SILAM-ECLIPSE and (d) Wet deposition of SILAM-MACCity

Dry deposition of particles near the Arctic areas seems to be noticeable. Particle seems to scavenged by wet deposition which are transported over seas and oceans. Both deposition in Asia is quite evident in both emission inventories. Some marking has been spotted in Europe and in Canadian Arctic areas. Also, Russian open fires seems to be dominant source of organic carbon in Arctic areas (Wang et al., 2011). Wet deposition by MACCity inventory shows spots around Russian areas and in Bering Sea.

4.4.2 Concentration comparison

Figure 21 shows modeled atmospheric concentration of BC for both seasons. Energy related combustion is higher where population density is the highest which can be observed by comparing European areas and Asian parts like India and China. Large BC concentration also occurs in and around biomass burning regions, especially in South America and Central Africa (Bond et al., 2013).
Figure 21. Concentration comparison of BC from May to October and November to April from (a) May-October SILAM-ECLIPSE, (b) May-October SILAM-MACCity, (c) November-April SILAM-ECLIPSE and (d) November-April SILAM-MACCity

There are only a few sources in Arctic so the existing BC is due to the transport from mid-latitudes. Summer season has lower emission and a higher deposition, which leads to lower
concentrations in figure 21. Concentration of BC found in Siberian Arctic during summer period is mainly due to wildfires. Winter period concentration seems to be evident where population density is higher and industrialized areas.

4.5 Carbon budget calculation

The carbon budget of the domain (Table 6) confirms that wet removal is the dominant mechanism in both seasons. The difference in the description of emission sectors in the database could explain difference in results.

Table 6. Comparison of deposition and concentration with seasonal variation from ECLIPSE and MACCity inventories

<table>
<thead>
<tr>
<th>Deposition and concentration results of BC with seasons</th>
<th>MACCity result</th>
<th>ECLIPSE result</th>
</tr>
</thead>
<tbody>
<tr>
<td>November to April</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry deposition</td>
<td>91.32 Kt/(Nov to Apr)</td>
<td>199 Kt/(Nov to Apr)</td>
</tr>
<tr>
<td>Wet deposition</td>
<td>1707.13 Kt /(Nov to Apr)</td>
<td>2110 Kt/(Nov to Apr)</td>
</tr>
<tr>
<td>May to October</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry deposition</td>
<td>65.91 Kt/(May to Oct)</td>
<td>39.5 Kt/(May to Oct)</td>
</tr>
<tr>
<td>Wet deposition</td>
<td>2136.5 Kt/ (May to Oct)</td>
<td>685 Kt/(May to Oct)</td>
</tr>
<tr>
<td>Concentration from November to April</td>
<td>0.23 μg/m³</td>
<td>0.26 μg/m³</td>
</tr>
<tr>
<td>Concentration from May to October</td>
<td>0.11 μg/m³</td>
<td>0.08 μg/m³</td>
</tr>
</tbody>
</table>

Deposition estimate from MACCity for winter period is 5.07% of dry deposition and 94.92% of wet deposition of BC. For ECLIPSE winter period is about 8.61% and 91.38% for dry and wet deposition. Concentration comparison shows that winter period concentration is higher for ECLIPSE driven run than MACCity and vice versa for summer period. Concentration due to stagnant condition and less precipitation during winter period leads to about double the concentration than during summer period. Also, residential combustion and domestic heating accounts for major emission during that period.
5. Concluding remarks

The aim of the research was to improve the knowledge on concentrations and depositions of the carbonaceous compounds, mainly BC, in Northern Europe and Arctic. Three emission inventories were compared, simulations with two of those – ECLIPSE and MACCity – were evaluated in details against the existing European and Arctic observations. A chemistry transport model SILAM was used to examine the seasonal variation of black carbon in the year 2010 over the Northern Hemisphere. Simulations included two emission inventories in order to understand the impact of the variability of emission inventories on the BC transport to the Arctic. The simulations were compared with daily deposition and concentration at European and Arctic observational stations.

Modeling of BC in the Arctic has improved compared to earlier studies. (Koch et al., 2009, Sharma et al., 2013; Eckhardt et al., 2015, AMAP,2011). Observations result of surface eBC at European stations shows some overestimation for the whole period with a correlation coefficient of 0.62 for the hourly observation. OC concentrations were usually underestimated in most stations for both inventories. The correlation coefficient of EC concentration is higher than other compounds. The overall performance for BC for European stations is substantially better than in the Arctic areas. Seasonal deposition and concentration of BC and OC is simulated for both inventories and seasonality has been analysed with higher concentration during winter than during summer period. We found a strong correlation between deposition and concentration. The overall performance of the runs driven by the MACCity inventory is somewhat higher than that of the run with the ECLIPSE inventory.

The modeling results indicate considerable uncertainties around BC emission. There is a possible underestimation of BC emissions from traffic in some areas and both underestimation and overestimation of BC emissions from residential combustion for some countries. The largest uncertainties lie in residential wood combustion in both emission inventories. Some, minor burning of backyard waste might produce huge local impact and somewhat contribute to the transport towards the Arctic areas.
Arctic areas are also sensitive to emissions in high-latitude Eurasia. The impact of wildfire emission and some local sources has been underestimated which could be seen from the Tiksi station. The contribution of fires to BC deposition ranges from 39% in May and 56% in July (Generoso et al., 2007). More relevant studies of wildfires in Russia and Siberia could give an estimate of uncertainty in the model run. Also, many backward trajectories has been suggested the Arctic BC during spring and winter seasons from industries are mainly transported from Eurasia and North America (Huang et al., 2010).

OC seasonality is still too weak in most results: the winter period, November- April, is underestimated. Changes in meteorology alone could not explain the observed trends, but were reflected in inter annual variations. During winter period transport from Canadian and US Arctic to pole areas is more evident in ECLIPSE than for MACCity inventory. During the summer period, monsoon in Southern Asia enforces the removal mechanism, which is distinct in the results. Due to higher emissions, the deposition of OC is higher for both dry and wet deposition. From deposition and concentration result it is found out that Arctic climate has significantly larger sensitivity of BC emitted within Arctic and near Arctic areas compared to BC emitted in mid-latitudes. So, even a small increment of BC in Arctic areas near would bring a global impact. Finally, this study provides the best model setup of SILAM-CTM for transport and deposition of carbonaceous aerosols especially BC analysis.
6. Discussion and future work

Black carbon is of great interest and its study reveals that, due to its absorption efficiency, an impact via snow/ice cycle has great potential to influence climate change and warm the atmosphere. The model measurement comparison showed that BC concentration is higher in most of the European stations than in the Arctic areas with a higher concentration being about 3 µg/m³ during the winter period.

Deposition for BC and OC shows that the seasonal transport of BC from source regions is evident in the Arctic and near Arctic areas. Pattern of dry deposition is higher during the winter period than during summer period.

During the study, an aged BC was assumed from the beginning, thus potentially increasing somewhat its removal intensity. Incorporation of this process has been completed but evaluation is yet to be made. Furthermore, deposition result were evaluated without the observational dataset from stations in the Arctic. Accordingly, intercomparing the result of model output obtained from both emission inventories with observational data might give an insight of extensive differences in the result. The research is limited to depositing BC, we were not able to calculate the impact of snow and ice cover (albedo change).

Our major challenge concerning BC was the emission inventories missing local sources, which in remote areas can substantially affect the observational sites. Also, the observations of BC deposition (both dry and wet) were missing, which complicated the evaluation of this important quantity. More observation data from the Arctic station would provide a good insight of evaluation of model measurement comparison of BC in Arctic snow and ice. For SILAM, including ageing would perform better for BC in Arctic areas.
Bibliography


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Appendix A. Time series of EC for two stations

Figure A1: Model measurement comparison of EC for the year 2010 from Station (DE0044R, Melpitz, Germany) with modeled result output of OC from MACCity and ECLIPSE emission inventories.

Figure A2: Model measurement comparison of EC for the year 2010 from Station (IT0004R, Ispra, Italy) with modeled result output of OC from MACCity and ECLIPSE emission inventories.
Appendix B: Time series of OC for three stations

Figure B1: Model measurement comparison of OC for the year 2010 from Station (CZ0003R, Kosetice, Czech Republic) with modeled result output of OC from MACCity and ECLIPSE emission inventories.

Figure B2: Model measurement comparison of OC for the year 2010 from Station (CH0002R, Payerne, Switzerland) with modeled result output of OC from MACCity and ECLIPSE emission inventories.
Figure B3: Model measurement comparison of OC for the year 2010 from Station (GB0036R, Harewell, Great Britain) with modeled result output of OC from MACCity and ECLIPSE emission inventories.

Figure B4: Model measurement comparison of OC for the year 2010 from Station (IT0004R, Ispra Italy) with modeled result output of OC from MACCity and ECLIPSE emission inventories.