CONSTRAINING A HISTORICAL BLACK CARBON EMISSION INVENTORY OF THE UNITED STATES FROM 1960-2000

BY

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THESIS
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ABSTRACT

In order to improve the accuracy of the black carbon (BC) historical emission inventory of the United States over the period 1960-2000, I studied the transport of global BC emissions and ambient air concentrations using the Community Atmosphere Model. I formulated the relationship between emissions and concentrations into matrices that allow reconstruction of emission for a time-varying ambient air concentration. When calculating the transport matrices from the model simulation result, I adjust the model meteorology according to the measured meteorology. This could avoid attributing model error to emission inaccuracy. With the transport matrices, I reconstructed the BC concentration in California and New Jersey where coefficient of haze data are available. The discrepancy between the modeled BC and the measurement identifies needed improvements to the emission inventory. However, BC concentrations simulated with different resolutions could vary by a factor of two. Therefore, I suggest that discrepancies in trends are more instructive than absolute discrepancies, especially for evaluating global model results. In the comparison, I examined magnitudes and seasonality of emissions. Emissions dominating BC concentrations in California and New Jersey have evolving seasonal pattern over 1960-2000. Heating Degree Days (HDDs) data are applied to estimate seasonal residential emissions. However, even the adjustment for HDDs does not fully explain the trend in seasonal variation of the measurement. Also, the mismatch of modeled BC and measurement in New Jersey reveals the inaccuracy of model meteorology in New Jersey. The magnitude of reconstructed emissions was decreasing throughout this period of time, while the original emission inventories peaked in the 1980s. This may indicate the shift of energy consumption between different technologies (shift to cleaner technology) which can cancel out the influence of energy consumption increase.
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I want to thank everyone that has helped me during my master study. It was an important transitional period of my life. This time for me likes take-off for a flight. Left the ground that I have been get used to, learned to fly myself to my destination and dealt with all the unpredictable turbulences. Flight is fragile during take-off. But it is the frangibility taking it to the sky.

Thank my parents, they are my best runway. They guided me to the direction to take-off and helped me well prepared for it. It is good to flying in the sky, but sometime I missed the ground and my runway a lot.

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CHAPTER 1
INTRODUCTION AND BACKGROUND

Human activities affect the energy balance of the Earth system by changing the chemical composition of the atmosphere. Greenhouse gases, such as CO$_2$, are known for their contribution to global warming. Aerosols, gaseous suspensions of fine solid or liquid particles, have also played an important role in altering the radiation balance, and this effect is critical in explaining the historical pattern of temperature change (Santer et al., 1995). The climate effect of aerosol is mainly dependent on its concentration in the atmosphere, optical properties, and interaction with clouds. Overall, aerosols have a net cooling effect on the earth system, since there are reflective aerosols scattering light back to space. However, some of them can absorb light, heat the atmosphere and oppose the cooling effect of the scattering aerosols. The effects of those warming aerosols were recognized (Charlson & Pilat, 1969) and are found to be mainly attributed to black carbon (BC) (Rosen, Hansen, Gundel, & Novakov, 1978). Later studies demonstrate the important influence of BC on the regional and global climate (Hansen & Nazarenko, 2004; Jacobson, 2002; Menon, Hansen, Nazarenko, & Luo, 2002; Ramanathan & Carmichael, 2008).

To further explore the spatial and temporal gradient of BC concentration in the atmosphere and its environmental consequence, confidence in an emission inventory and trends of BC are of great importance. Emission inventories are required inputs for atmospheric model simulation. Moreover, they are also necessary for the policy development of air quality and climate change since they contain information on feasibility and mitigation potential for all sources of emission.

1.1. Black carbon and its impact

Black carbon (BC) is a type of carbonaceous material produced by incomplete combustion of carbon-based fuel. Primary sources include emissions from incomplete combustion of fossil fuels for transportation, biofuel for industrial and residential uses, and open burning of biomass (Bond et al., 2007; Bond et al., 2013; Dickerson et al., 2002). BC is refractory and vaporizes at nearly 4000K. It is stable in the atmosphere and mainly removed by wet deposition. The way it forms makes it a good tracer of certain kinds of incomplete combustion (e.g. diesel vehicle and low-technology solid fuel burning).
It has been widely acknowledged that exposure to particulate matter (PM) is correlated with increased mortality (Lelieveld, Barlas, Giannadaki, & Pozzer, 2013; Pope, Ezzati, & Dockery, 2009). As part of particulate matter and good tracer of incomplete combustion, BC is a valuable additional air quality indicator to evaluate adverse health effects of air pollution dominated by combustion. Furthermore, a meta-analysis of health risks found that the health response to BC was more robust than the health response to PM mass. Estimated health effects of a 1μg/m³ increase in exposure were greater for BC than for PM₁₀ or PM₂.₅. The estimated increase in life expectancy of a hypothetical traffic abatement measure was four to nine times higher when expressed in BC compared with an equivalent change in PM mass (Janssen et al., 2011). Also, BC is a major component of particles produced by wood and vegetation burning. Health study in Seattle, Washington demonstrated that combustion-derived PM, which is mainly contributed by burning vegetation and mobile sources, is associated with airway inflammation in adult subjects with asthma (Jansen et al., 2005).

BC plays an important role in the climate. BC can influence the Earth’s radiative balance by absorbing visible light and warming the atmosphere (Bond, Zarzycki, Flanner, & Koch, 2011; Hansen & Sato, 2001; Schulz et al., 2006; Smith, Wigley, & Edmonds, 2000). BC’s lifetime ranges from several days to weeks (Chung & Seinfeld, 2002; Schulz, et al., 2006; Zhang et al., 2012). The short lifetime produces quick response of ambient air concentration to emission reductions, which means climate forcing from BC is transient. Therefore, reducing black carbon could present quicker climate benefits compared with the greenhouse gas of which lifetime is much longer. The warming potency of sources emitting BC vary due to co-emitted climate cooling agents such as organic carbon and sulfur species (Bond, et al., 2013; Bond, et al., 2011; Jacobson, 2001; Schulz, et al., 2006). Therefore, emissions that are rich in BC have larger warming effects than other sources.

BC has an influence on the formation and properties of water and ice clouds. This is an indirect, cooling climate effect with large uncertainties (Chung & Seinfeld, 2005; Hansen et al., 2005; Johnson, Shine, & Forster, 2004). Additionally, BC particles that deposit on snow and ice surfaces decrease the albedo and accelerate snow and ice melting (Bond, et al., 2013; Flanner, Zender, Randerson, & Rasch, 2007; Hansen, et al., 2005; Jacobson, 2004; Koch et al., 2009). The loss of snow and ice cover further reduces the planetary albedo because underlying surfaces
(e.g. soil or open water) are darker than snow. This is a positive feedback process that amplifies the global warming effect. Land-based glacier melting could raise the sea level and threaten the life and property of coastline residents. The loss of glacier melt would decrease the water supply to large river and lake systems that support millions of people globally (Barnett, Adam, & Lettenmaier, 2005; Menon et al., 2009).

1.2. Constraining historical emission

The aim of this work is to improve the accuracy of the black carbon (BC) historical emission inventory of the United States over the period of 1960-2000. To accomplish this goal, I interpreted the emission into observable variables and compared them with observations. Our constraining process differs from those comparisons in two ways. First, I acknowledge that atmospheric transport is a key factor in determining concentrations. Without an evaluation of the effects of transport, no conclusions can be drawn about emissions (Wang et al., 2014). Transport in global models is investigated in the context of interpreting observations. Second, improvements in emissions of BC are being interpreted in terms of the technology that would produce those emissions. I explicitly represent activities that have taken place throughout history, including changes in combustion technology. Changes in emissions imply changes in fuel use and emission factors of technology. When there is a mismatch between models and measurements, and the emission inventory is at fault, an error in estimated fuel use or technology is implicated. This in turn affects emission estimates of all pollutants. I intend to use the annual and seasonal trend in the observations of BC to constrain our current representation of fuel use and emission factors and then provide more accurate tabulations of emission quantities of BC and other species.
CHAPTER 2
METHODOLOGY

The purpose of this work is to improve the accuracy of historical BC emission estimation based on the corresponding measurements. To achieve this goal, I firstly interpreted emission into concentration and then analyzed the discrepancy between the modeled concentrations and its measurements. There are three main calculations in the method. The flow chart in Figure 1 describes the calculation process. First, transport matrices are calculated. A tagging run of the Community Atmospheric Model (CAM) was conducted starting with a BC emission inventory (Bond, et al., 2007) to simulate ambient air concentration. In the tagging run, emissions from different source regions are tagged as different “species” in the simulation. In this way, I can distinguish BC concentrations originating from different sources. Using the model simulation result, the relationship between emissions and ambient air concentration was then formulated into transport matrices. Here, I use the word “transport” to indicate all the chemical and physical processes occurring between emission and the resulting ambient air concentration in any location. This calculation is further described in Section 2.1. Secondly, historical emission inventories are reconstructed (modeled) into concentration with transport matrix and historical emissions so that they are comparable with the BC concentration measurements. I reconstructed the BC concentrations in California and New Jersey where coefficient of haze (COH) data for the 1960s to the 2000s are available. Third, potential errors in historical emissions are compared with reconstructed and measured historical BC concentrations. The discrepancy between the reconstructed ambient air concentrations and the measurements identifies required adjustments to the emission inventory.

The simulation of CAM on which the transport matrix is based and the simulation investigating of stability of long-term historical transport was run by Mark Flanner and Chaoyi Jiao from University of Michigan, Ann Arbor. The COH data and BC reconstruction is provided by Thomas Kirchstetter from Lawrence Berkeley National Laboratory and University of California, Berkeley. I also discuss an investigation of spatial resolution using a simulation of the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem). This model was run by Wayne Chang from University of Illinois. The remaining analysis described in this thesis is my contribution to this project.
2.1 Transport Matrix

The aim of calculating the transport matrix is to develop a formal method for comparing modeled and measured concentration trends without performing a simulation for each period. The concept and function of our transport matrix is same as the Jacobian matrix in the inverse modelling method. This treatment, with expansion to include uncertainties, plays a role in the classical inverse modeling formulation (Jacob, 2007). The Jacobian matrix, defined as $K=\frac{\partial y}{\partial x}$, represents the sensitivity of the concentration ($y$) to the emission ($x$) (Jacob, 2007). It builds a connection between the emission and the ambient air concentration driven by the emission. In our study, I assume this relationship is linear and that the transport matrix does not depend on the magnitude of emission. Therefore, I used the same set of transport matrices to interpret varying emissions into concentrations and vise versa. The transport matrices were numerically constructed from a simulation over the period of 2000-2006, conducted with the NCAR CAM version 4.0 running at 1.9x2.5 degree horizontal resolution, forced with transient sea surface temperatures (SSTs) over 1850-2005. The bulk aerosol model was used for the aerosol module. The model tracked 23 BC tracers originating from 23 regions, of which 17 are fossil and biofuel emission sources and 6 are biomass emission sources.

In Equation (1), average ambient air concentrations of $m$ receptor regions are denoted as the vector $\vec{C} = [c_1, c_2, ..., c_m]$, and emissions from $k$ source regions are denoted as $\vec{E} = [e_1, e_2, ..., e_k]$. Then, the transport matrix is the $m \times k$ matrix transferring the emission vector $\vec{E}$ into the concentration vector $\vec{C}$:

$$\vec{C} = T\vec{E} \quad (1)$$

As mentioned before, each element in the transport matrix is the sensitivity (response) of the concentration of receptor region to the emission of a source region. Since the transport is influenced by meteorology, I calculate the result on a monthly basis. In this case there are 23 emission source regions and two receptor regions. Emissions from three regions—U.S. fossil fuel and biofuel, North American biomass and Central American biomass—account for 94% of the total concentration of California. Over 99.7% of the total sensitivity of New Jersey originates
from U.S. fossil fuel and biofuel, Canadian fossil fuel and biofuel and North American biomass. The full transport matrices for California and New Jersey are shown in Appendix A.

One important concern about the transport matrix is whether it changes during a long-term historical time. To explore this, Mark Flanner and Cahoyi Jiao from University of Michigan examined variations in BC deposition within a GCM simulation over the period 1850-2005, conducted with CAM3.5 running at 1.9×2.5 degree horizontal resolution. It prescribed interannually-changing SSTs over this period, and applied the same BC emissions each year. Decadal variability in normalized deposition was found to be quite small (standard deviation <0.3) everywhere, except in one area of the South Pacific where absolute deposition is small and out of the scope of this study (see Figure 2). Though normalized interannual variability can be large due to El Nino Southern Oscillation (ENSO) and other high frequency variability, these fluctuations have less relevance for interpreting long-term changes in observational records of BC. In summary, transport variability has only a second-order impact on observed decadal-scale variability in BC deposition over 1850-2005, relative to variability in emissions. Therefore, I can reasonably apply our transport matrix extracted from a 7 year simulation to a 50-year study.

The value of representing transport, emissions, and concentrations as a matrix equation is twofold. First, the equation can be inverted to obtain emissions, given observed concentrations. However, the number of observations needs to be equal to or greater than the number of emissions to constrain. This can be achieved by merging emissions from less influential source regions. Second, since the transport matrix is relatively invariant for long-term, the forward version of this equation can be used to predict historical concentrations given emissions in different regions, thus avoiding a series of long model runs.

2.2 Measurement Data

BC ambient air concentration data are estimated from COH data acquired from the California Air Resources Board (CARB), the New Jersey Department of Environmental Protection (NJDEP), and the Environmental Protection Agency’s Air Quality System. COH was one of the first measures of particulate matter air pollution, which was introduced in the early 1950s (Hemeon et al., 1953). The measurements of COH are similar to those of BC (Hansen et al., 1984). Both of them are based on the quantification of light transmission through a filter with
particles on it. Studies indicated that COH is a reasonable measure of BC and that past BC concentrations can be inferred from records of COH concentration (Allen et al. 1999; Kirchstetter et al. 2008). In this study, \( \text{BC (\( \mu g \cdot m^{-3} \)) = 6.7COH + 0.1} \) is used for conversion of COH to BC. The COH data are available for California and New Jersey during 1963-2011 and 1967-2005, respectively.

2.3 Analysis of Discrepancy between Estimation and Observation

There are four sources of discrepancy between the BC concentrations estimated from emission inventory and the observed BC concentrations: inaccuracy of model meteorology, seasonality of emissions, average magnitude of emission, and measurement methods. In this study, I mainly address the first three types of discrepancy. I will first identify the error of meteorology in the model and suggest corrections. Then, I will focus on discrepancies in the seasonality and magnitude of emissions, which is the primary aim of this study.

2.3.1. Investigation of Model Meteorology

Errors in model meteorology could cause discrepancy between estimation and measurements, which should not be attributed to the inaccuracy of the emission inventory. Therefore, I analyzed those meteorological parameters that could influence the modeled concentration by comparing the modeled values with the real world measurements. Measurements of wind speeds (U) and mixing heights (H) for 2007-2009, are acquired from NASA’s Modern-Era Retrospective Analysis for Research and Applications (MERRA). U and H are compared with the wind speed and the planetary boundary layer (PBL) in the model respectively. Both wind speeds are vertical averages within the mixing height or planetary boundary layer. Figure 3 shows the comparison of modeled and measured meteorology parameters in the Bay Area in California (one grid box in the model) and New Jersey (two grid boxes in the model). PBL and H could be different by a factor of 2-3. Atmosphere with higher mixing height has stronger dispersion and lower BC concentration. However, the wind speed has a seasonal difference between the model and measurements. Considering that the modeled concentration is lower than the measurements, especially in winter, the correction of wind speed is not explanatory for the discrepancy in California. The reason is that the seasonality of modeled concentrations are smaller than that of the observation, and the correction of wind speed will make the modeled seasonality even
smaller. Therefore, I did not adjust the modeled wind with the measurements at this time. Only PBL is adjusted with the measurement of H. I explored the possibility of adjusting PBL with distinct equations individually for each month and grid box within California. I found little increase in R-square when adding month and grid box number as categorical variable into the regression of BC concentration on PBL. This result means that it is satisfactory to adjust it with uniform equation for all months and grid boxes. Therefore, I used the same equation to adjust the model output with the measurement of H. I assume that BC concentration would inversely proportional to the mixing height or PBL when emission is constant. Then the adjustment is reflected in the T matrix as $T' = T \times \frac{PBL}{H}$. The adjustment is done individually for each of the 15 grid boxes in California based on observations in that grid box. New Jersey is covered by only two boxes; the same adjustment of PBL is conducted for both since there is not enough measurement data and there is little difference between the PBL of the two boxes in New Jersey.

Besides U and H, I also investigated the influence of precipitation on the BC concentration. An adjustment for precipitation amount would not be straightforward, because concentrations can be removed at any point between emission and deposition, so that precipitation in many grid boxes could affect the concentration in the receptor box. Nevertheless, I found that this influence would not explain the observed discrepancy, because a correction would introduce the opposite seasonality to the observation. Figure 4 shows the monthly precipitation of San Francisco County, California. There is more rainfall in winter than summer.

2.3.2 Trend Comparison

Resolution has always been a concern when comparing observation with model result. Observed data at a point are not equivalent to the modeled grid box average, especially for measurements of the urban sites where BC concentration gradient is steep. I investigated the spatial heterogeneity of the modeled BC concentration with different size of grid boxes to explore the influence of the resolution. I used WRF-Chem to model BC mass concentration over Los Angeles, California, with a resolution of 0.04°×0.04 °. Then lowered the resolution of the data by averaging the concentrations of small grid boxes into the concentration into larger grid boxes. Figure 6 shows the comparison of BC concentration using different grid box sizes, for Los Angeles, California, which is an urban region with big BC concentration gradient. It shows that BC concentrations simulated at different resolutions may vary by a factor of two. Therefore,
I suggest that discrepancies in trends are more instructive than absolute discrepancies, especially for evaluating global model results.

2.3.3 Constraining Seasonal and Baseline Emissions with Measurements

After transport matrices were adjusted with meteorological observations, I reconstructed the BC ambient air concentrations for 1960-2000. At this stage of analysis, there is no seasonal variation in the seasonal section of our emission inventory, which means that all emissions are constant throughout a year. I used the invariant seasonal emission to evaluate the magnitude of seasonal and baseline emission, as derived below. After quantifying the magnitude of the seasonal emission, I applied Heating Degrees Day (HDD) data to estimate the variation of seasonal emission (Section 2.3.3). By analyzing the trend of discrepancy between the estimation of BC ambient air concentration (reconstructed BC concentration) and observation (measured BC concentration), potential improvements in the historical emission inventory are identified. All calculations are conducted on a decadal basis. Emissions are divided into two parts based on seasonality: seasonal emissions that are varying with time and the baseline emissions that are constant throughout the year. The most plausible explanation for seasonality in emissions is related to winter heating supply. Therefore, the seasonal emissions are assumed to zero in summer, because there is no emission from heating. Baseline emissions and seasonal emissions are constrained separately, assuming that seasonal emission is zero in summer.

Assuming that BC ambient air concentration is linearly proportional to emission, the ratio of the measured BC concentration to the modeled BC concentration each month, is equal to the ratio true BC emission and estimated emission. Furthermore, emission has a baseline and a seasonal component, as shown in equation (2). Because the modeled concentrations had invariant seasonality, the denominator is constant for each month.

For the $i^{th}$ month in a decade:

\[
R_i = \left( \frac{C_{m}}{C_{re}} \right)_i = \frac{E_{b} + E_{s}'}{E_{b} + E_{s_i}} \quad \text{for } i = 1, 2, \ldots, 12
\]
where $C_m$ and $C_{re}$ are the measured and modeled BC concentrations. $E_b'$ and $E_b$ are the true and estimated baseline emission, respectively. $E_{s_i}'$ and $E_{s_i}$ are the true seasonal emission for each month and the annually averaged estimation of the seasonal component of emission.

The average of the monthly ratio between the measurement and the estimation, $\bar{R}_t$, is the ratio of magnitudes of the true emission to the estimated total emission, as shown in equation (3). The summer ratio, $R_{summer}$, is the ratio of true value to the estimation of baseline emission, since zero seasonal emission is assumed, as shown in equation (4). Here, data for June and July are used for summer. Additionally, by identifying sources that are likely to be seasonal, I can estimate the ratio between baseline emission and seasonal emission in the original emission inventory, denoted as $k$ (equation (5)). Original emission inventory is the emission before our adjusting and constraining work. It is opposite to adjusted emission inventory, which is the adjusted and constrained inventory. Combining $\bar{R}_t$, $R_{summer}$ and $k$, I can express the best estimate of the true emission. Equations (6) and (7) give the magnitudes of baseline and average seasonal emissions, based on the observational constraints.

\[
\bar{R}_t = \frac{E_b' + E_{s_i}'}{E_b + E_{s_i}} = a \quad (3)
\]
\[
R_{summer} = \frac{E_b'}{E_b} = b \quad (4)
\]
\[
\frac{E_b}{E_{s_i}} = k \quad (5)
\]
\[
E_b' = bE_b \quad (6)
\]
\[
\overline{E_{s_i}'} = (k(a - b) + a) E_{s_i} \quad (7)
\]

2.3.4 Seasonal Variation of Emission.

Two components are required to depict seasonal emission: magnitude and seasonality. Magnitude is the average of seasonal emission over all months, while seasonality is the variation about the average. I use a time factor to represent this variation, defined as a set of monthly coefficients with an average of one. This time factor represents variations in the seasonal emission.
Because I assume that seasonally varying emissions are associated only with heating, I use heating degree days to estimate this fluctuation. Equation (8) gives the definition of HDD, which describes the heating energy used to bring the house’s temperature to a desired temperature. According to the definition, more energy will be consumed when the ambient temperature is lower (Kaynakli, 2008; Quayle & Diaz, 1980; Stohl et al., 2013).

\[ HDD = \sum_{d=1}^{n} \max(T_{set} - T_d, 0) \]  

where HDD stands for monthly degree-days; \( T_d \) is daily temperature; and \( T_{set} \) is the temperature of the indoor environment.

Figure 5 shows the HDD data from the National Oceanic and Atmospheric Administration (NOAA) National Climate Data Center for the time period of this study (NOAA 2013). I use the value of HDD to calculate a time factor, as shown in Equation (9), and the best bottom-up guess of monthly emission is calculated by multiplying the average residential emission with this time factor.

\[ TF(HDD)_i = \frac{HDD_i}{HDD} \quad i = 1,2,...,12 \]  

where TF(HDD)_i is the time factor of HDD for the \( i^{th} \) month.
2.4 Figures and Tables

Figure 1 Flowchart of the method

Figure 2 Normalized standard deviation in decadal averaged BC between 1850 and 2000, from the CAM3.5 simulation with varying SSTs and non-varying BC emissions. Decadal variability in normalized deposition is quite small (standard deviation <0.3) everywhere, except the South Pacific where absolute deposition is small and not much affected by the emissions. Simulation are conducted by Mark Flanner and Chaoyi Jiao from University of Michigan, Ann Arbor.
Figure 3 Monthly average of the wind speed vertical average, mixing height (measurement) and planetary boundary layer (model) of (a) Bay Ares in California (b) New Jersey. Measurements of wind speeds (U) and mixing heights (H) for 2007-2009, are acquired from NASA’s Modern-Era Retrospective Analysis for Research and Applications. Meteorological variables from the model are from the CAM 4.0 model simulation run by Mark Flanner and Chaoyi Jiao from University of Michigan, Ann Arbor.
Figure 4 Average monthly precipitation over 1971-2000 in San Francisco County, California. (Data acquired from http://www.rssweather.com/climate/California/San%20Francisco%20County/)

Figure 5 Monthly average of HDD for California and New Jersey over each decade during 1960-2000. HDD shows a bigger seasonal difference in New Jersey than California because of colder winter there. Data are calculated from National Oceanic and Atmospheric Administration (NOAA) National Climate Data Center
Figure 6 Comparison of BC concentration distribution for an urban region with different resolution. Result with higher resolution shows higher gradient within the same grid box. BC with different resolution could vary by a factor of two. This implies the uncertainty of absolute magnitude comparison and indicates that trend comparison could be more instructive. This simulation of the Weather Research and Forecasting model coupled with Chemistry was run by Wayne Chang from University of Illinois
CHAPTER 3
CONSTRAINING EMISSION PROCESS AND THE ADJUSTED EMISSION INVENTORY

The constraining work was conducted with two emission inventories. The first emission inventory is from Bond et al., (2007) with fossil fuel use data from United Nations (1995) through 1980 and International Energy Agency (IEA) (2004) after 1980, as well as biofuel data from Fernandes et al., (2007). The second one is an updated version of the first emission inventory, with two major differences (Liu et al. 2014). First, fuel consumption data are taken from the Energy Information Administration (EIA, 2010), the United States national agency responsible for collecting energy statistics. These data are disaggregated by state, whereas the IEA data are given for the whole nation. The second difference is the use of the SPEW-Trend vehicle fleet model (Yan, Winijkul, Jung, Bond, & Streets, 2011) to estimate the number of vehicles built according to different standards that are emitting in each year. This fleet-model approach gives a more realistic estimate of trends, because emissions do not cease when emission standards come into force, but rather when old vehicles retire from the road.

Figure 7 shows comparison of United States BC emissions between the two emission inventories for (a) vehicle emission (b) residential emission. For the vehicle sector, the emission inventory from Bond et al. (2007) has a sharp peak in 1975, while that from Liu et al. (2014) gradually peaked at 1980-1985 and then decreased. The ratio of the sector emission between the new inventory and the old emission inventory is showed as the green line in Figure 6. The ratio become higher in recent decade and the average of it over the 50 years is 1.8 for vehicle emission. For the residential emission, both emission inventories show a gradual decrease during 1960-2000 except the new emission inventory has a bump in 1980-1985. Again, the ratio of this sector increased with time and the average ratio is 3.1. Although the ratio of the sector emissions between these two emission inventories increased with time, the gap between them actually deceases since the magnitude of emissions is getting smaller. The sharper decrease in the emission inventory from Liu et al. (2014) caused by the more realistic implementation of vehicle standards. Figure 8 shows the comparison of trends in these two emission inventories. The magnitude of total US BC emission inventory from Bond et al. (2007) is much lower than that from Liu et al. (2014). The emission estimation from the latest fuel use data is about a factor of three higher than the previous inventory for the time before 1990. The first inventory peaks at
1975 with a gentle decrease throughout 1960s-2000s. The trend in the second emission inventory, however, shows a strong decrease in the same period with a bump in 1980-1990.

Additionally, there is high heavy fuel oil consumption for international shipping in the transport sector in the emission inventory using new EIA data. It contributes to the emissions in transport sector (off road) which are different from the vehicle (on road) emission. Emissions from the transport sector, which is dominated by the international shipping emission, account for over 50% of the total emission before 1995. However, the international shipping emission would occur outside the U.S. that would have little contribution to the US BC concentration. Therefore I did not include this part of emission when interpreting the emission into concentration (not shown in Figure 8)

3.1 Constraining the Adjusted historical emission inventories

The measurement data, original modeled BC, adjusted modeled BC and the intermediate modeled BC for California and New Jersey are shown in Figure 9 and Figure 10. In both cases (California and New Jersey), BC directly modeled from the original emission inventory is represented by the solid blue line. Comparison of these original modeled BC concentrations (solid blue line) with the measurements (green line), shows discrepancies in magnitude, seasonality and trend. The magnitudes of BC concentration modeled from both emission inventories are lower than the measurements. Secondly, BC concentrations modeled from both emission inventories show limited seasonal variation, because of attributed to the inaccuracy of the meteorology in the model and the absence of seasonality in the emission inventory. Finally, the trend of the measurement is decreasing throughout the study period, while both modeled BC from both inventories shows a peak in 1980s.

As discussed in Section 2.3.1, an adjustment for the model PBL was included. Since the PBL in the model is higher than the measured mixing height, correcting the meteorology increases estimated BC concentrations. This adjustment increases both the magnitude and seasonality (high in winter and low in summer) as indicated by the dashed blue lines in Figure 9 and Figure 10. The remaining discrepancy between the modeled BC and measurement is attributed to the baseline and seasonal emissions of the two inventories. Modeled BC from the first emission
inventory matches the magnitude of measurement during the 1980s and later for both receptor regions.

In the final step, I first used the remaining discrepancy to calculate adjustments for the baseline emission and seasonal emission in the adjusted emission inventory with the method described in section 2.3.2. HDDs were used as a time factor for the sector expected to be seasonal (residential sector). For California, the final modeled BC matches the measurement well with an expectation of small overestimation of seasonality in 1980s. The reason for this mismatch in the 1980s is that seasonality in the modeled BC is equal to the seasonal emission multiplied by HDDs, and the seasonal emission is high early in the comparison time.

Since the HDDs remain about the same for the decades under study (see Figure 5), higher adjusted emissions resulted in greater seasonality in the modeled BC. However, the seasonality in the measurements (gap between winter and summer) remains about the same although the total emissions are decreasing. This difference indicates that HDDs cannot fully represent the changing seasonality in the residential emissions. Other factors that describe changing emissions should be incorporated to reproduce the historical BC trend. For New Jersey, BC modeled from the adjusted emissions does not reproduce the observed decreasing seasonality in measurements. Unlike California, New Jersey does not have stagnant meteorology conditions in winter. Therefore the seasonality of BC concentration should be mainly contributed by the seasonal emission. The decreasing seasonality demonstrates that seasonal emissions were decreasing due to pollution control. To reproduce this trend I will need more than HDDs. Additionally, the model meteorology does not perform well for New Jersey, although I adjusted the PBL in the model. The dashed blue line shows a strong seasonality when the HDDs have not been included, which is not consistent with observations in New Jersey. More model investigation of the New Jersey and other east coast region should be conducted.

3.2 The adjusted emission inventory

The adjusted emission inventories are shown in Table 1 and Table 2. Although the two sets of priori emissions differ a lot, the adjusted versions are similar because both are adjusted to observations. For California, the two sets of the adjusted emission agree well. For New Jersey, where the modeled meteorology does not fit the seasonality of the observation well, there is a
mismatch of the adjusted emission especially for 1990s and 2000s. This observation indicates the importance of well-reproduced atmospheric transport and meteorology, which is also the basis of our constrained calculation process described in 2.3.3. Figure 11 and Figure 12 describe the comparison between the adjusted emissions and the original emissions for California and New Jersey. Although both figures show proposed adjustment of the current US emission inventories, they are dominated by different local emissions, since they are calculated with measurements from two different states. Emissions within the state will affect the local BC concentration most. Therefore, the difference in the adjusted US emission based on measurement of California and New Jersey could be explained by the emission composition in these two states. In California, the adjusted ratios of seasonal emission for different decades are all higher than the baseline emission, indicating that there should be more seasonal emission for California. Besides the residential sector, there may be seasonal emission from other sectors. For New Jersey, the adjusted seasonal is much lower and even below zero in recent decades. This unreasonable result indicates the problem of the seasonality reproduction from the inaccuracy for model meteorology and the HDDs for New Jersey. As to the trend, the adjustment ratios all show a drop for 1980s to construct a decreasing trend for the 40 or 50 years. In this current emission inventory, there is not change of split between normal emitter and super emitter for the heavy oil consumption of international shipping which account for about 50% of the total emission. Also, the split between fireplace and heating stove remains same for the wood burning in residential sector, which dominates the bump of the emission during 1980s. To fully explore the constraining of the emission inventory, more work on the fuel consumption and the distribution of them among different technologies with district emission factors need to be done.
3.3 Figures and Tables

Figure 7 Comparison of United States emissions of black carbon between Bond et al (2007) and Liu et al (2014) for (a) Vehicle emissions (b) Residential emissions
Figure 8 United States emissions of black carbon  
(a) from Bond et al (2007)  
(b) from Liu et al (2014)
Figure 9 Modeled and Measured BC concentration result for California with (a) emission inventory from Bond et al., (2007) and (b) updated emission inventory.
Figure 10 Modeled and Measured BC concentration result for New Jersey with (a) emission inventory from Bond et al. (2007) and (b) emission inventory from Liu et al. (2014)
Figure 11 Comparison between the adjusted US BC emission and the original US BC emission in the case study of California with (a) emission inventory from Bond et al. (2007) and (b) emission inventory from Liu et al. (2014)
Figure 12 Comparison between the adjusted US BC emission and the original US BC emission in the case study of New Jersey with (a) emission inventory from Bond et al. (2007) and (b) emission inventory from Liu et al. (2014)
Table 1: The adjusted US BC emission based on the measurements of California

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Adjusted result of emission inventory from Bond et al. (2007)

Table 2: The adjusted US BC emission based on the measurements of New Jersey

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Adjusted result of emission inventory from Liu et al. (2014)
CHAPTER 4
CONCLUSION AND FUTURE WORK

To improve the accuracy of US historical BC emission estimation for 1960-2000 based on the corresponding measurements. I firstly calculated transport matrices based on the simulation result of a tagging run of CAM 4.0. Then I reconstructed (modeled) historical BC concentrations for California and New Jersey with transport matrix and the current historical emissions inventories, where BC measurements are available. Third, I analyzed the discrepancy between the reconstructed ambient air concentrations and the measurements to identify required adjustments to the emission inventory.

Based on a comparison between modeled and measured concentration, I hypothesize that US BC emissions were underestimated by Bond et al. (2007) and overestimated when applying the EIA 2010 data for BC estimation. The overestimation of the updated emission is probably due the high energy consumption in international shipping, which does not affect concentrations in the US. Therefore, it cannot be used as part of US emission input for the transport matrix since it has different transport comparing with the emission within US. In the trend comparison, there is a peak in the 1980s observed in both emission inventories but not in the measurement. This may indicate that we should consider a change of splits among different technologies (shift to cleaner technology) which can cancel out the influence of energy consumption increase.

The modeled BC with the adjusted emission fits better in California than New Jersey. Also, the adjusted emissions from the two set of emission inventories converge better for California than New Jersey. These results may be affected by the inaccuracy of model meteorology in New Jersey. Unlike California, there is no stagnant winter condition in New Jersey; however, high winter BC prediction is observed from the modeled BC before applying HDDs for seasonal emissions. More understanding of the behavior of the model in east coast would be valuable for this kind of emission constraining work.

An accurate emission inventory has always been important for atmospheric model study and policy making. During this study, I applied the principle of inverse modeling, into a method that could utilize the BC concentration measurement as constraint to adjust the current emission inventory of BC. This method provides a relatively simple process to reconstruct concentration from emission, thus avoiding long historical model simulations. I also investigated the role of
atmospheric transport and meteorology in the emission-measurement comparison process and quantified its influence based on the current available information.
REFERENCES

Allen, GA; Lawrence, J; Koutrakis, P (1999) Field validation of a semi-continuous method for aerosol black carbon (aethalometer) and temporal patterns of summertime hourly black carbon measurements in southwestern PA, Atmos. Environ. 33, 817–823.


Climate for San Francisco County, California, data retrieved from http://www.rssweather.com/climate/California/San%20Francisco%20County/


## APPENDIX A TRANSPORT MATRIX

Table 3Transport matrix with the receptor region of California and 23 global emission source regions

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### Table 4 Transport matrix with the receptor region of New Jersey and 23 global emiss Constraining the Adjusted historical emission inventories ion source regions

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