

Analysis of Spatial and Temporal Trends of Black Carbon in Boston

George Allen

**Prepared by
NESCAUM**

January, 2014

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Acknowledgments

NESCAUM gratefully acknowledges the funding support provided by the Massachusetts Department of Environmental Protection (MassDEP) and the U.S. Environmental Protection Agency. BC Data from the Countway Library Brigham Circle site was provided by the Harvard School of Public Health.

This report was a joint effort between staff at NESCAUM and MassDEP. NESCAUM is an association of the eight northeast state air pollution control programs. NESCAUM provides technical guidance and policy advice to its member states.

NESCAUM also thanks the following individuals for providing review and comments on this report:

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Printed: January 2014

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Executive Summary

NESCAUM, with funding from MassDEP and the U.S. EPA, conducted a study to evaluate spatial patterns and temporal trends of black carbon soot (BC) in Boston.

Key Findings

1. Concentrations of black carbon (BC, or soot) in Boston decreased substantially between 2002 and 2004. During this time period, diesel bus retrofit and State diesel Inspection and Maintenance (I/M) programs were implemented in Boston. The observed decrease in BC is likely due to these programs, and demonstrates the effectiveness and benefit of such programs.
2. From 2005 to 2012, BC concentrations in Boston have not dropped at two of the three long-term monitoring sites.
3. BC was substantially higher in the urban area where there are more mobile sources.
4. Urban BC was associated with time of day and day of week traffic patterns.
5. Within the urban area, there was substantial local scale heterogeneity.
6. Of the four BC sites in operation since 2009, the highest annual mean BC was observed at the NESCAUM office South St. site. Wind analysis suggests traffic emissions from the Southeast Expressway as the likely cause of the higher BC concentrations at this location.
7. Point sources of BC (e.g., emergency diesel gensets) can produce very large short-term (~20 minute) spikes of BC.

ES-1. Introduction

This report presents an analysis of spatial and temporal trends of black carbon soot (BC) in Boston, Massachusetts. BC is an optical measurement of how dark an aerosol is, and has been shown to be well correlated with elemental carbon (EC) measurements. BC is a useful indicator of local mobile source aerosol emissions in urban areas.

Local mobile sources in large urban areas contribute to elevated levels of a wide range of air pollutants associated with adverse health effects reported in a wide range of epidemiological studies. More recently, BC has been cited as a factor in near-road health effects.

From a policy perspective, an improved understanding of the spatial patterns (gradients) and long-term temporal trends of mobile source-related PM in large urban areas, as represented by BC, can inform control strategy assessment and implementation. It can also aid in understanding exposure dynamics of potential environmental justice-related hot-spots, such as the Dudley Square area of Roxbury (Boston) and can contribute to better understanding and improving estimates of exposures used in health effect studies. This project analyzed BC in the metropolitan Boston area, with the goal of better understanding urban gradients and temporal trends of BC (i.e., locally generated mobile source aerosols, including diesel PM) over the last decade.

In December 2002, NESCAUM started a one-year exploratory study to investigate the spatial extent of elevated BC from traffic, using two existing long-term BC monitoring sites (Roxbury and Brigham Circle) and four new sites. This project reanalyzed these data, using the full year of data and with improved data processing techniques. Site locations spanned from downtown Boston to the town of Stow, which is just inside the I-495 loop. Sites were located to be representative of neighborhood-scale concentrations, and avoid influence from heavily trafficked roads. Table ES-1 lists the six sites and the MassDEP North End site that started taking BC measurements in July 2003. Distance from the Joy Street Beacon Hill site, considered the centrally located site, and a general description of site characteristics are presented.

Table ES-1. Description of Core Monitoring Sites.

7 Core Site Locations	Km from Beacon Hill Site	Site Description
Joy Street, Beacon Hill (Boston)	0.0	Urban Residential (near State House)
North End (Boston)	1.1	Urban Residential/Commercial
Roxbury (Boston)	3.5	Urban Residential/Commercial; EJ
Brigham Circle (Boston)	4.1	Urban Residential/Commercial
Brighton (Boston)	7.0	Semi-Urban Residential
Waltham	14.9	Suburban Residential/Light Commercial
Stow	35.3	Semi-rural, open land, Regional Background Site for Metro Boston

Figure ES-1 presents the distribution of hourly BC for periods noted. BC concentrations at the four urban sites (Joy Street, North End, Roxbury, and Brigham Circle) were all higher than the two suburban and one background sites. The North End and Roxbury sites were similar to each other. As expected, the Stow semi-rural background site was the lowest, with mean BC being roughly one-fourth of the North End and Roxbury sites.

Figure ES-1. Distribution of 2003 Hourly BC for Seven Core Sites.

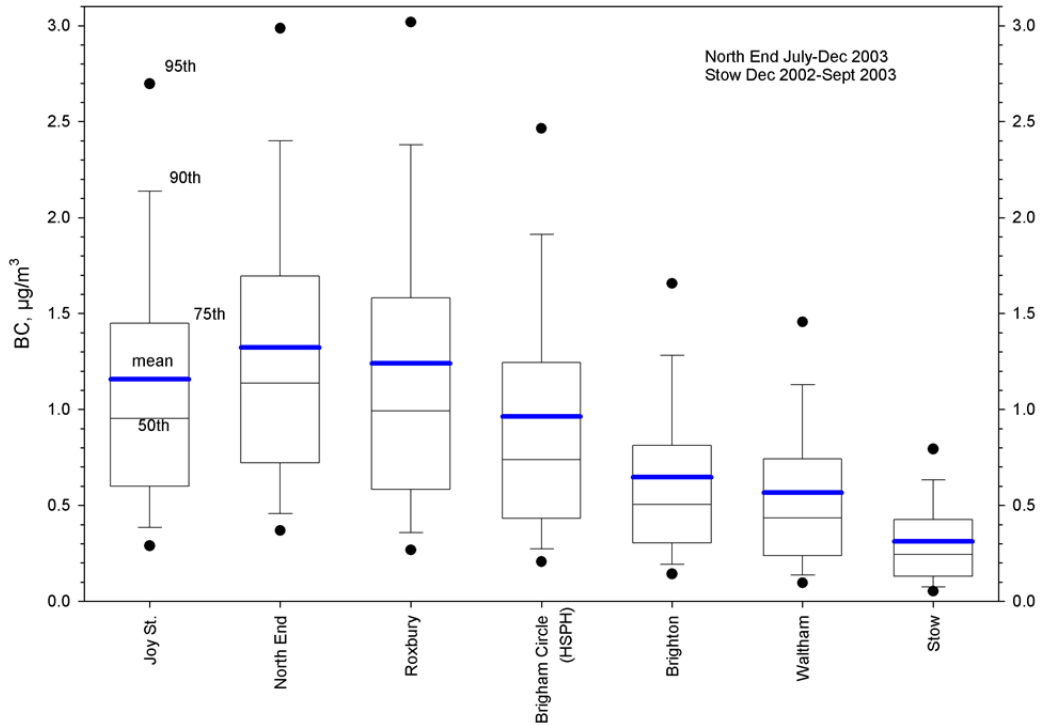
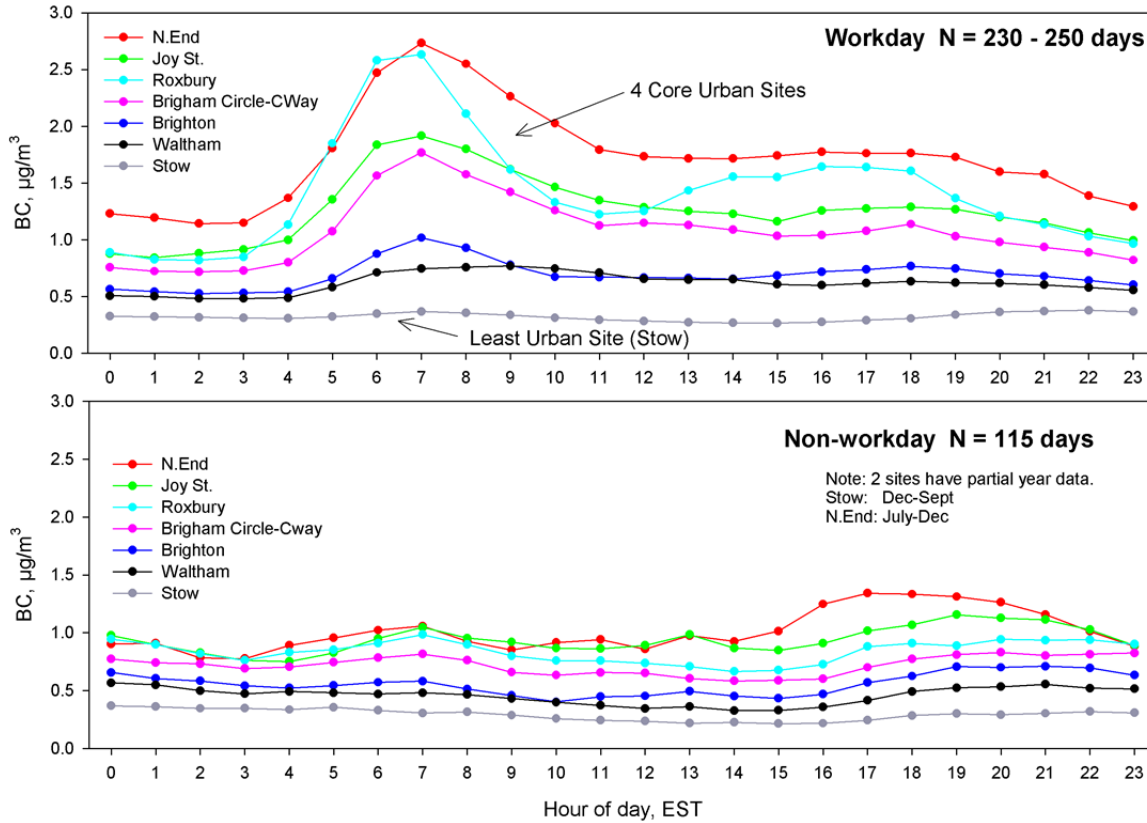


Figure ES-2 shows the diurnal BC pattern for the seven sites, with work days plotted separately from weekend days and holidays. For the weekday plot, the Roxbury and the North End sites had the highest morning rush-hour BC concentrations; the Joy Street and Brigham Circle sites were somewhat lower. The two suburban sites (Brighton and Waltham) showed a smaller morning rush hour peak relative to the rest of the day. The background site (Stow) showed no strong BC pattern for the entire day, consistent with its semi-rural location upwind of Boston.

For the non-weekday plot, there was no strong diurnal pattern, even for the core urban sites. This is consistent with expected non-weekday traffic patterns. The North End site showed its daily maximum in the early evening, from 5:00 to 7:00 pm. The multi-season weekday/non-weekday diurnal analysis provides increased confidence that BC is reasonably specific to local mobile source aerosol at these sites, minimizing concerns related to potential interferences at these sites from other sources of BC, such as oil-fired space heating and woodsmoke.

Figure ES-2. 2003 Diurnal Plot of Seven Core Sites.



A Boston Neighborhood Scale study was conducted for two months during the summer of 2003. Five additional sites were added to explore BC gradients within the urban core of Boston. Of all 12 sites, 10 were in Boston; 9 of which were within a radius of 2.5 km and in very urban settings. The Brighton site, while in the City of Boston, is in a semi-urban setting. Siting was representative of neighborhood scale (rather than hotspot/microscale) exposures. Table ES-2 presents, for each of the 10 Boston sites, their distance from the Joy Street site and general site characteristics.

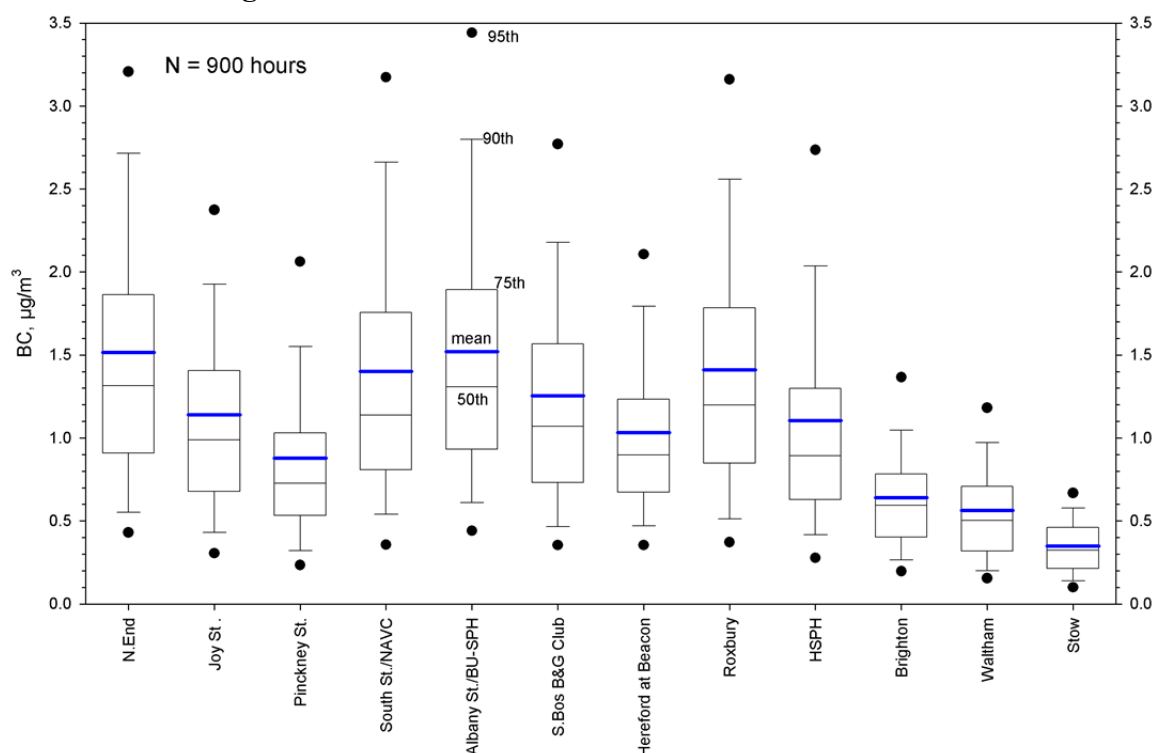
Table ES-2. Description of Sites for Summer Intensive.

Site Locations	Km	Site Description
Joy Street	0.0	Urban Residential/Commercial. (Beacon Hill, near State House)
Pinckney Street	0.3	Urban Residential (Beacon Hill)
North End	1.1	Urban Residential/Commercial (near the I-93 Expressway)
South Street	1.0	Urban Commercial (near South Station bus and train terminals)
Hereford Street	1.9	Urban Residential (Back Bay)
Albany Street	2.4	Urban Commercial (BU School of Public Health)

South Boston	2.9	Urban Residential
Roxbury	3.5	Urban Residential/Commercial
HSPH	4.0	Urban Residential/Commercial (urban scale)
Brighton	7.0	Semi-Urban Residential

Figure ES-3 shows the BC distributions for all 12 monitoring sites, limited to days where all sites had data. Approximately 20 days were excluded because two sites each had a 10-day period of missing data attributable to equipment malfunction and/or data loss attributable to sample collection and processing issues.

Figure ES-3. BC Distributions of 12 Summer Intensive Sites.

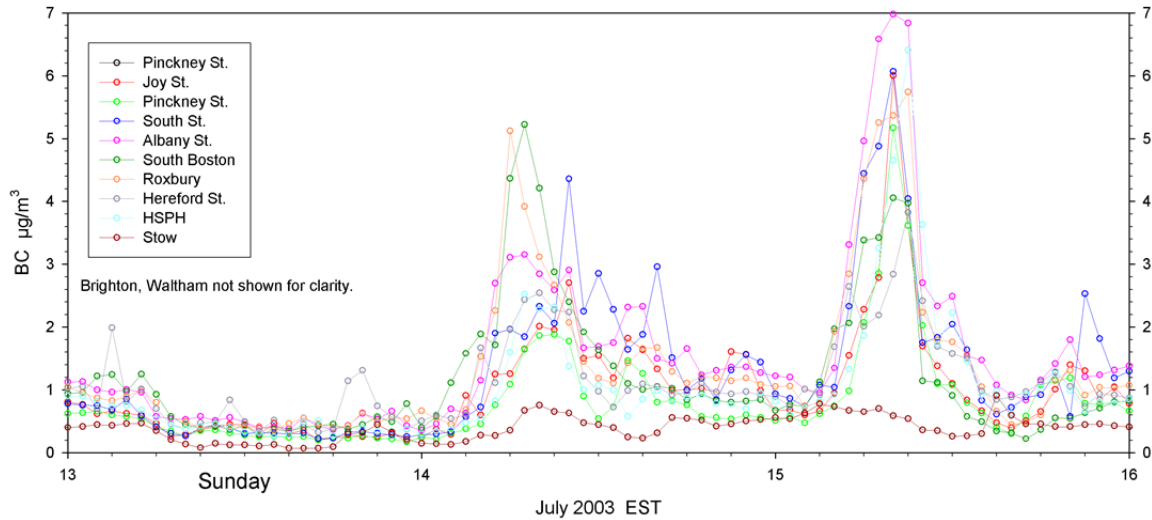


There were substantial gradients for mean BC at neighborhood scale-oriented sites in Boston. The observed variation across sites may be attributable to variability in monitor siting, mobile source strength gradients, and microscale meteorology. The North End and Albany Street sites were the highest, and the Roxbury and South Street (near South Station) sites were slightly lower. The urban Boston sites with the highest and lowest BC were North End and Pinckney Street (on Beacon Hill) locations. Although these sites are only 1.3 km (0.8 miles) apart, they exhibited a BC ratio of 1.7. The measured BC at the Hereford Street site was also relatively low, as it is near both Storrow Drive, where truck traffic is prohibited, and the Charles River.

Figure ES-4 is a time series plot that shows short-term patterns and gradients of hourly BC across the Boston area for July 13-15, 2003. A distinct “clean Sunday and dirty work-day” effect was observed. Tuesday, July 15, 2003 was one of the dirtier BC days of the summer;

several sites exceeded 4 micrograms per cubic meter ($\mu\text{g}/\text{m}^3$) BC for several hours during the morning. The ratio of these sites to the Stow background site for this peak period was approximately 10.

Figure ES-4. July 13-15, 2003 Event.



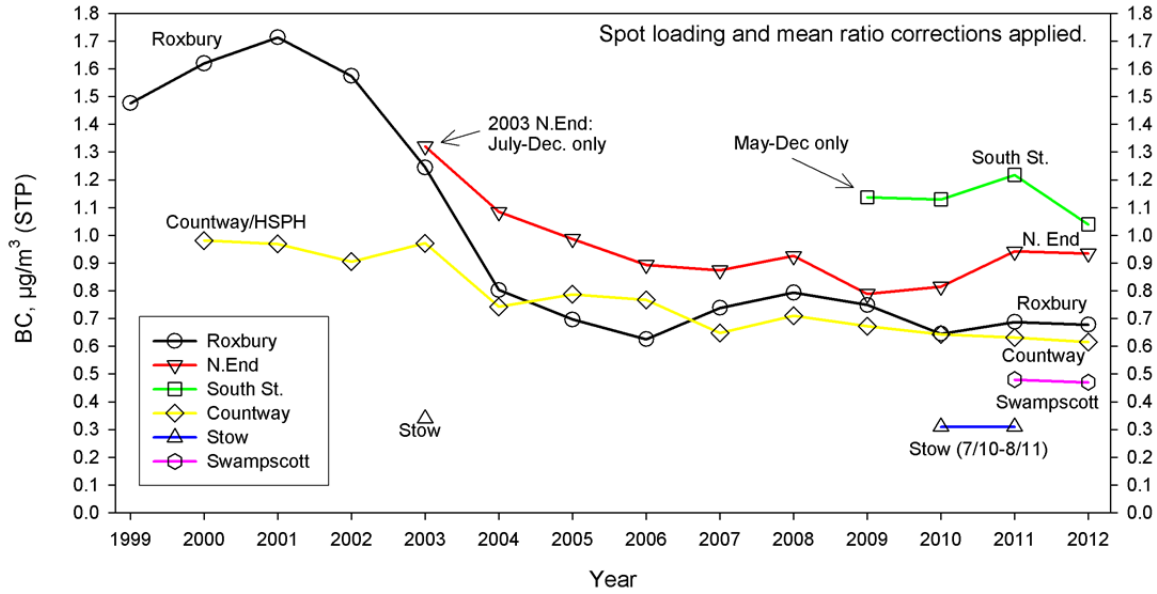
ES-2. Updated BC Spatial Analysis

The 2003 Spatial Study was conducted during a year when substantial reductions in heavy-duty diesel PM occurred due to the installation of PM control technologies on the MBTA and Boston school bus fleet. By spring 2005, both these fleets were entirely controlled for PM emissions. Trend analysis has shown a substantial drop in BC at the two long-term sites, and these changes may have affected the spatial patterns of BC.

From 2009 to 2012, additional BC monitoring sites were run specifically for this study to provide an updated assessment of the spatial scale of Boston BC. The Stow regional background site was run for two years (July 2009-August 2011), a permanent site at NESCAUM's South Street offices started in May 2009, and BC was monitored in Swampscott (21 km northeast of Boston) for the period 2011 through 2012.

Figure ES-5 shows the trend for the four Boston BC sites, along with the Stow 2003 and 2010 to 2011 means, and the 2011 to 2012 BC means for Swampscott. BC at the Stow regional background site dropped from 0.34 to 0.31 $\mu\text{g}/\text{m}^3$ over seven to eight years, consistent with reduced heavy-duty diesel PM emissions on a regional basis. Swampscott BC concentrations were lower than the Boston sites, but higher than the Stow site. Swampscott is downwind of Boston, and Stow is upwind; this may explain the relative BC concentrations at these two non-urban sites.

Figure ES-5. Trend for Four Boston BC Sites, Stow, and Swampscott.

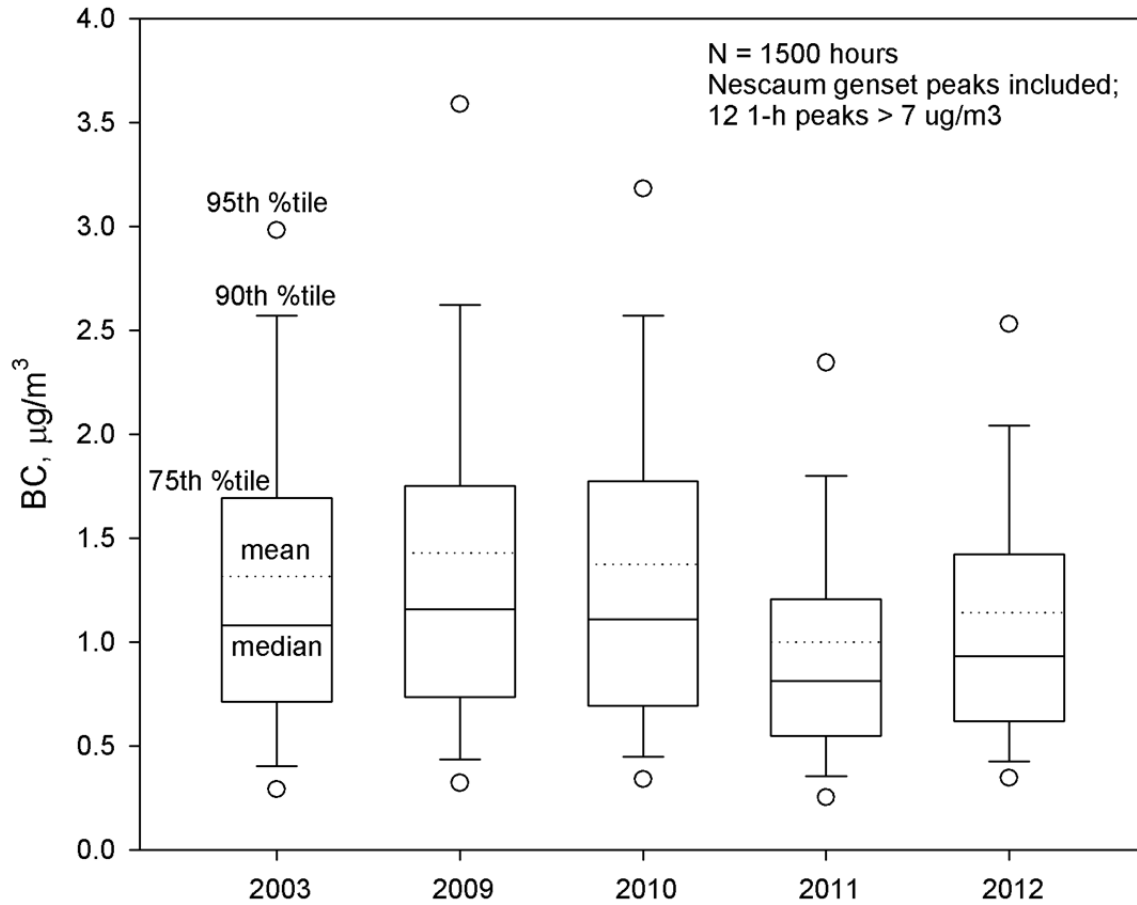


The NESCAUM South Street site measured the highest BC of all Boston sites for 2009 to 2012 (Figure ES-5). Mean BC was $1.13 \mu\text{g}/\text{m}^3$. The means for the other Boston BC sites ranged from 0.64 (HSPH) to 0.87 (North End) $\mu\text{g}/\text{m}^3$. The mean for Springfield was $0.87 \mu\text{g}/\text{m}^3$. South Station, a major transportation hub for rail and intercity buses, is 300 meters to the southeast of the South Street monitoring site, and was considered to be a potential source of the BC at South Street. However, analysis of BC and wind direction data clearly showed that winds from the south-southwest and southwest accounted for more BC on average than any other direction, and winds from South Station (southeast) contributed a relatively small amount of BC measured at the site. The lack of influence from South Station diesel sources may be due in part to the use of an active ventilation system for commuter rail engines and the bus depot that vents their exhaust at high velocity through four stacks above the roof of the bus depot parking garage. The exhaust is diluted and dispersed approximately 37 meters above street level. The Massachusetts Turnpike (I-90), the Southeast Expressway (I-93), and the large interchange between the two, are all to the south-southwest and southwest of the South Street site by approximately 650 meters. The large amount of traffic activity from this area and the first kilometer of the I-93 Expressway above ground to the south-southwest is a likely reason for the high mobile source-related BC observed at this site.

The South Street site used in 2003 (located at the rear of 112 South Street) and the NESCAUM site at 89 South Street site are 65 meters apart, and thus close enough to be compared over time. Figure ES-6 shows that the summer spatial intensive BC distribution for the 2003 site, along with the 2009-2012 BC data from the NESCAUM South Street site, matched for the same days of the year. The 2009 to 2010 BC distributions were similar to the 2003 data. The 95th percentiles were higher for 2009 to 2010, in part due to a local source.¹

¹ A diesel genset weekly test produced very high BC, up to $> 100 \mu\text{g}/\text{m}^3$ for one-minute concentrations, once a week for approximately 15 minutes.

Figure ES-6. Summer 2003 and 2009-2012 South St. BC.

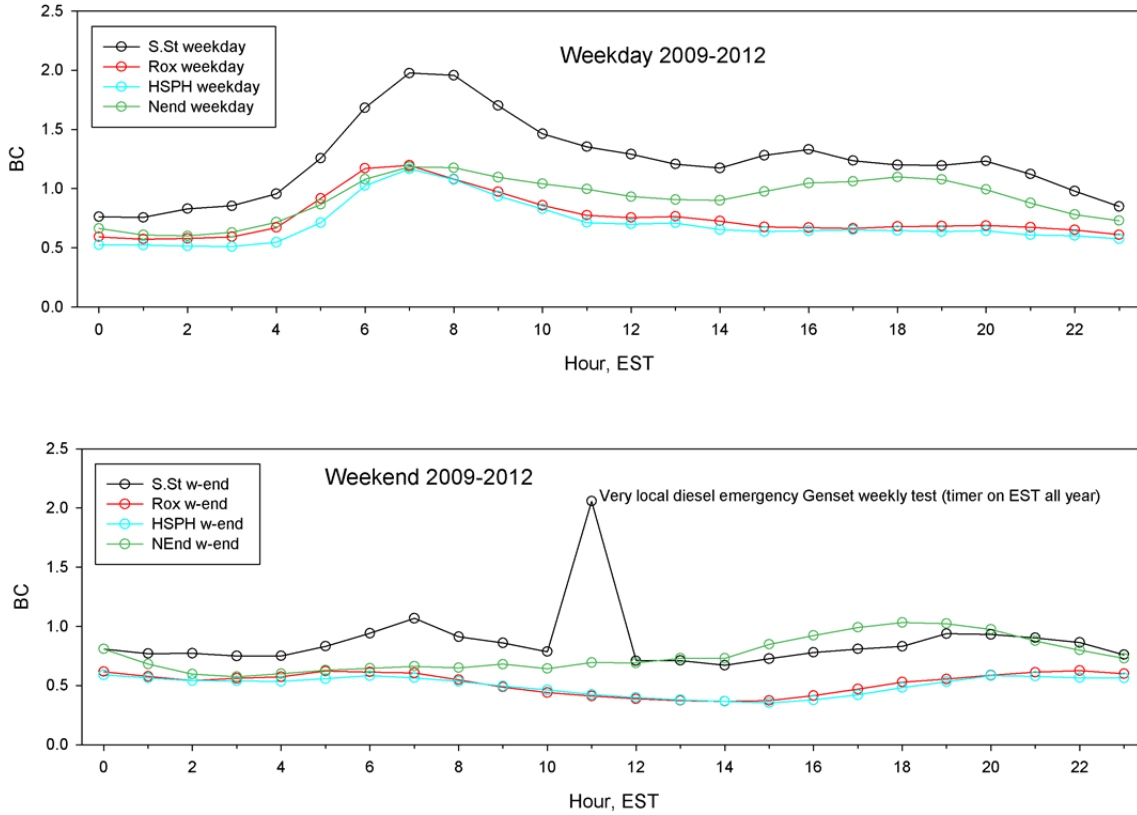


ES-2.1. Diurnal plots for 2009-2012, four Boston sites.

The weekday and weekend diurnal BC plots shown in Figure ES-7 have been updated for data from 2009 through 2012 for three of the Boston sites used in 2003. The NESCAUM South Street site was also added. The general patterns were unchanged, but as expected, the Roxbury site appeared to behave more like the HSPH site, with a smoother and less pronounced morning rush-hour peak that is characteristic of an urban-scale site.

The South Street site was highest for all hours during weekdays and weekends. The North End site was higher than Roxbury and HSPH for most hours of the day. As with the 2003 diurnal plots, there was a clear morning rush hour peak on weekdays, and no peak on weekends, except at South Street. That site also showed the effect of the weekly test of an emergency diesel genset on Saturdays at hour 11:00 a.m. EST.

Figure ES-7. Updated Diurnal Plots.

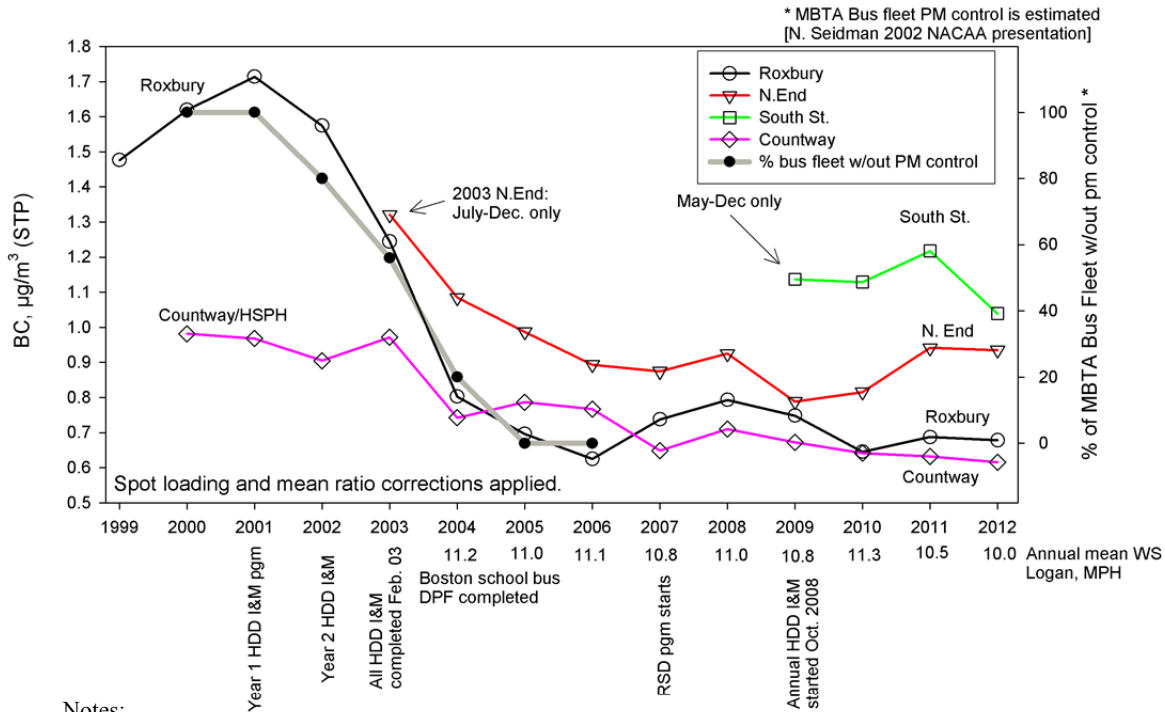


ES-3. Temporal Trends of Boston BC

There are several sites with multiple years of BC measurements in Boston. The Roxbury site started in February 1999, and the HSPH site started in October 1999. BC measurements were added to the MassDEP's North End site in July 2003. As part of this project, BC measurements commenced at NESCAUM's South Street, sixth-floor office (overlooking Tufts Street) in May 2009. Note that this is not the same South Street location used in the 2003 Boston Neighborhood Scale spatial summer intensive study.

Trend analysis was based primarily on annual mean BC concentrations. Figure ES-8 shows the BC trend data for the four Boston sites that had multiple years of data. Dates of the MassDEP's heavy-duty diesel control programs and the MBTA and Boston school bus fleet control programs are also indicated.

Figure ES-8. BC Trend Data for Boston Long-term Sites.



Notes:

- “HDD I&M” is the MassDEP heavy duty diesel inspection and maintenance program.
- “DPF” is diesel particulate filter
- “RSD program” is the MBTA’s bus garage Remote Sensing Device program for bus emissions.
- “Spot loading and mean ratio corrections” are measurement method artifacts that varied over time and could affect trend analysis if not controlled for.

There was a striking drop in Roxbury BC over the three-year period 2002 through 2004. This correlates with the progress of the MBTA’s bus fleet PM emission control program (the grey line from 2000 to 2006 in Figure ES-8), which resulted in the clean-up of 100% of the fleet between late 2002 and the end of 2004 (Seidman, 2002). The percentage of buses without PM controls is shown on the right axis of Figure ES-8. The entire Boston school bus fleet was also retrofitted with PM controls between 2003 and spring of 2005. Another factor in the reduction of Roxbury BC may have been the closing of the MBTA’s Bartlett Street bus garage near Dudley Square at the end of 2003.

ES-3.1. Post-retrofit Progress.

Starting in 2005 after heavy-duty diesel PM emissions from the MBTA bus and the Boston school bus fleets were fully controlled and the Roxbury Bartlett Street MBTA bus garage was closed, there was no clear trend in BC indicated at the North End and Roxbury sites. The annual mean BC concentrations for 2005 and 2012 for these two sites were essentially identical, at 0.70 and 0.68 $\mu\text{g}/\text{m}^3$ for Roxbury, and 0.99 and 0.94 $\mu\text{g}/\text{m}^3$ at the North End. Figure ES-9 shows the regression of annual mean BC versus year at the North End from 2005 through 2012. It shows no clear trend. The Roxbury BC trend from 2007 through 2012 (Figure ES-10) had a downward trend, but the regression was not significant ($p = 0.11$). The trend from 2005-2012 was weaker ($p = 0.8$, not shown).

Figure ES-9. Regression of Annual Mean BC vs. Year 2005-2012 for North End.

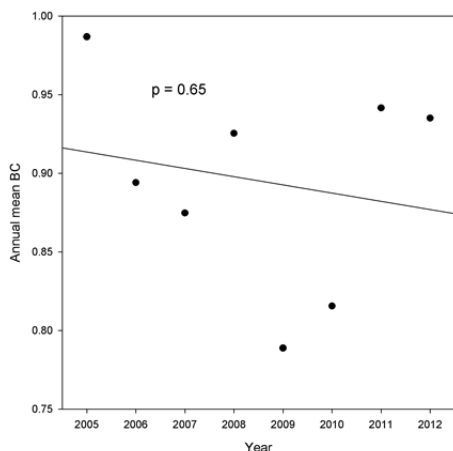
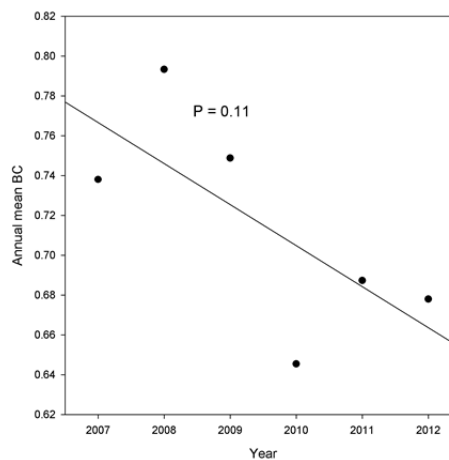
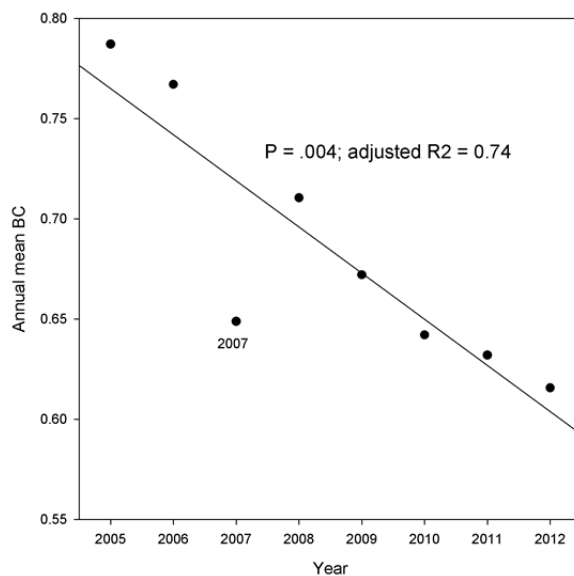


Figure ES-10. Regression of Annual Mean BC vs. Year 2007-2012 for Roxbury.



The same trend analysis for the HSPH Brigham Circle BC data showed a robust downward trend over time for 2005-2012 (Figure ES-11). It is not clear why the HSPH site showed a stronger trend for 2005-2012 than the Roxbury site. Both sites were urban scale over the 2005 to 2012 period, and had a similar mean BC ($0.70 \mu\text{g}/\text{m}^3$ for Roxbury and $0.68 \mu\text{g}/\text{m}^3$ for HSPH).

Figure ES-11. HSPH Trend 2005-2012.



ES-4. Future work.

BC will continue to be monitored at four existing Boston sites, allowing assessment of future trends. The new MassDEP near-road monitoring site on the inbound Southeast Expressway, two kilometers east of the Roxbury site, includes BC measurements, for a total of five Boston BC monitoring sites. As mobile source BC emissions continue to be reduced, similar to the dramatic reduction of carbon monoxide over the last two decades, the utility of BC as a mobile source marker is also likely to decrease. While this is desirable from a health and exposure perspective, there are other mobile source pollutants of concern that may not be reduced, and it may be difficult to find another easily measured marker of mobile sources.

1. INTRODUCTION

This report presents an analysis of spatial and temporal trends of black carbon (BC) in Boston, Massachusetts. BC is an optical measurement of how dark an aerosol is, and has been shown to be well correlated with elemental carbon (EC) and coefficient of haze (COH) measurements (Allen et al., 1999). It is a useful indicator of local mobile source aerosol emissions in urban areas (Janssen et al., 1997). BC measurements were made at three long-term sites in Boston; these data were used to assess temporal trends and the impact of various diesel PM control strategies. Additional BC measurements made in 2003 and from 2009 through 2012 were assessed to evaluate spatial patterns across the metropolitan Boston area.

1.1. Sources and Health Effects of BC

Local mobile sources in large urban areas contribute to elevated levels of a wide range of air pollutants, including particulate matter (PM) from automotive (spark ignition) and diesel vehicles. PM from local mobile sources can be highly toxic, and is considered a major factor in the observed PM health effects reported by a wide range of epidemiological studies over the last decade (von Klot et al., 2011; Janssen et al., 2011). More recently, BC has been cited as a factor in near-road health effects (Brunekreef et al., 2009; Vette et al., 2013; Knibbs and Morawska, 2012).

From a policy perspective, an improved understanding of the spatial patterns (gradients) and long-term temporal trends of mobile source-related PM in large urban areas, as represented by BC, can inform control strategy assessment and implementation. It can also aid in understanding exposure dynamics of potential environmental justice-related hot-spots, such as the Dudley Square area of Roxbury (Boston). Moreover, it is critical for better understanding and improving estimates of exposures used in health effect studies. For example, Harvard University's EPA Boston PM Center has used the Massachusetts Department of Environmental Protection's (MassDEP's) air pollution data in several studies over the last decade.

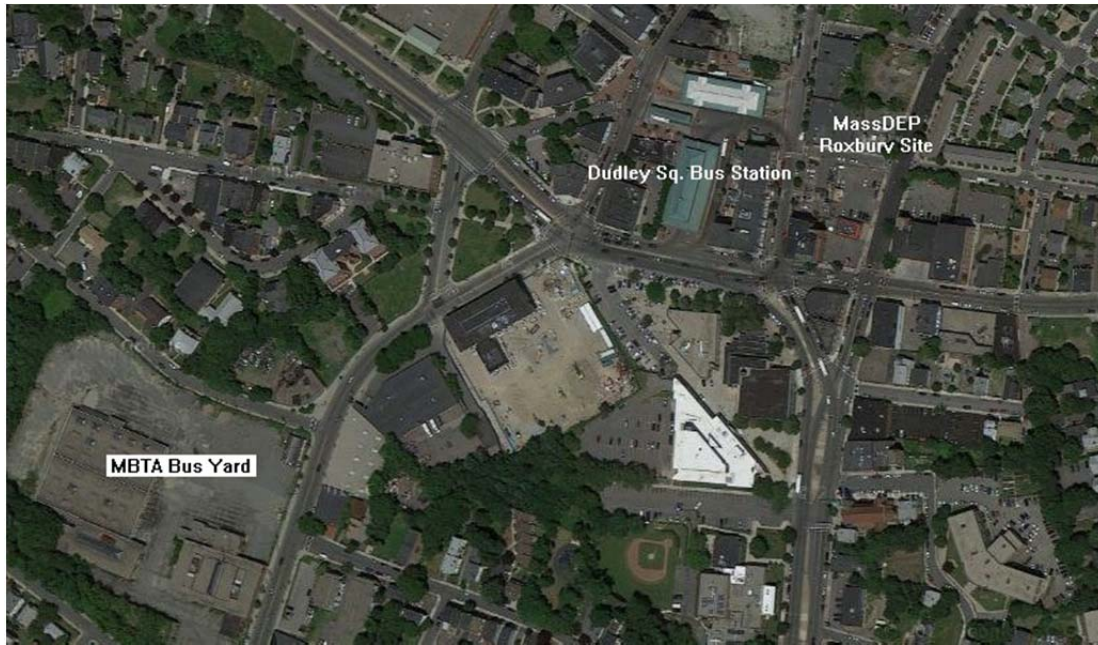
When data from a single monitoring site are used to represent concentrations of locally generated pollutants such as BC for a metropolitan area, exposure misclassification may occur since variation in pollutant concentrations can be substantial over the urban area. Unless the spatial component of exposures is taken into account, this error can bias estimates of health effects toward lower (less hazardous) values (Kunzli et al., 2005). This project analyzed BC in the metropolitan Boston area, with the goal of better understanding urban gradients and temporal trends of BC (i.e., locally generated mobile source aerosols, including diesel PM) over the last decade. Quantifying BC spatial gradients and temporal trends are related tasks, as both affect characterization of mobile-source aerosol exposures.

1.2. Previous Related Work in Boston.

Since 1999, BC measurements in Boston have been collected at two sites: (1) in Roxbury at the intersection of Harrison Ave. and Ziegler St. near Dudley Square by MassDEP (February 1999); and (2) near Brigham Circle on the roof of the Harvard University Countway Library by the Harvard School of Public Health (HSPH) (October 1999). When Roxbury measurement efforts started, the Dudley Square area was considered a hot-spot for diesel pollution from the Dudley Square Bus Station and the nearby Massachusetts Bay Transit Authority (MBTA) Bartlett Street bus yard and garage. At that time, the MBTA bus fleet was old. Much of the fleet

dated back to the mid- and late-1980's, and included some two-stroke diesel engines. Buses had no emission controls, and thus contributed to high levels of pollution, including soot. Figure 1-1 shows an aerial view of Roxbury, with the bus yard, the Dudley Square Bus Station, and the MassDEP Roxbury monitoring site labeled.

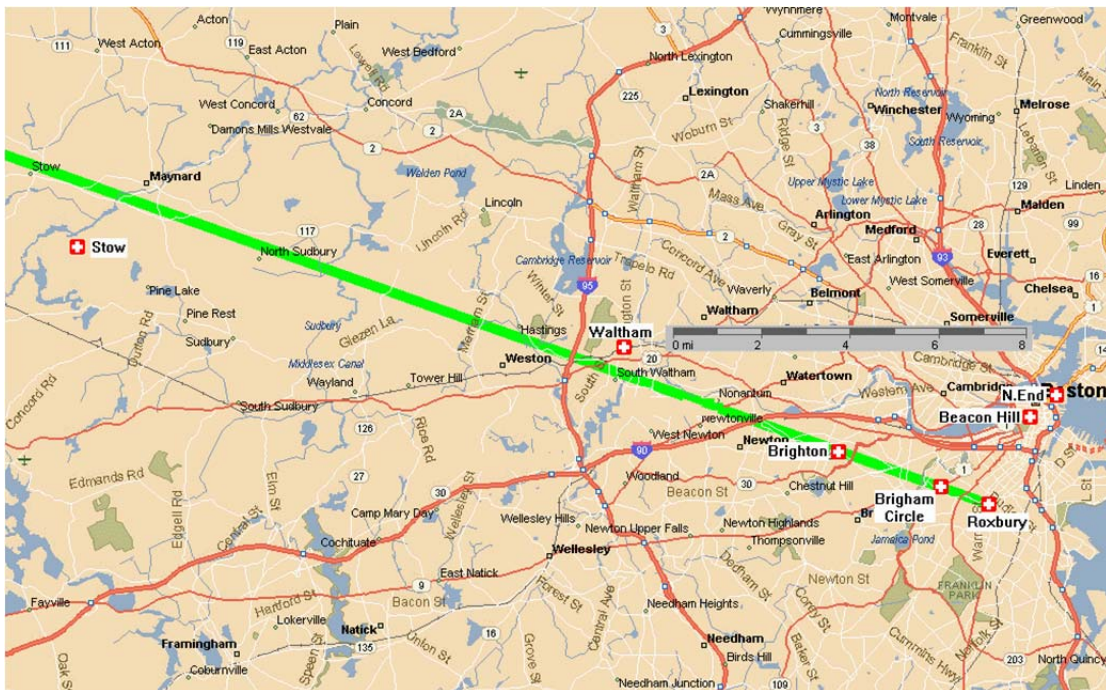
Figure 1-1. Aerial View of Roxbury.



Beginning in 1999, local environmental justice (EJ) groups worked with MassDEP to install a comprehensive monitoring site one block from Dudley Square. The EJ groups, MassDEP, the Harvard School of Public Health, and NESCAUM received funding from the U.S. EPA for an EMPACT project (Environmental Monitoring for Public Access and Community Tracking) to develop outreach tools for the community (Loh, 2002). The resulting AIRBEAT project provided the public access to real-time ozone and PM data through a web site and telephone hotline. The AIRBEAT web site, which is hosted by NESCAUM, remains operational as of January 2014, and the project was chosen by EPA as a technology-transfer case study.²

In December 2002, NESCAUM started a one-year study to investigate the spatial extent of elevated BC from traffic, using the two existing monitoring sites and four new sites. Their locations spanned from downtown Boston to the town of Stow, which is just inside the I-495 loop. Sites were located to be representative of neighborhood-scale concentrations and avoid influence from heavily trafficked roads. Figure 1-2 shows the site locations of these six year-long sites plus the MassDEP North End site that started July 2003.

² United States EPA. Planning and Implementing a Real-time Air Pollution Monitoring and Outreach Program for Your Community The AirBeat Project of Roxbury, Massachusetts. 2002. Accessed May 21, 2013. <http://airbeat.org/airbeat-tech-xfer-final.pdf>.

Figure 1-2. Map of Seven Year-long Sites.

A related pilot study that included three additional monitoring sites ran for six weeks, from January 11 to February 26, 2003. Subsequently, during the summer of 2003, six additional sites were added in order to look at gradients within the Boston urban core, resulting in a total of ten urban core sites. Previous preliminary analyses for these efforts, conducted prior to this assessment, are included in Appendix A and B.

1.3. Limitations of Previous Work

There were several limitations to the previous preliminary analysis. The scope of the analysis was limited due to resource constraints. The lack of long-term data did not allow for proper seasonal analysis. The last four months of data were not analyzed, and the North End site was not included. Moreover, a substantial artifact to the BC measurement method was not accounted for.

The BC artifact produces a reduced instrument response at higher filter particle loadings. This error can vary from none to a factor of two for some hours. The error varies with filter loading, instrument configuration, and the aerosol composition, which may have strong seasonal and spatial components (Virkkula et al., 2007; Park et al., 2010; Turner et al., 2007; Coen et al., 2010). In 2003, this artifact was not understood. NESCAUM authored a report for MassDEP on Aethalometer BC artifacts in 2007, entitled “Evaluation of the Aethalometer BC Spot Matrix Effect” that characterized different aspects of BC measurement errors.³ This artifact has been accounted for in the following assessment.

³ <http://www.nescaum.org/documents/madep-aeth-spot-effect-12dec2007.pdf/view>

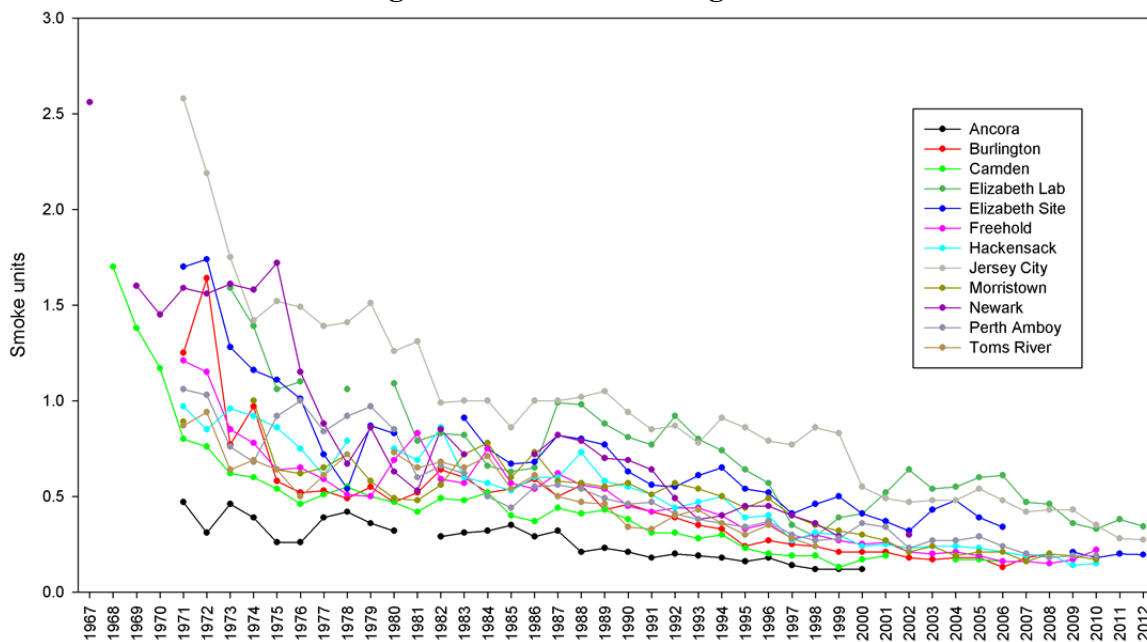
2. APPROACH AND METHODS

Data sources for this project included BC monitors at on-going MassDEP sites and at the HSPH’s EPA PM-center, as well as BC measurements at sites that were run specifically for this project.

2.1. Aethalometer Method

The primary metric used for this analysis was BC from Aethalometers™ (Magee Scientific, Berkeley, CA), a measurement that is commonly used by the EPA National Air Toxics Trends Stations (NATTS) program. The Aethalometer measures how dark the aerosol is: the more sampled graphitic carbon soot (which comes primarily from diesel in urban areas as well as from spark ignition vehicles), the higher the reported BC concentration. Aethalometer BC has been shown to be highly correlated with thermal elemental carbon (EC) methods, such as those used in EPA’s Chemical Speciation Network (CSN), and with the classic smoke shade coefficient of haze (COH) measurement that has been in use since the 1960's (Allen et al., 1999). One COH unit is approximately 5 micrograms per cubic meter ($\mu\text{g}/\text{m}^3$) BC. The New Jersey Department of Environmental Protection (NJDEP) has been measuring COH at multiple monitoring sites since 1967. Figure 2-1 shows COH trend data for New Jersey from 1967 through 2012, which demonstrates that, over the last several decades, there has been a dramatic reduction of COH (or BC), reflecting the cleanup of the on-road vehicle fleet (Kirchstetter et al., 2008).

Figure 2-1. NJ COH Long-term Trend.



2.2. Data Processing and Analysis

For the spatial BC analysis, annual means were used as the primary metric. Correlations of hourly concentration across sites were done for specific short-term periods of a few days.

Annual mean BC was also used for the temporal trend analysis. The monthly pattern of BC was examined for sites used in the trend analysis.

Data from the Roxbury and HSPH sites required evaluation of changes in instrument configuration and data handling over the 13 years of BC data. Most of these changes were taken into account by the data reprocessing technique that minimizes the filter spot loading artifact noted above. The reprocessing requires the original instrument data. For Roxbury, much of these original data were not available between 1999 and 2002, so BC data from the MassDEP data acquisition system (DAS) were used. There were periods in 1999 and 2000 when the original instrument data files were available; these were used to determine the relationship between the re-processed BC data and the MassDEP DAS data. Based on this analysis, a correction factor of 1.2 was applied to the DAS data, consistent with the bias expected from uncorrected BC data.

Instrument bias, the potential difference between two different Aethalometers even when operating properly (Müller et al., 2011), is another factor to be considered in trend analysis. At the North End site, the same instrument was used over the entire time-period used in this analysis (Springfield also used the same instrument) other than brief periods for repairs, removing this factor for trend analysis. The HSPH/Countway site used two different instruments over the 13 years; the change of instrument was evaluated, and there was minimal effect on trend data. The Roxbury site used several different instruments during the October 2004 to August 2007 period, and there was no evaluation (e.g., collocation) of possible effects of these changes on the reported BC data. These changes could introduce artifacts in the BC trend at this site. However, the Roxbury Aethalometer was not changed from the start of monitoring in 1999 to October 2004 (the period with a large decrease in BC concentrations) and since September 2007 other than for brief periods for repair. This and the observed decreases in BC at the HSPH and North End sites during 2003-2004 provides confidence that the BC trend during this period is not an artifact from using different instruments at the Roxbury site. Table 2-1 shows the dates and instrument serial numbers for the Aethalometers used for Roxbury BC measurements.

Table 2-1. Roxbury Aethalometer History (serial numbers)

Date	Instrument serial number
4/1/99-10/1/04	sn199
11/5/04-3/24/05	sn456
3/24/05-4/11/05	sn367
4/24/05-8/3/05	sn456
9/9/05-10/24/05	sn199
10/24/05-8/9/06	sn641
8/9/06-8/27/07	sn684
8/27/07-10/16/07	sn801
10/16/07-11/21/07	sn766
11/21/07-9/4/08	sn801
9/4/08-9/16/08	sn380
9/16/08-4/14/11	sn801
4/14/11-5/25/11	sn199
5/25/11-12/31/13	sn801

Another instrument factor that was controlled for was the “Mean Ratio” (MR) setting. Data from the MassDEP North End and Springfield sites used an MR value of 1.00 for the first several years, as that is how the instruments were originally configured by the Aethalometer manufacturer. Subsequently, the manufacturer determined that the MR value was approximately 0.85. This change was implemented in the instrument configuration in November 2009 for the North End site, and in March 2011 for the Springfield site. Data in this report before this change have thus been corrected by a factor of 0.85. The official MassDEP BC data do not reflect this change.

Similarly, the official HSPH data from the Countway Library site have not been corrected for the spot loading artifact. The data presented here were corrected, and therefore differ from the HSPH data. Appendices C and D provide examples of the differences between the reprocessed and original BC data sets for the HSPH and North End sites.

The original 2003 spatial analysis did not include the North End BC measurements, which started on July 1, 2003. For the re-analysis, reflectance measurements were performed on Federal Reference Method (FRM) sampler Teflon filters from the site to fill in BC data for the first half of 2003 (Heal and Quincy, 2012).

The reflectance data resulted in a mean BC concentration of $1.35 \mu\text{g}/\text{m}^3$ for February through April 2003. The mean Aethalometer BC concentration for July through December 2003 was $1.32 \mu\text{g}/\text{m}^3$, which was used as the annual mean for the North End site. Originally, we expected to be able to fill in BC data back to 2001 using this method, but the FRM filters for 2001-2002 could not be located. Additional details on the reflectance method are in Appendix E.

There are several sites in Boston for which there are many years of BC measurements. Roxbury started in 1999, and HSPH in 2000. BC measurements were added to the MassDEP’s North End monitoring site in July 2003. As part of this project, BC measurements were started at NESCAUM’s South Street (South St.), sixth-floor office (overlooking Tufts Street) in May 2009. Note that this is not the same South St. location used in the 2003 spatial summer intensive analysis. This report’s trend analysis was based primarily on annual mean BC concentrations (with reflectance data fill in for the North End 2003 BC mean).

3. RE-ANALYSIS OF 2003 SPATIAL BC DATA.

Since the initial analysis in 2003, substantial effort has been put into improving data post-processing techniques to reduce the Aethalometer data artifact. This has resulted in a “binned” correction method. This correction method is described in a presentation from the 2012 National Air Monitoring Conference.⁴

All BC data used for this project were corrected for filter loading errors with this method in a consistent manner. Additional details on the binned correction method and examples of its effect are in Appendix D and Turner 2011, Appendix F.

3.1. Seven Site One-Year Re-Analysis.

For this study, the preliminary analysis conducted in 2003 was repeated, using a full year of corrected BC data from the six original sites and with data from the MassDEP’s North End site.

The Stow background site has data from December 2002 through mid-September 2003, and the North End site started July 1, 2003. The other sites have a full year of data, with reasonable data capture. Table 3-1 lists the seven sites, their distance from the Joy Street site, and a description of the land use.

Table 3-1. Description of Seven 2003 Year-long Sites.

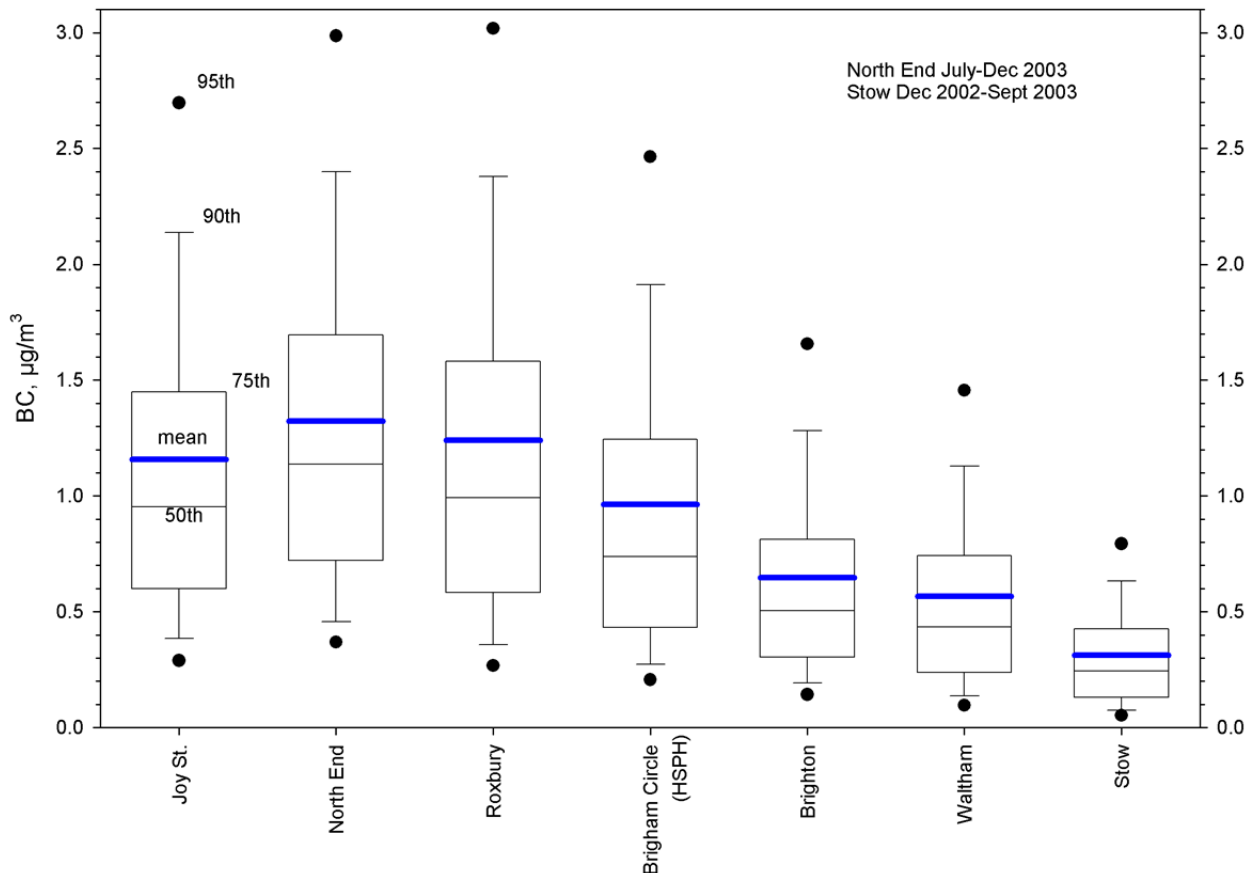
7 Core Site Locations	Km	Site Description
Joy Street (Boston)	0.0	Urban Residential (near State House)
North End (Boston)	1.1	Urban Residential/Commercial
Roxbury (Boston)	3.5	Urban Residential/Commercial; Environmental Justice
HSPH (Boston)	4.1	Urban Residential/Commercial (urban scale)
Brighton (Boston)	7.0	Semi-Urban Residential
Waltham	14.9	Suburban Residential/Light Commercial
Stow	35.3	Semi-rural, Open Land; (Regional Background site for Metro Boston)

Figure 3-1 shows the distribution of hourly BC for the study period for each site. The four urban sites (Joy Street, North End, Roxbury, and HSPH-Brigham Circle) were all higher than the three suburban and background sites. The North End and Roxbury sites were similar.

⁴ Allen, George, and J. Turner. “Aethalometer Data Post Processor (‘Masher’) Update: Spot Loading Correction.” Presentation, National Air Monitoring Conference, Denver, CO, May 16, 2012. Accessed May 21, 2013, <http://www.epa.gov/ttnamti1/files/2012conference/3C01Allen.pdf>.

As expected, the Stow background site was the lowest, with mean BC about one-fourth of the North End and Roxbury sites.

Figure 3-1. Distribution of 2003 Hourly BC for Seven Core Sites.



Note that tests for significant differences in mean BC across sites were not performed, since the very large sample size typically shows all means to be different at $p = 0.05$.⁵ This result is misleading for these data since the between instrument bias for the Aethalometer is typically between 10 and 20%. Thus, any difference less than approximately 15 to 20% could be an artifact of the measurement.

Figure 3-2 shows the diurnal pattern for the seven sites, with work-days plotted separately from weekend days and holidays. For the weekday plot, Roxbury and the North End site had the highest morning rush-hour BC concentrations; Joy Street and HSPH were somewhat lower. The two non-urban core sites, Brighton and Waltham, showed a smaller morning rush hour peak relative to the rest of the day. Stow, the background site, showed no strong BC pattern for the entire day, consistent with its semi-rural location upwind of Boston.

For the non-weekday plot, there was no strong diurnal pattern even for the core urban sites. This is consistent with the expected different non-weekday traffic patterns. The North End site showed a daily maximum in the early evening from 5:00 to 7:00 pm. This multi-season

⁵ Normally ANOVA on ranks followed with an all Pairwise Multiple Comparison Procedure would be used for this purpose.

weekday/non-weekday diurnal analysis provides increased confidence that BC is reasonably specific to local mobile source aerosol, minimizing concerns related to potential interferences at these sites from other sources of BC, such as oil-fired space heating and woodsmoke.

Figure 3-2. 2003 Diurnal Plots of Seven Core Sites.

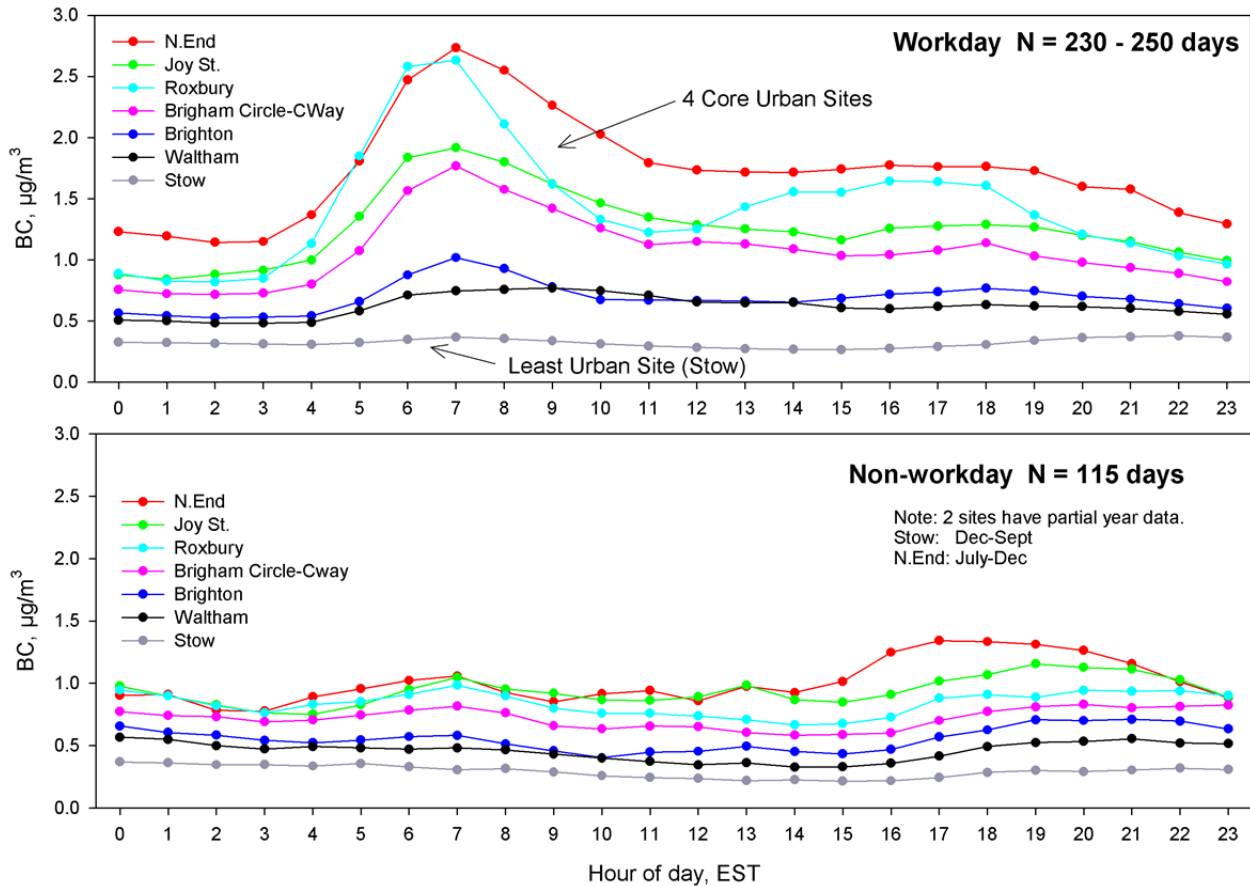
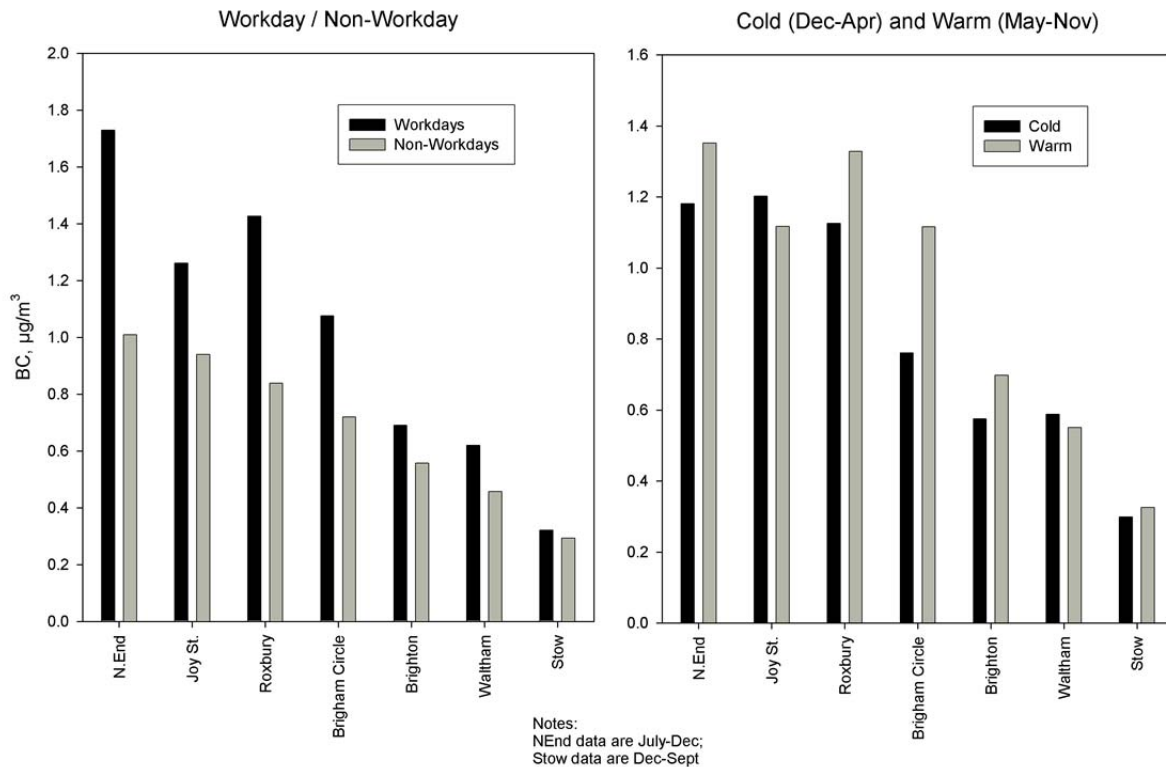


Figure 3-3a shows the average BC for these seven sites on work- and non-workdays. The relative difference decreased as the sites become less urban, with minimal difference for the Stow background site. Figure 3-3b shows the average BC for cold (i.e., December-April) and warm (i.e., May-November) months. There was no clear seasonal pattern. Joy St. was slightly higher than the other urban sites during the winter.

Figure 3-3a and 3-3b. BC for Work/Non-Workdays and Warm/Cold Season.



The long-term trend data, which are discussed in Section 5, Temporal Trends, provide more detail than the simple warm versus cold-season mean BC shown above.

3.2. Twelve Site Summer Study.

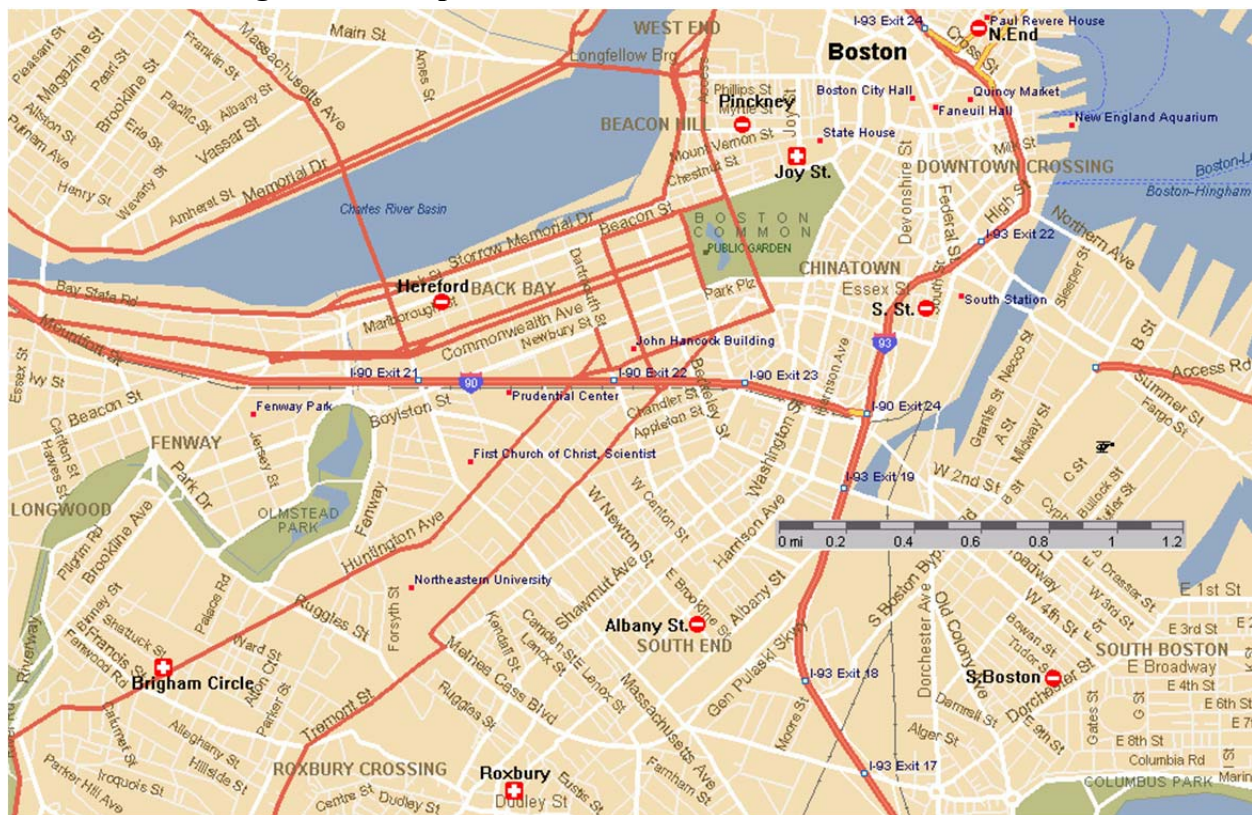
During the summer of 2003, a neighborhood scale study was conducted by NESCAUM in Boston for two months to explore gradients within the urban core. Ten of the twelve sites were in Boston, of which nine were within a radius of 2.5 km. Siting was representative of neighborhood scale (i.e., not hotspot/microscale) exposure. Table 3-2 shows the distances from the State House (Joy Street) for each site. Figure 3-4 shows the locations of the sites; those marked with a “+” are year-long, and “-” indicates summer spatial intensive sites.

Table 3-2. Description of 2003 Summer Intensive Sites.

Site Locations	Km	Site Description
Joy Street	0.0	Urban Residential/Commercial. (Beacon Hill, near State House)
Pinckney Street	0.3	Urban Residential (Beacon Hill)
North End	1.1	Urban Residential/Commercial (near the I-93 Expressway)
South Street	1.0	Urban Commercial (near South Station bus and train terminals)

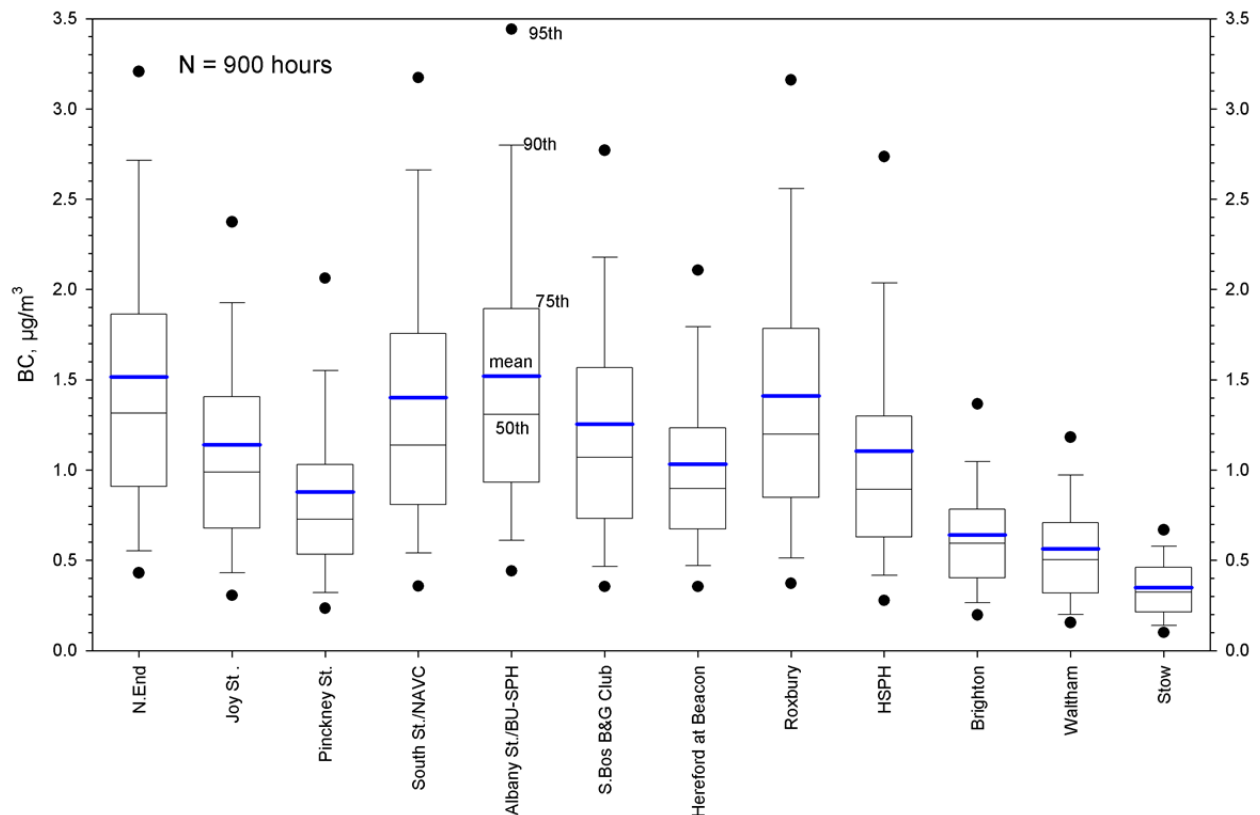
Hereford Street	1.9	Urban Residential (Back Bay)
Albany Street	2.4	Urban Commercial (BU School of Public Health)
South Boston	2.9	Urban Residential
Roxbury	3.5	Urban Residential/Commercial
HSPH	4.0	Urban Residential/Commercial (urban scale)
Brighton	7.0	Semi-Urban Residential

Figure 3-4. Map of 9 of the Boston Summer Intensive Sites.



Sites marked with a square “+” are year-long monitoring sites; those with a circle “-” symbol are sites that ran only during the summer intensive period. The Brighton (Boston) semi-urban site is not shown on this map.

Figure 3-5 shows the distributions for all 12 BC monitoring sites, limited to days where all sites had data. Approximately 20 days were excluded, because two sites had a 10-day period of missing data.

Figure 3-5. BC Distributions of all 12 Sites for 2003 Summer Intensive.

There were substantial gradients for mean BC at neighborhood scale-oriented sites in Boston. The observed variation across sites could be influenced by variability in monitor siting, mobile source strength gradients, and microscale meteorology. The North End and Albany Street sites had the highest gradients, with Roxbury and South Street (near South Station) just slightly lower. The highest and lowest sites, North End and Pinckney Street, respectively, are 1.3 km (0.8 miles) apart, with a ratio of 1.7. The Hereford Street site was relatively low, as it is near both Storrow Drive, where truck traffic is prohibited, and the Charles River.

3.2.1. Population Density and BC

The association between population density and mean BC was explored for the year-long study and the summer intensive study of core Boston sites. Population estimates were generated using LandView5 software, a database application created by the U.S. EPA, Census Bureau, Geological Survey, and NOAA. LandView's population estimator function uses block data from the 2000 U.S. Census, and can generate demographic information for circular areas using block centroids whose coordinates fall within a prescribed radius. Geographic coordinates for the nine Aethalometer monitoring sites were used as center points to estimate population density.

Figures 3-6 and 3-7 show regressions of BC versus population density (1-mile radius) for these two cases. The year-long study showed that BC decreased with population density (Figure 3-6), but the regression was not significant ($p = 0.07$). Removing the Brighton site increased the adjusted R^2 to 0.68 ($p = 0.03$). Brighton may have a high amount of student and multifamily housing relative to the other sites.

The summer 10 urban-site regression shown in Figure 3-7 had the opposite slope from the year-long sites, with $R^2 = 0.68$ ($p = 0.003$). This could be explained by core commercial and transit corridor areas such as the South St. site near South Station having lower population density but high traffic activity.

Figure 3-6. Regression of BC on Population Density for Year-Long.

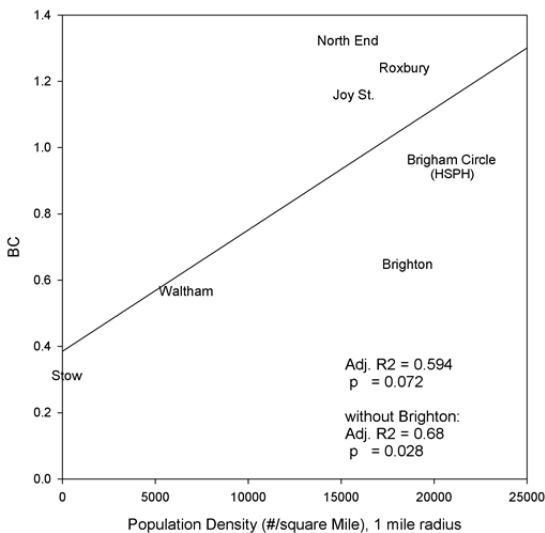
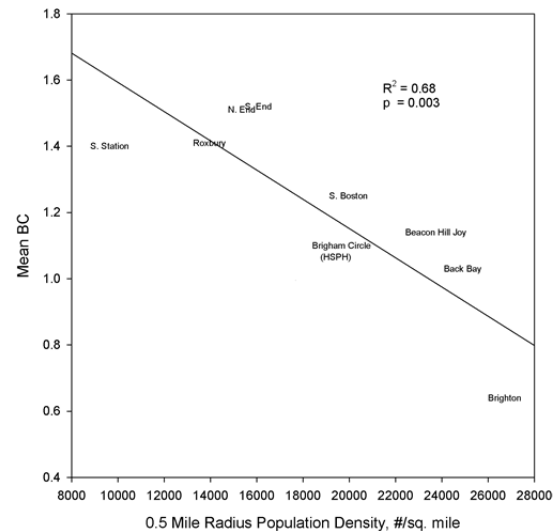


Figure 3-7. Regression of BC on Population Density for Summer Intensive.



3.2.2. Two Time-Series Case Study Examples.

Two time series plots, Figures 3-8 and 3-9, show examples of short term patterns and gradients of hourly BC across the Boston area. Both showed a distinct “clean Sunday and dirty work-day” effect. Figure 3-8 presents data for the nine core Boston sites and the Stow background site during July 13-15, 2003. Tuesday, July 15, 2003 was one of the dirtier days of the summer, with several sites exceeding $4 \mu\text{g}/\text{m}^3$ BC for several morning hours. The ratio of the Boston to the background sites for this peak period was approximately 10, which is similar to that observed during events from the 2003 winter pilot project.

Figure 3-9 presents data for all 12 sites during the period August 6 through 11, 2003. Thursday, August 7 showed a distinct evening rush-hour peak, which is not a common feature. The very high peak in South Boston on Friday, August 8 at 7 a.m. EST was substantially higher than the other sites, although the other urban sites peak at the same hour. The South Boston site may have been influenced by local marine diesel sources, as major Boston Harbor piers are about one mile away to the north-northeast. Winds at Logan Airport were north-northeast to northeast at a few miles per hour during this time. The peak hour was influenced by two contiguous very high 5-minute BC values (22 and $13 \mu\text{g}/\text{m}^3$, not shown). Without those values, the mean for this hour would have been $6 \mu\text{g}/\text{m}^3$, which is more consistent with the other sites.

Figure 3-8. July 13-15, 2003 Time-Series.

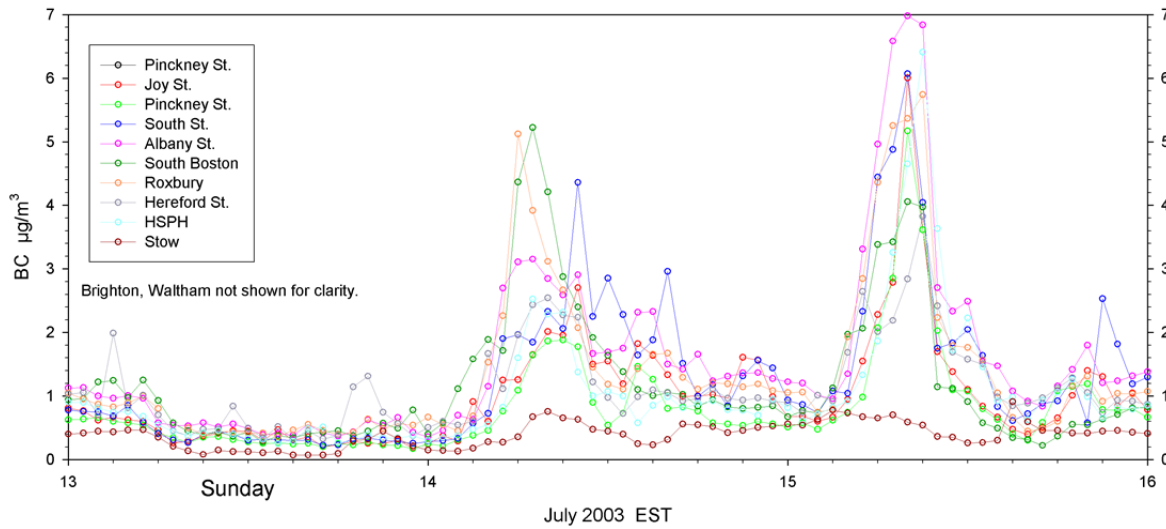
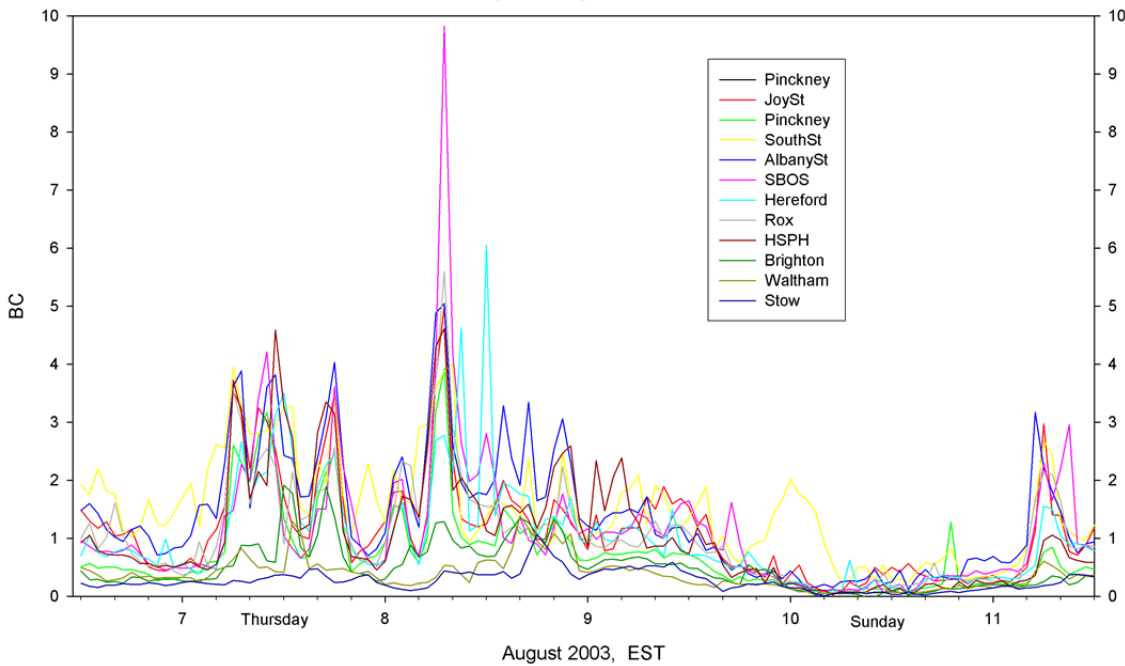


Figure 3-9. August 6-11, 2003 Times-Series.



August 6-11 case study: hourly scatter plots and correlation matrix

Scatter plots for hourly BC for six site-pairs presented in Figure 3-10, and R^2 values for all site pairs during the August 6-day period shown in Table 3-3, are examples of the short-term (hourly) relationships across different spatial scales. There was a wide range of correlation from high ($R^2 = 0.85$ for the two Beacon Hill sites) to very low (0.07 for Stow and South Street). All regressions are significant at $p = 0.05$.

Distance between sites was not always a good predictor of how well they were correlated. Joy Street and Roxbury, 3.5 km apart, had an R^2 of 0.79. The R^2 for Hereford and South Streets, 2.4 km apart, was 0.29 for the same time period. Some sites, such as South and Hereford Streets,

were not well correlated with other urban sites. Others, such as Roxbury, Joy Street, and Albany Street, seemed to be reasonably well correlated with most urban sites.

The scatter plots show interesting patterns for some of the site-pairs. The two downtown Boston sites with the lowest mean BC, Hereford and Pinckney Streets, were well correlated when levels were below about $1 \mu\text{g}/\text{m}^3$ BC. When levels were high at either site, however, they tended to be temporally decoupled. The South Boston and South Street sites were clearly influenced by different sources. North End and Stow, the highest and lowest sites in the study, were largely decoupled at this time scale. Note that the scatter plot axes are not scaled the same, and the bottom line is the 1:1 line. As would be expected, essentially all hours at North End were at or above the Stow BC levels.

Figure 3-10. Scatterplot for Hourly BC for Six Site-pairs During August 2003 Event Period.

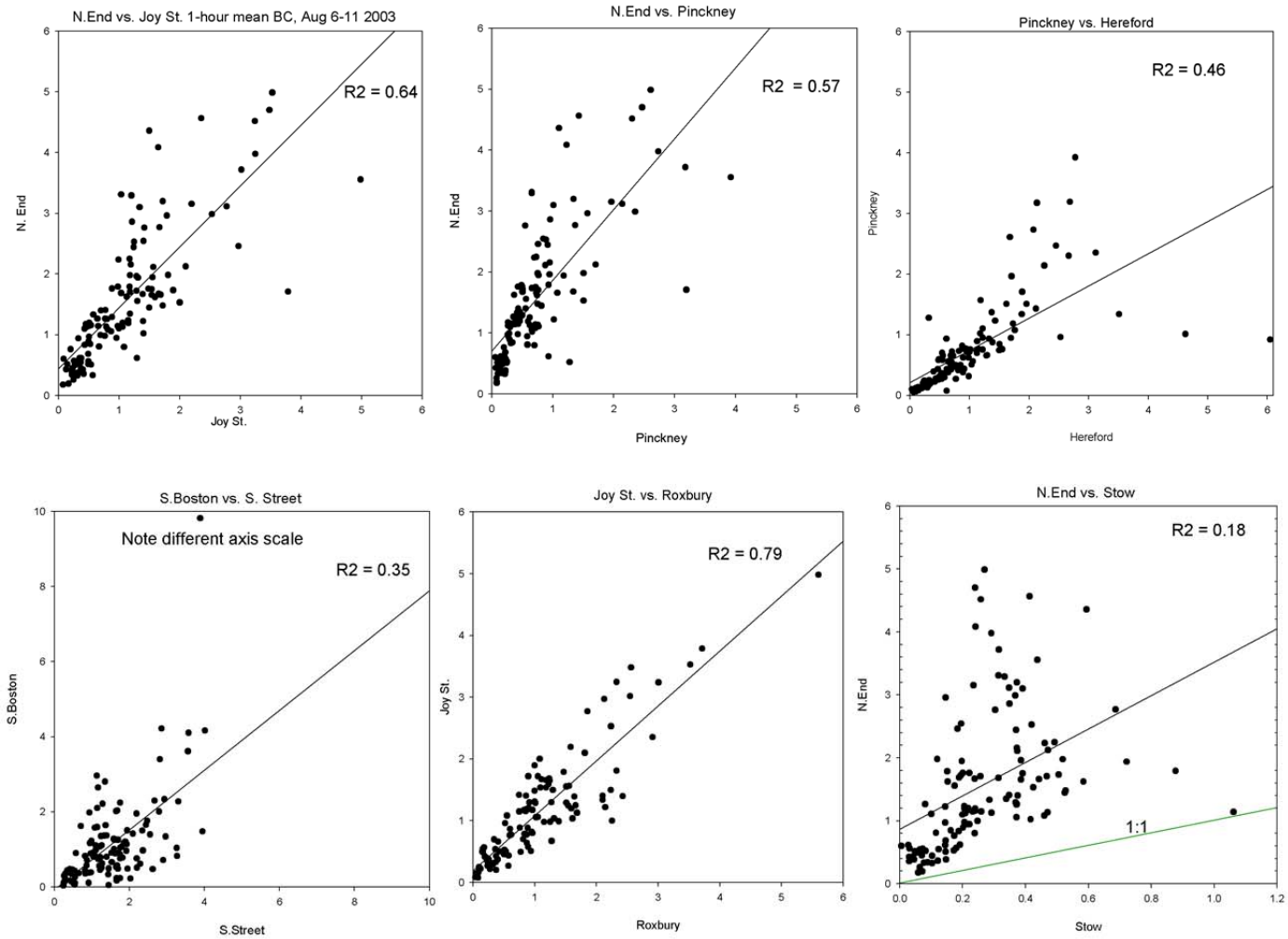


Table 3-3. One-Hour R² Matrix, August 6-11, 2003

(R² **0.70 or higher** R² **0.50 to 0.69**)

	North End	Joy St.	Pinckney St.	South St.	Albany St.	South Boston	Hereford St.	Roxbury	HSPH-Brigham Circle	Brighton	Waltham	Stow
North End	X	0.64	0.57	0.53	0.58	0.43	0.46	0.59	0.54	0.44	0.33	0.18
Joy	0.64	x	0.85	0.59	0.77	0.66	0.43	0.79	0.67	0.41	0.21	0.14
Pinckney	0.57	0.85	X	0.54	0.77	0.63	0.46	0.73	0.73	0.46	0.19	0.13
South St.	0.53	0.59	0.54	x	0.57	0.35	0.29	0.57	0.52	0.34	0.20	0.07
Albany	0.58	0.77	0.77	0.57	x	0.53	0.49	0.77	0.76	0.55	0.36	0.19
S. Boston	0.43	0.66	0.63	0.35	0.53	x	0.30	0.57	0.44	0.24	0.09	0.10
Hereford	0.46	0.43	0.46	0.29	0.49	0.30	x	0.45	0.52	0.48	0.33	0.18
Roxbury	0.59	0.79	0.73	0.57	0.77	0.57	0.45	x	0.69	0.40	0.23	0.12
Brig. Cir.	0.54	0.67	0.73	0.52	0.76	0.44	0.52	0.69	x	0.62	0.29	0.21
Brighton	0.44	0.41	0.46	0.34	0.55	0.24	0.48	0.40	0.62	x	0.51	0.39
Waltham	0.33	0.21	0.19	0.20	0.36	0.09	0.33	0.23	0.29	0.51	x	0.45
Stow	0.18	0.14	0.13	0.07	0.19	0.10	0.18	0.12	0.21	0.39	0.45	x

4. UPDATED BC SPATIAL ANALYSIS, 2009-2012

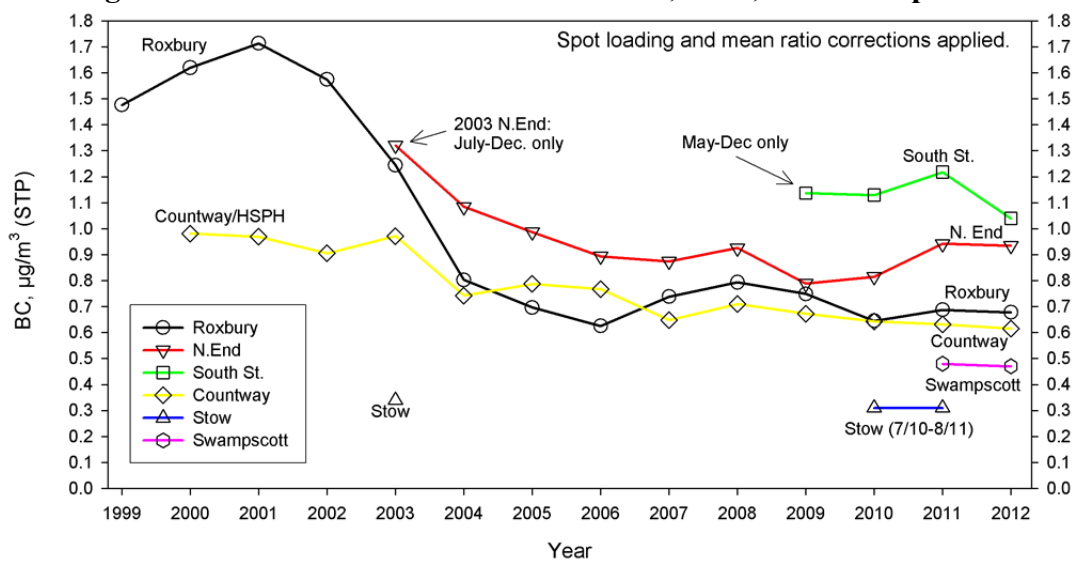
In retrospect, the 2003 spatial study was conducted during a year when substantial reductions in heavy-duty diesel PM emissions were occurring from changes to both the Boston public transit system (MBTA) and the Boston school bus fleets. The changes in BC concentrations and details on the bus fleet cleanups that continued through spring 2005 are discussed in Section 5, Temporal Trends.

Because these changes may have affected spatial patterns of BC in Boston, additional BC monitoring sites were set up and run for this project by NESCAUM between 2009 and 2012. This was done in order to provide a limited assessment of the more recent spatial scale of Boston BC. The Stow regional background site was run for two years (July 2009 through August 2011), a permanent site at NESCAUM’s South Street offices started in May 2009, and a BC monitor ran in Swampscott (21 km northeast of Boston) from 2011 through 2012. The existing long-term Boston BC sites at North End, Roxbury, and HSPH were also included in this analysis.

4.1. Trends.

Figure 4-1 shows the trend for the four Boston BC sites, along with means for the Stow and Swampscott sites. BC at the Stow regional background site dropped from 0.34 to 0.31 $\mu\text{g}/\text{m}^3$ over seven to eight years. This is consistent with reduced heavy-duty diesel PM emissions on a regional basis. The Swampscott site’s BC concentrations were lower than the Boston and higher than the Stow sites. That Swampscott is downwind of Boston and Stow is upwind may explain the relative BC concentrations at these two non-urban sites.

Figure 4-1. Trend for four Boston BC Sites, Stow, and Swampscott.



In 2010, the ratio of North End to Stow mean BC was 2.6. When comparing Stow with the 2011 North End BC data, the ratio increases to 3.0. This is similar to the ratio observed in 2003.

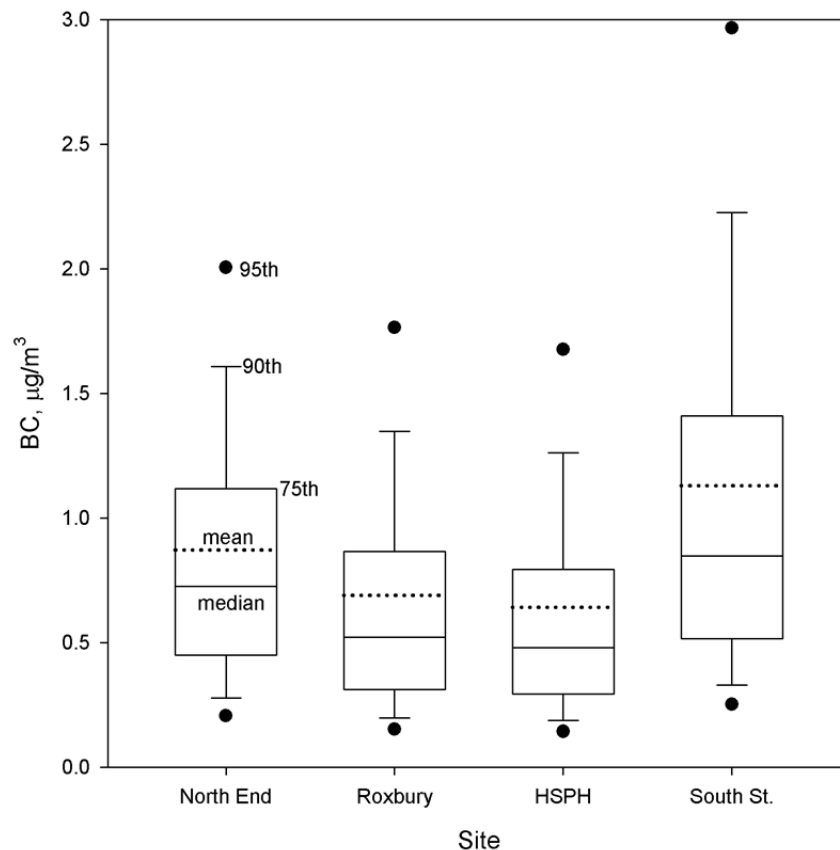
The ratio of the 2010 mean BC between the South St. and Stow sites was 3.6, compared to 3.9 using 2011 data. These comparisons suggest that the gradient from urban to upwind has not changed much over the last seven to eight years.

4.2. Sources of BC at the South St. Site.

The South Street site measured the highest levels of all Boston BC sites between 2009 – 2012, at $1.13 \mu\text{g}/\text{m}^3$ (Figure 4-1). Means for other Boston BC sites ranged from 0.64 at HSPH to $0.87 \mu\text{g}/\text{m}^3$ at North End), while the Springfield site's mean was also $0.87 \mu\text{g}/\text{m}^3$. South Station, a major transportation hub for rail and inter-city buses, is located 300 meters to the southeast of the South St. monitoring location. Pollutant wind rose analysis suggests that the station's bus and train activity is not a major factor in the elevated BC levels, as the wind was not usually from that direction.

Distributions of hourly BC for the four Boston sites from 2009-2012 are shown in Figure 4-2. Roxbury and HSPH distributions are similar, with the North End site somewhat higher. South Street is distinctly higher, with the 95th percentile at $3.0 \mu\text{g}/\text{m}^3$. This is not driven by the Saturday diesel genset test events, because the one-hour per week was only 0.6% of hours.

Figure 4-2. Distribution of 1-hour BC at Four Boston Sites, 2009-2012.



Additional information on sources of BC in downtown Boston is available from source apportionment analysis done for the North End site (Turner, 2008; *see* Appendix G). Although the BC was primarily from mobile sources, a significant amount was from biomass combustion, e.g., woodsmoke from space heating or recreational use.

Figures 4-3 and 4-4 show South Street pollutant roses using wind data from the MassDEP Roxbury site 3.3 km to the southwest. Figure 4-3 shows BC up to $4 \mu\text{g}/\text{m}^3$, which is more than 95% of hours during this period. Note that hours with wind from south to southeast were not common nor associated with these levels of BC relative to other directions.

Figure 4-4 shows BC data for hours greater than $3 \mu\text{g}/\text{m}^3$, including one-hour values up to $19 \mu\text{g}/\text{m}^3$. Winds for hours with BC between 3 and $5 \mu\text{g}/\text{m}^3$ were predominately from the southwest to east. These plots suggest that the elevated BC at this site was from many local sources, indicative of an active area of downtown Boston with respect to heavy-duty diesel vehicles.

Figure 4-3. NESCAUM South St. BC Pollutant Rose, less than $4 \mu\text{g}/\text{m}^3$.

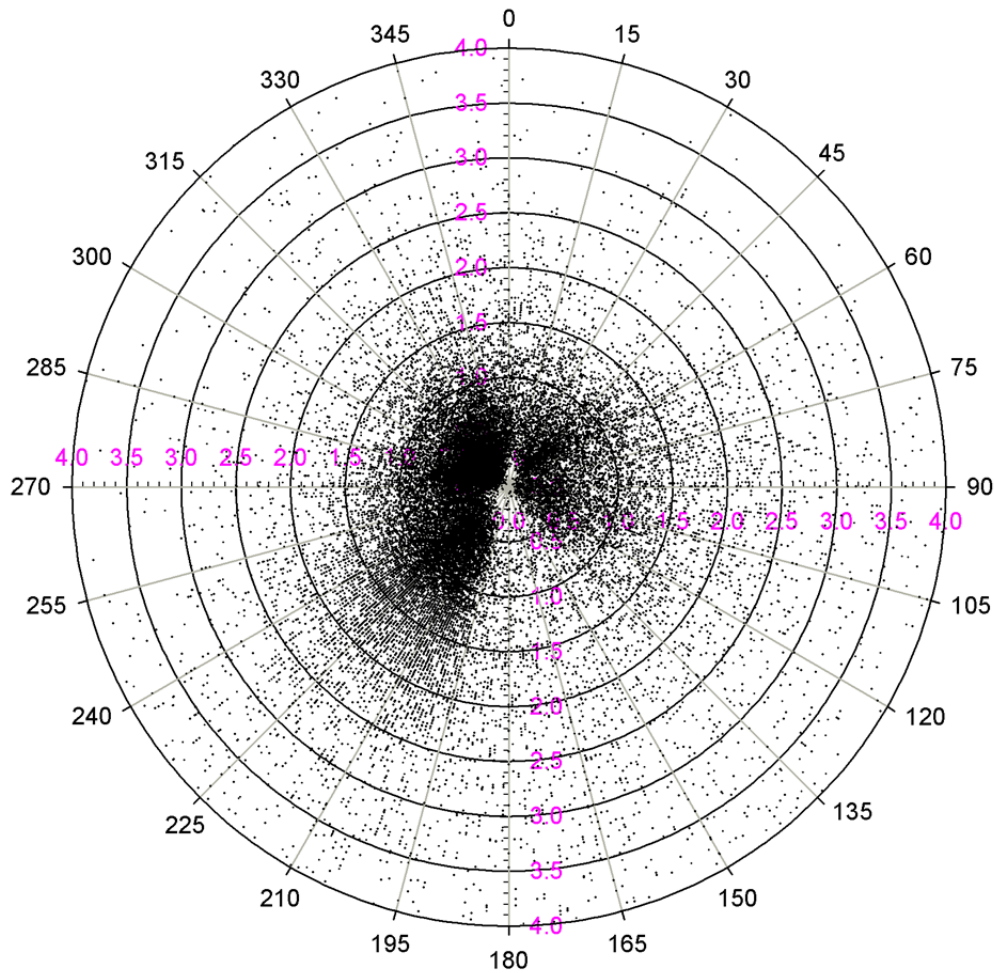


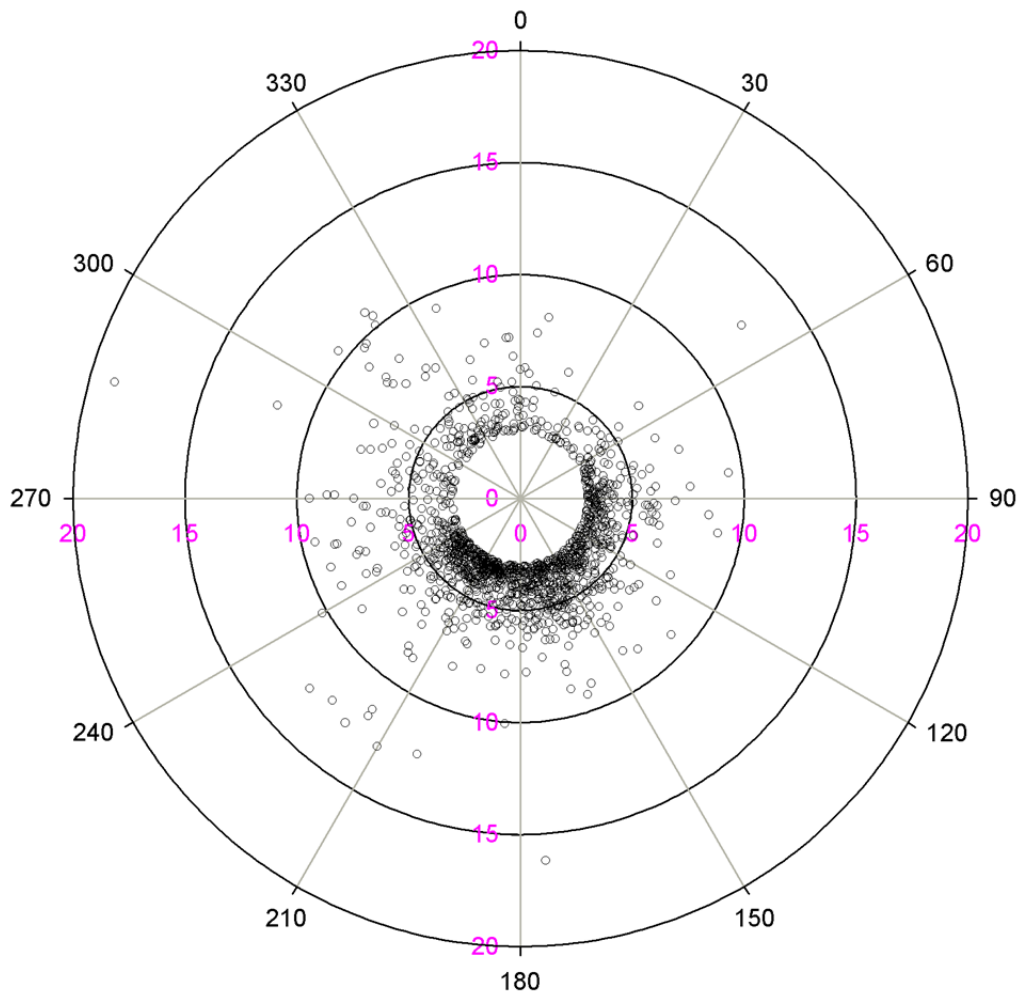
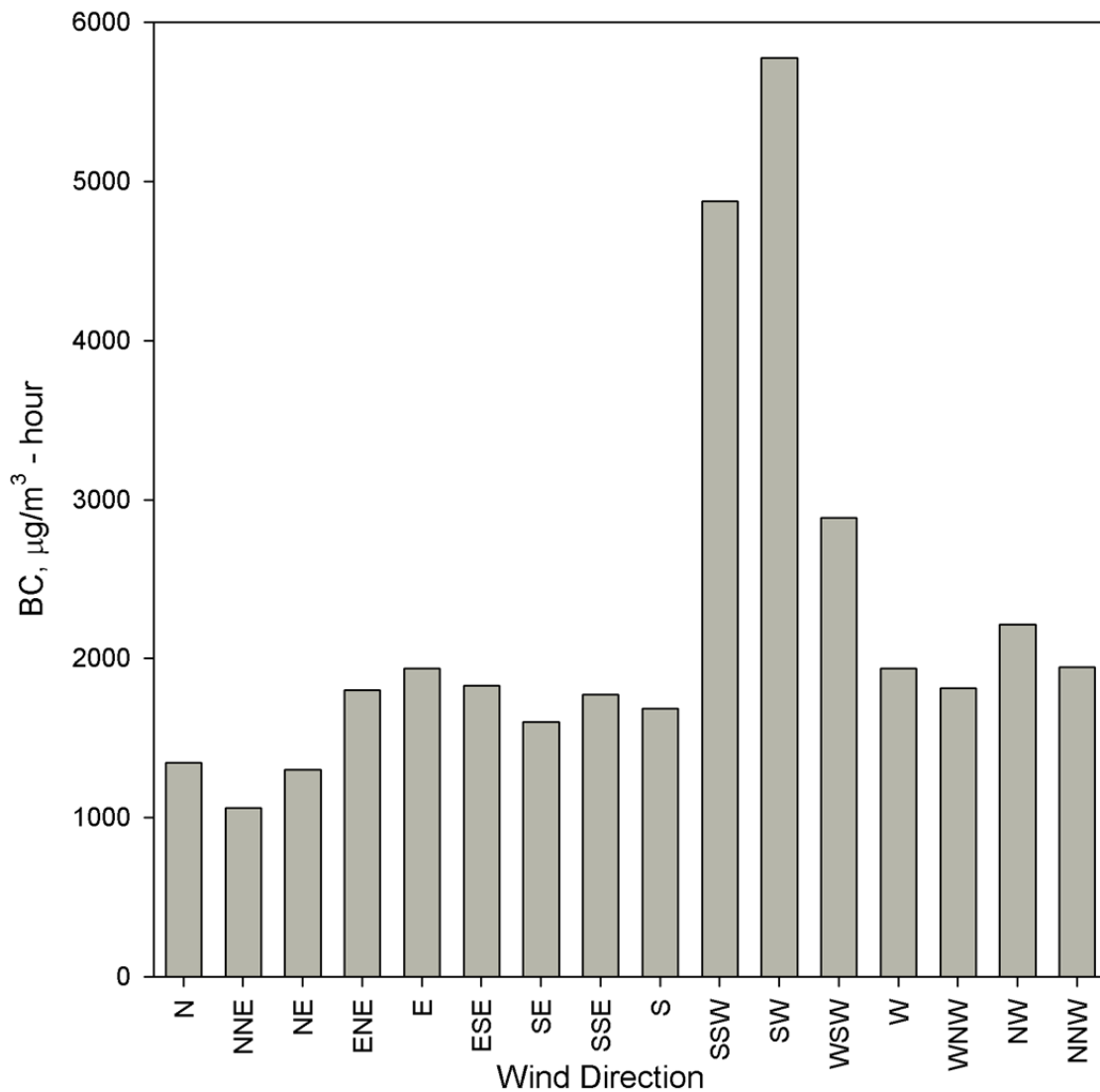
Figure 4-4. NESCAUM South St. BC Pollutant Rose, greater than $3 \mu\text{g}/\text{m}^3$.

Figure 4-6 shows BC concentration times the number of hours, binned by wind direction using Roxbury wind data. This approach is an exposure metric, taking into account both the average concentration from each direction and the number of hours in each wind direction bin. It is very clear that winds from the south-southwest and southwest account for more BC than any other direction, and winds from South Station (southeast) contribute a relatively small amount of BC measured at this site. This is in part due to prevailing winds being from the south to southwest. The map in Figure 3-4 shows that the Massachusetts Turnpike (I-90), the Southeast Expressway (I-93), and the large interchange between the two are all to the south-southwest and southwest of the South Street site within approximately 650 meters. The large amount of traffic activity from this area and the first km of the I-93 Expressway above ground to the south-southwest are likely reasons for the high mobile source-related BC observed at this site. Figure 4-5 more clearly shows this section of the Expressway, the prevailing wind direction (black line), and the NESCAUM South St. site location.

Figure 4-5. Prevailing wind direction alignment with Expressway and the NESCAUM

South St. BC monitoring site (red dot).



Figure 4-6. South St. BC times Hour by Roxbury Wind Direction.

The 2003 study's South Street site, located at the rear of 112 South Street, is 65 meters from the current South Street Site (at 89 South Street). These two sites are close enough to be compared over time. Figure 4-7 shows the summer spatial intensive BC distribution for the 2003 site along with the 2009-2012 BC data from the NESCAUM South Street site matched for the same days of the year. Except for 2011, BC data distributions were very similar to the 2003 data. The contemporary South St. data include the influence from a local source (a diesel genset weekly test) that produced very high BC once a week for approximately 15-20 minutes. Figure 4-8 shows one-minute data from such an event on October 22, 2011 when the peak 1-min BC concentration measured $100 \mu\text{g}/\text{m}^3$. The rapid fluctuations in concentration at this time-scale are indicative of a source that is very close (within ~ 50 meters or less).

Figure 4-7. Summer BC Distribution for South Street.

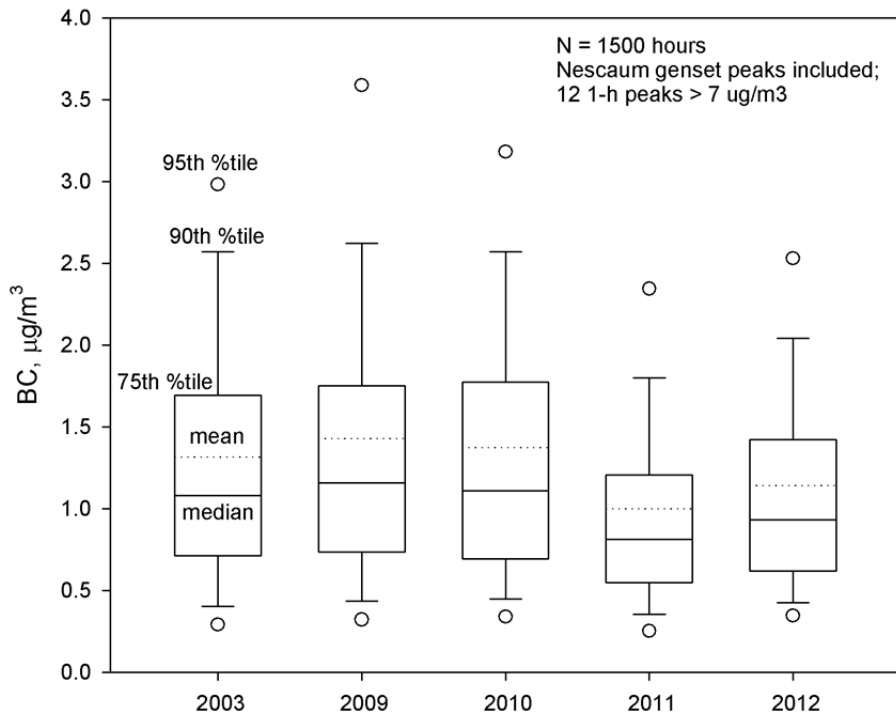
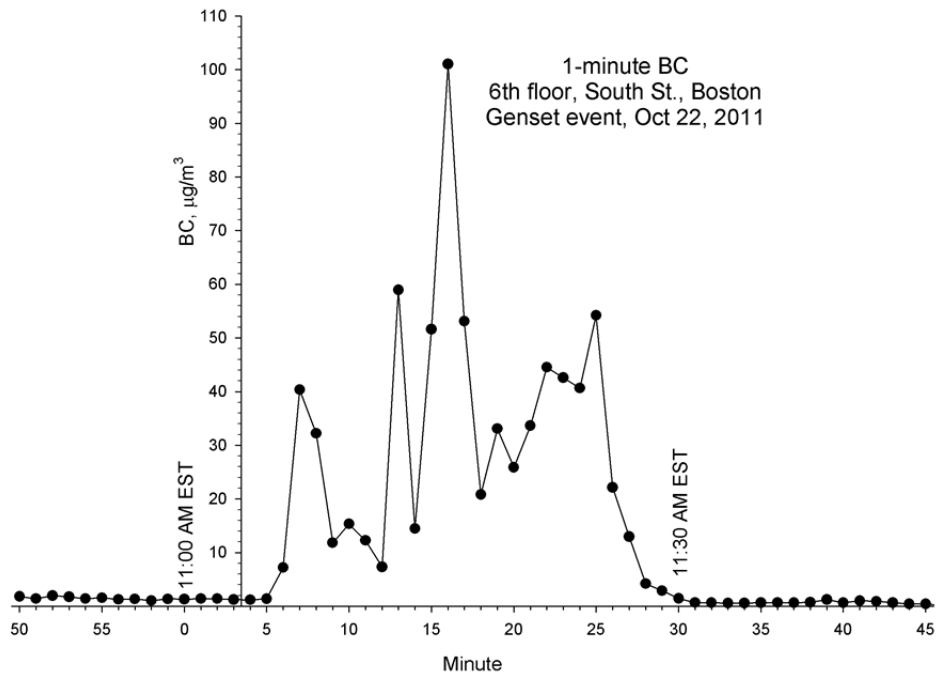


Figure 4-8. October 22, 2011 Diesel Genset Event.

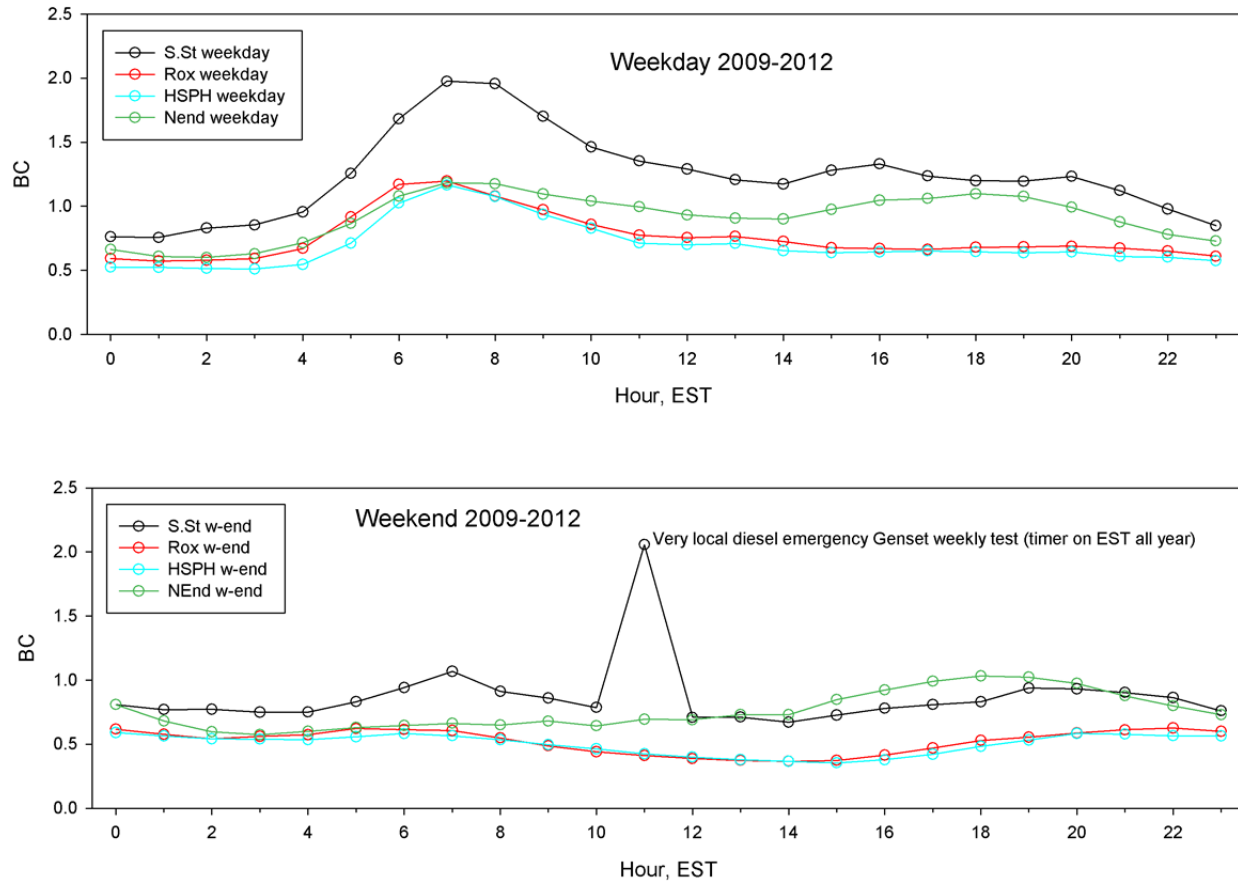


4.3. Diurnal plots for 2009-2012, four Boston sites.

The weekday and weekend diurnal BC plots were updated for data from 2009 through 2012 for three of the Boston sites used in 2003. The South Street site data were also added. These are presented in Figure 4-9. While the general patterns were unchanged relative to 2003,

the Roxbury site behaved more like the HSPH site, with a smoother and less pronounced morning rush-hour peak characteristic of an urban-scale site. The South Street site measured highest concentrations for all hours, during weekdays and weekends. The North End site was measuring higher BC concentrations than the Roxbury or HSPH sites for most hours of the day. As with the 2003 diurnal plots, there was a clear morning rush hour peak on weekdays and no peak on weekends, with the exception of the South Street site. That site also showed the effect of a weekly test of the emergency diesel genset on Saturday at 11 a.m. EST.⁶

Figure 4-9. Four-Site 2009-2012 Updated Diurnal Plots.



4.4. Additional HSPH Monitoring Near Street Level.

Limited additional monitoring was performed near the HSPH Countway Library roof site to assess the effect of elevation on BC concentrations from that site (approximately 26 meters above ground level). From January 6 to May 30, 2010, BC was measured at 635 Huntington Avenue, from the second floor of an office building. This monitor was located on the same block as the HSPH Huntington Avenue site, but closer to Longwood Avenue. Figure 4-10 shows the sample location, which was outside of the second floor window on the left.

⁶ The genset timer is on EST year round, resulting in only hour 11 EST being elevated instead of hours 11 and 12.

Figure 4-10. Location of Street Level HSPH Sampler.

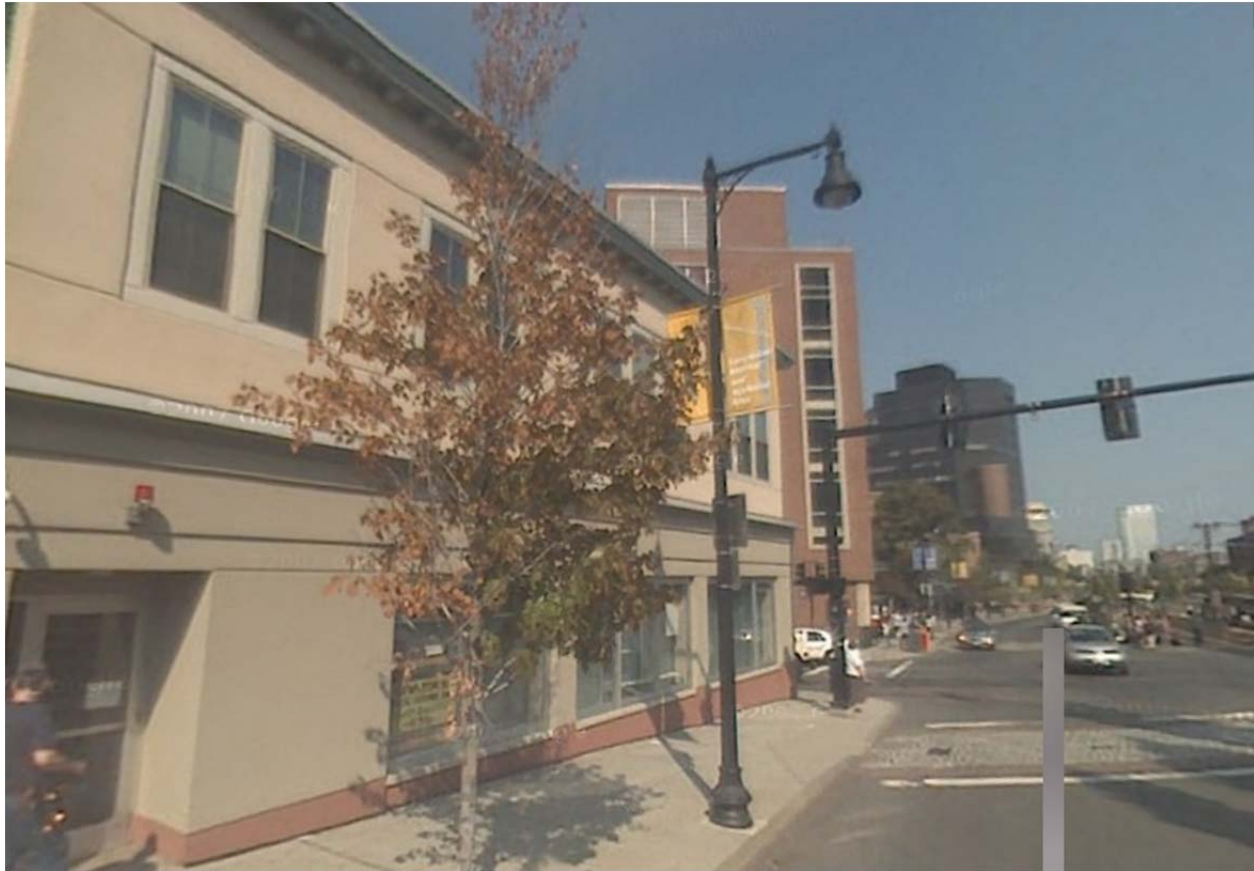
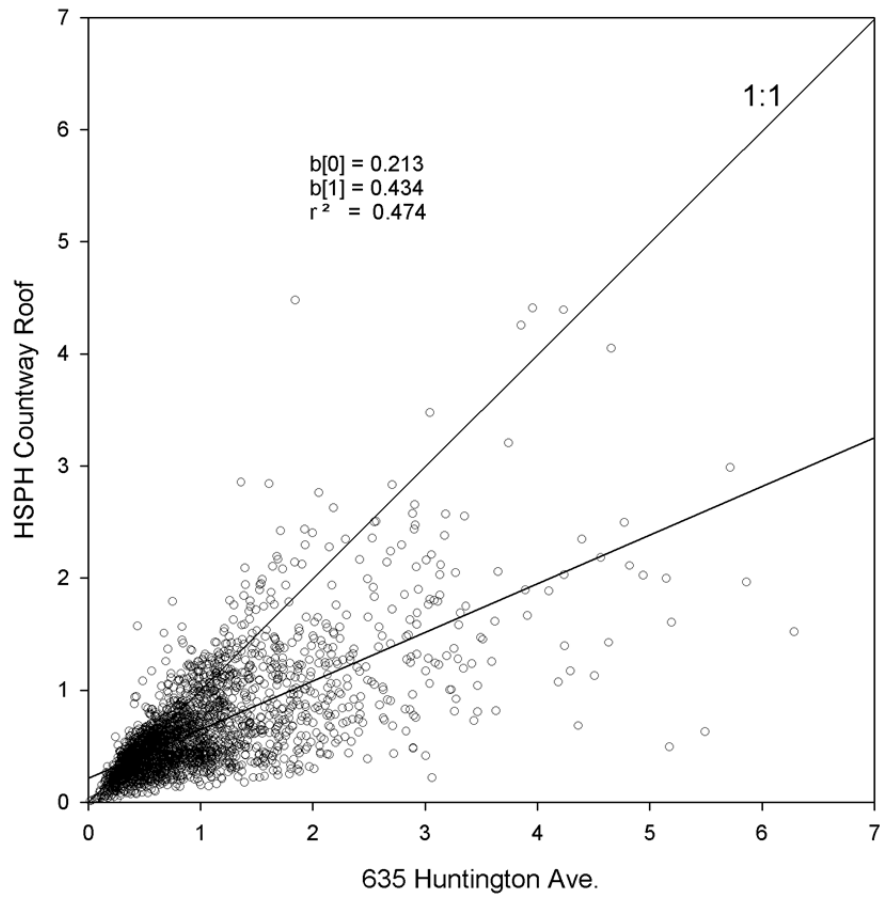


Figure 4-11 shows the relationship of hourly BC between the HSPH and the 635 Huntington Avenue sites. Mean BC was $0.85 \mu\text{g}/\text{m}^3$ for the street-level site and $0.69 \mu\text{g}/\text{m}^3$ for the HSPH site, a ratio of 1.23. The hourly R^2 was 0.47. As expected, BC at the street-level site was generally but not always higher than the HSPH site.

Figure 4-11. HSPH Roof vs. Street Hourly BC.



5. TEMPORAL TRENDS OF BOSTON BC

There are four Boston sites with multiple years of BC data. BC trends have both a strong seasonal pattern but a weak long-term trend at most sites.

5.1. Seasonal Patterns of BC.

There was a strong seasonal pattern for monthly mean BC for the urban sites. BC was distinctly lowest in the late winter or early spring, and highest in the summer for all long-term sites except Springfield. This pattern is consistent with monthly wind speeds being lowest in summer and highest in the winter, as dispersion of local ground level pollutants is expected to be driven by wind speed. Figure 5-1 shows monthly mean BC for the four Boston sites (using all available years of data) with the inverse of monthly average wind speed. The patterns for wind speed and BC were stronger for some sites than others. Seasonal patterns for the South Street and the North End sites were strongest and very similar ($R^2 = 0.74$, $p < 0.001$), with a distinct summer peak. These sites have a substantial influence from local traffic and the highest mean BC. Figure 5-2 shows good correlation between the Roxbury and HSPH sites ($R^2 = 0.85$, $p < 0.001$). These sites have a weaker summer peak, consistent with their more urban scale siting. Nine months of the year have a similar pattern, but May, June, and October were different, for unknown reasons. South Street and HSPH are poorly correlated due to the different scales of influence for these sites ($R^2 = 0.21$, $p = 0.14$). For the same reason, Roxbury and South Street are also poorly correlated ($R^2 = 0.14$, $p = 0.12$)

Springfield showed a very different seasonal pattern than the Boston sites, perhaps due to the valley topography. Figure 5-3 shows a late winter minimum and then an increase in BC through November with no clear summer peak. All sites had a peak in November that is not readily explained.

Figure 5-1. Monthly Mean BC for Four Sites with Inverse Wind Speed.

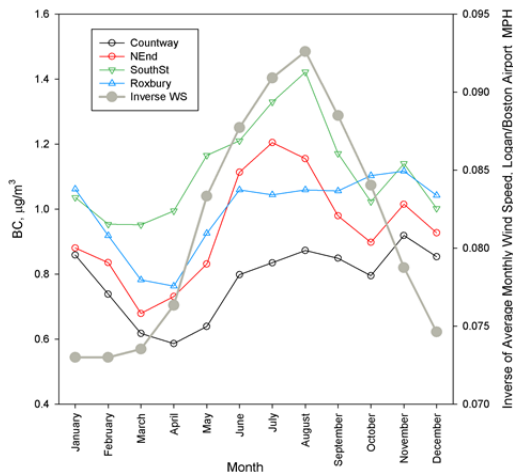


Figure 5-2. Correlation of Monthly Mean BC for HSPH and Roxbury.

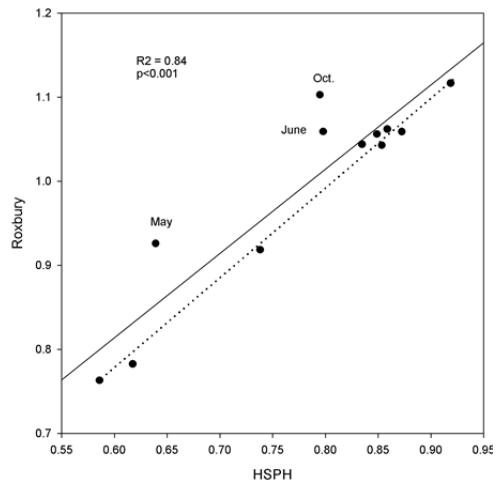
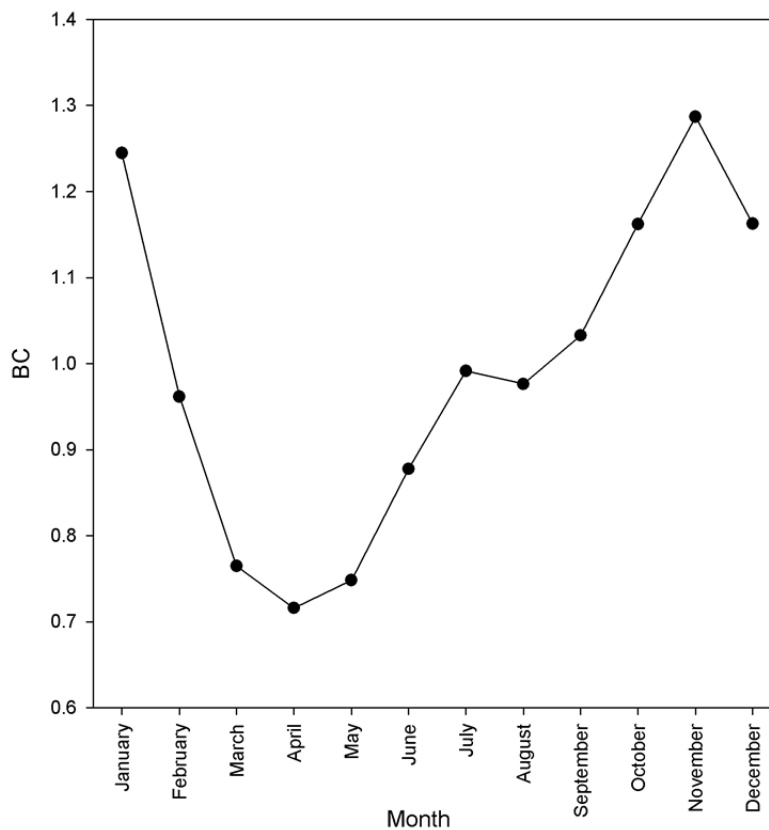


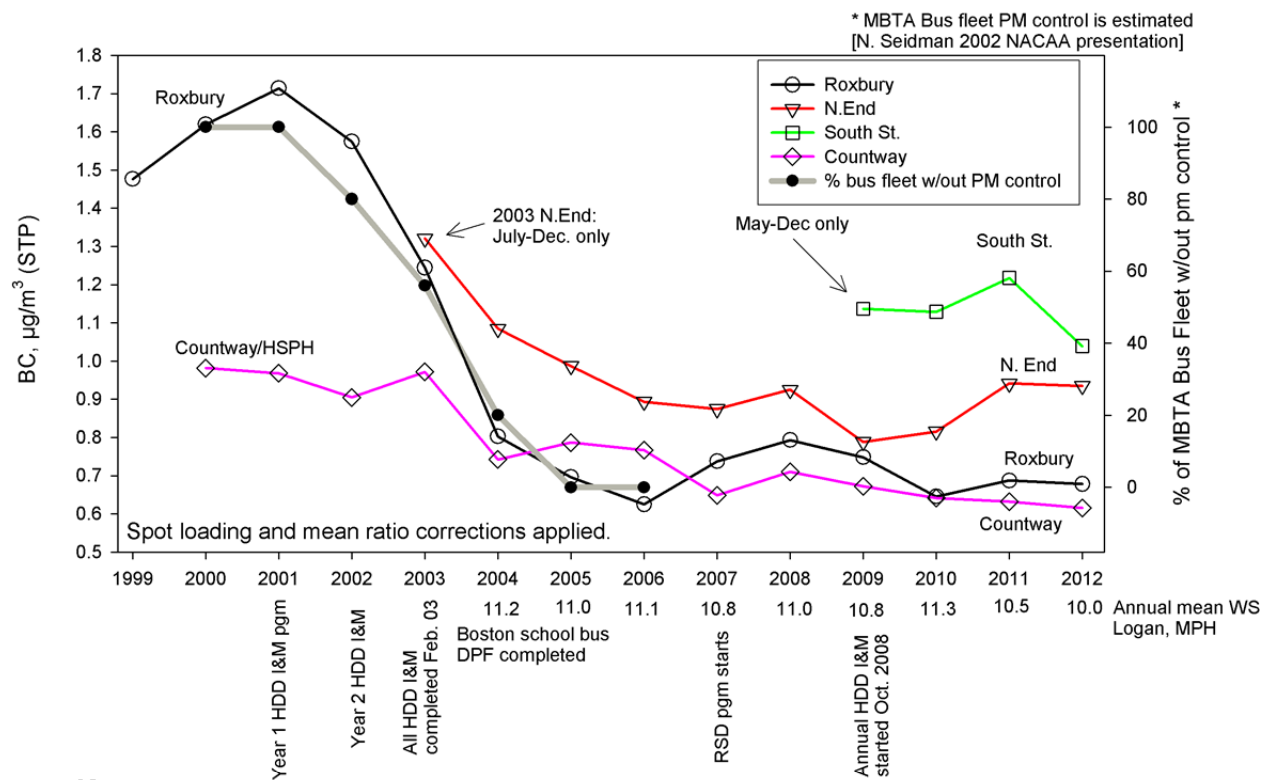
Figure 5-3. Monthly Mean BC for Springfield, 2006-2012.

5.2. Trends of Annual Mean BC.

Figure 5-4 shows the annual mean BC trend data from 1999 to 2012 for the four Boston sites. The dates of MassDEP's heavy-duty diesel control programs and the timing of the MBTA and Boston school bus fleet clean-up are noted. There was a striking drop in Roxbury BC over a three-year period (2002-2004). This directly matched the progress of the MBTA bus fleet clean-up effort, which controlled PM emissions, for the first time, on 100% of the buses between late 2002 and the end of 2004 (Seidman, 2002). The percentage of buses without PM controls is shown on the right axis of Figure 5-4.⁷ Note that because of strong seasonal BC patterns that differ across sites, BC trend analysis must be done on annual means.

⁷ Annual average wind speed from Logan Airport is shown in Figure 5-4 for 2004 and later, as higher wind speed tends to result in lower BC concentrations (due to improved dispersion) and may explain some of the year-to-year variation in mean BC at some sites. Appendix H shows the seasonal and year-to-year variation in wind roses from Logan Airport for 2000-2012.

Figure 5-4. Four Site Boston Trend.



Notes:
 “HDD I&M” is the MassDEP heavy duty diesel inspection and maintenance program.
 “DPF” is diesel particulate filter
 “RSD program” is the MBTA’s bus garage Remote Sensing Device program for bus emissions.
 “Spot loading and mean ratio corrections” are measurement method artifacts that varied over time and could affect trend analysis if not controlled for.

The MBTA bus fleet clean-up effort is best explained in the following summary, by Nancy Seidman of MassDEP. This is from the National Association of Clean Air Agency’s (NACAA) summary of its fall 2002 meeting.⁸

The Clean Bus Program sprung from 3 events: Governor William Weld’s pledge in the mid-1990s that MBTA would not purchase any new diesel buses, Central Artery air quality mitigation commitments to purchase 200 additional and 200 replacement clean buses and a consent decree requiring that 200 additional buses use compressed natural gas (CNG) or, if diesel, be retrofitted. MBTA will have 358 CNG buses by early 2003; as of May 2002, all of its diesel buses use ultra-low-sulfur diesel fuel, and 400 of its buses are scheduled to be retrofitted with diesel particulate filters (DPFs) between now and 2004. All of the oldest (1989) buses will be retired as of December 31, 2004. As a result of these efforts, by 2004, PM emissions are expected to decrease almost 90 percent from 2000 levels.

⁸ NACAA, formerly the State and Territorial Air Pollution Program Administrators/Association of Local Air Pollution Control Officers (STAPPA/ALAPCO). Accessed May 21, 2013. <http://www.4cleanair.org/Oldmembers/members/FallMembership02.pdf>.

Another factor that likely contributed to this rapid drop in Boston BC was the retrofit of the entire Boston school bus fleet with DPFs between 2003 and 2005. This work was funded by two EPA Supplemental Environmental Projects,⁹ and retrofitted the entire fleet of 600 Boston school buses with DPFs starting in 2003 and ending in the spring of 2005. The MBTA closed the Bartlett Street bus garage near Dudley Square at the end of 2003. This may also have been a factor in the reduction of Roxbury BC during this time period.

The “Big Dig” Central Artery downtown tunnel opening occurred during 2003 and thus is another factor to consider when evaluating BC trends. The Northbound lanes of the I-93 O’Neill tunnel (replacing the elevated downtown section of the Expressway) opened in March 2003, and southbound lanes opened in December 2003. Although overall downtown Boston mobile source emissions would not have been expected to change due to the new tunnels, the spatial patterns did, with tunnel exhaust vents at several points along the route. These changes could have affected BC at the North End site, but this is difficult to assess since that site did not start monitoring BC until July 1, 2003. Even after the December 2003 tunnel opening, substantial construction continued on the surface, including demolition of the elevated highway and construction of additional surface access roads in the North End. The opening of the Big Dig tunnels would not be expected to have any influence at the other two Boston BC sites (Roxbury and HSPH/Brigham Circle).

5.3. BC Trends after 2005.

The next step in the trend analysis was to evaluate changes after the large bus fleets were controlled for PM emissions. Starting in 2005 after heavy-duty diesel PM from the MBTA and Boston school bus fleets was controlled and the Bartlett Street Bus garage in Roxbury was closed, there was no clear trend in BC at the North End and Roxbury sites. The annual mean BC for 2005 and 2012 for these two sites was essentially identical: 0.70 and 0.68 $\mu\text{g}/\text{m}^3$ for Roxbury, and 0.99 and 0.94 $\mu\text{g}/\text{m}^3$ at the North End. Figures 5-5 and 5-6 show the regression of annual mean BC versus year at these two sites from 2005 through 2012. Although there was an indication of a very small downward trend over these years, the slopes of the regressions were not significant ($p = 0.65$ and 0.8 for North End and Roxbury respectively). The Roxbury BC trend from 2007 through 2012 (Figure 5-7) did have a downward trend, but the regression was still not significant ($p = 0.11$). When all Roxbury BC years are included (Figure 5-8), there was a very significant trend ($R^2 = 0.71$, $p < 0.001$) driven by the large difference in pre- and post-retrofit year BC concentrations.

⁹ United States Department of Justice. “U.S. Enforcement Case Brings Clean Air Benefits to Boston: \$6 Million Settlement Includes Largest School Bus Pollution Control Project In Country.” Last modified January 30, 2004. http://www.justice.gov/opa/pr/2004/January/04_enrd_058.htm.

Figure 5-5. Regression of Annual Mean BC vs. Year 2005-2012 for North End.

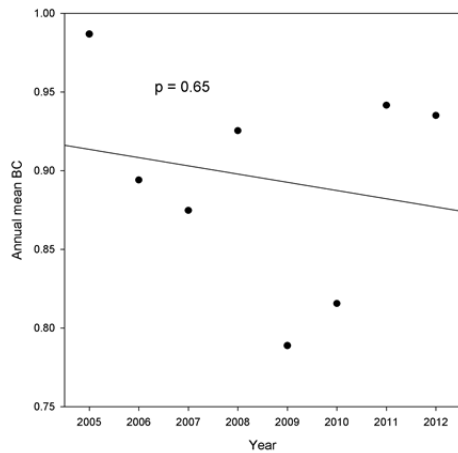


Figure 5-6. Regression of Annual Mean BC vs. Year 2005-2012 for Roxbury.

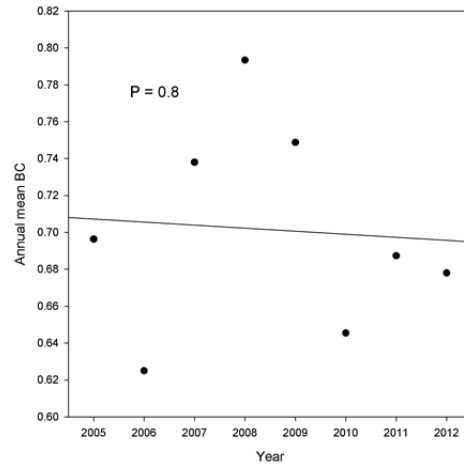


Figure 5-8. Roxbury BC Trend from 2007-2012.

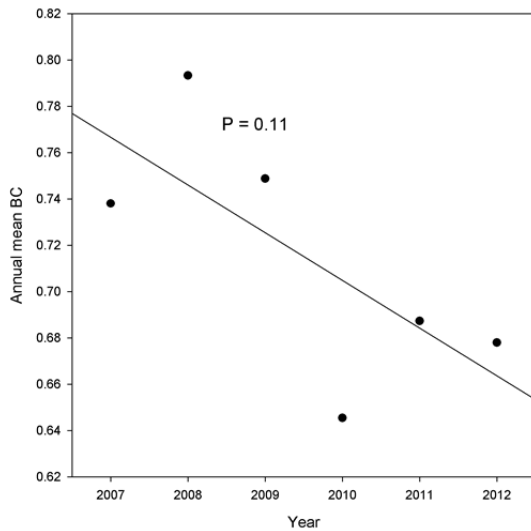
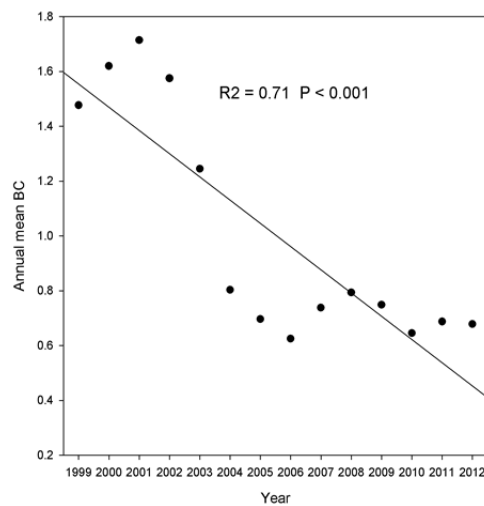


Figure 5-7. Roxbury BC Trend from all years.



The same trend analysis for the HSPH Brigham Circle BC data showed a robust downward trend over time for both all years and limited to 2005 to 2012. Figures 5-9 and 5-10 show the regression of BC versus year for 2000 to 2012 and restricted to 2005 to 2012. It is not clear why the HSPH site showed a trend for 2005 to 2012 and the Roxbury site did not. Both sites are urban scale over the 2005 to 2012 period and had similar mean BC, $0.70 \mu\text{g}/\text{m}^3$ for Roxbury and $0.68 \mu\text{g}/\text{m}^3$ for HSPH.

Figure 5-9. HSPH Trend 2000-2012.

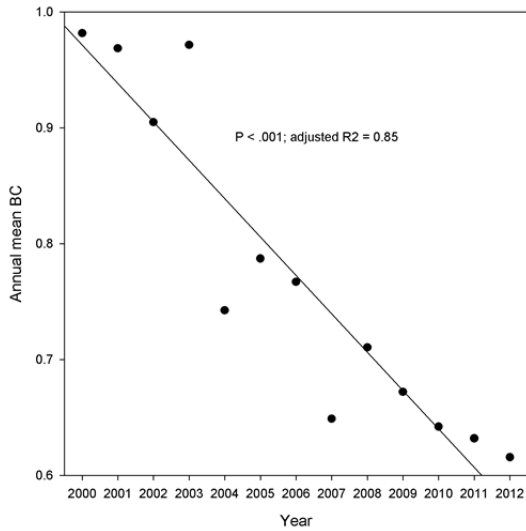
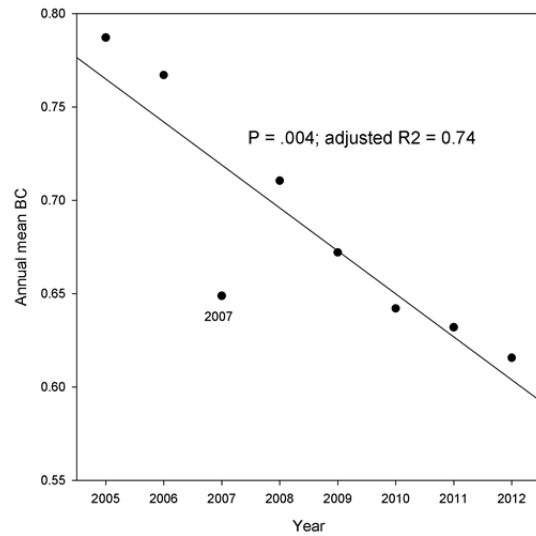
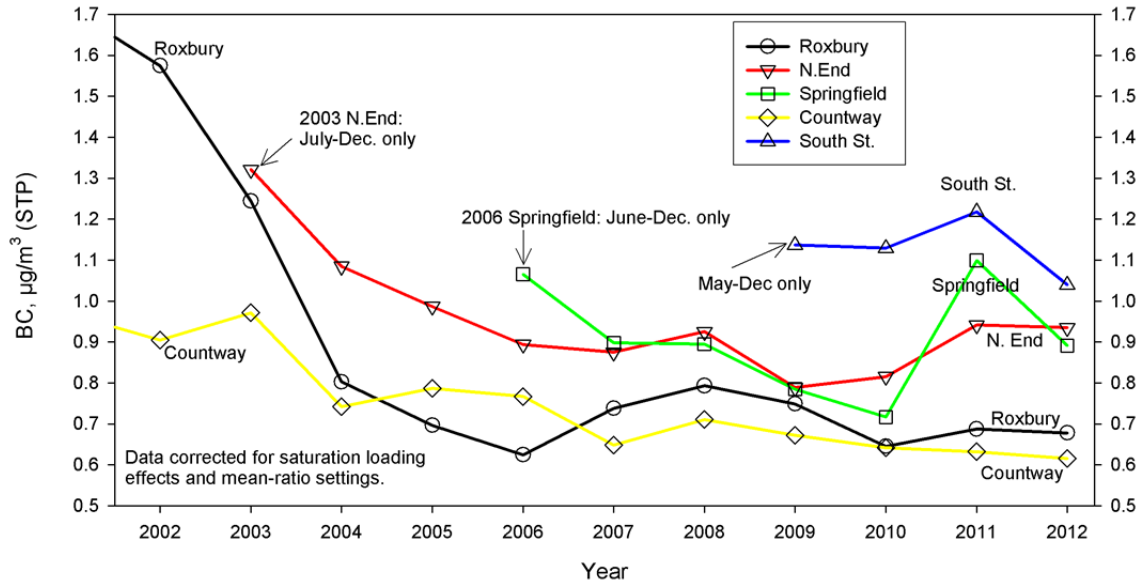


Figure 5-10. HSPH Trend 2005-2012.



MassDEP started monitoring BC in Springfield in the middle of 2006. Figure 5-11 shows BC for the Springfield and the four Boston sites. While there was a slight downward BC trend in Springfield from 2006 to 2010, 2011 was slightly higher than the seven-month mean in 2006 ($1.10 \mu\text{g}/\text{m}^3$ for 2011, and $1.07 \mu\text{g}/\text{m}^3$ for 2006). All BC sites, except the HSPH site, increased from 2010 to 2011, implying a weather-related factor. In 2012, BC levels at the South Street and Springfield sites dropped slightly, while levels in the North End did not change.

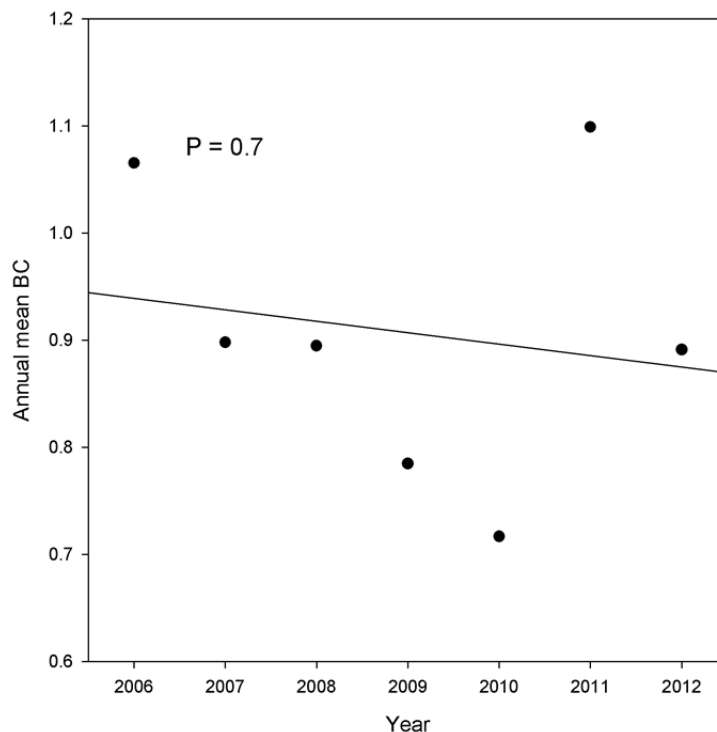
Figure 5-11. BC Trend for Four Boston BC Sites with Springfield.



The seven years of BC data for the Springfield site presented no trend. Figure 5-12 shows BC versus year from 2006 through 2012. The cause of the large increase at Springfield from 2010-2011 is unknown, although all sites except Countway did increase at the same time,

implying a regional process at least in part. Weather patterns would be one possible factor, but not sufficient alone to explain the observed increase in Springfield BC.

Figure 5-12. BC vs. Year for Springfield.

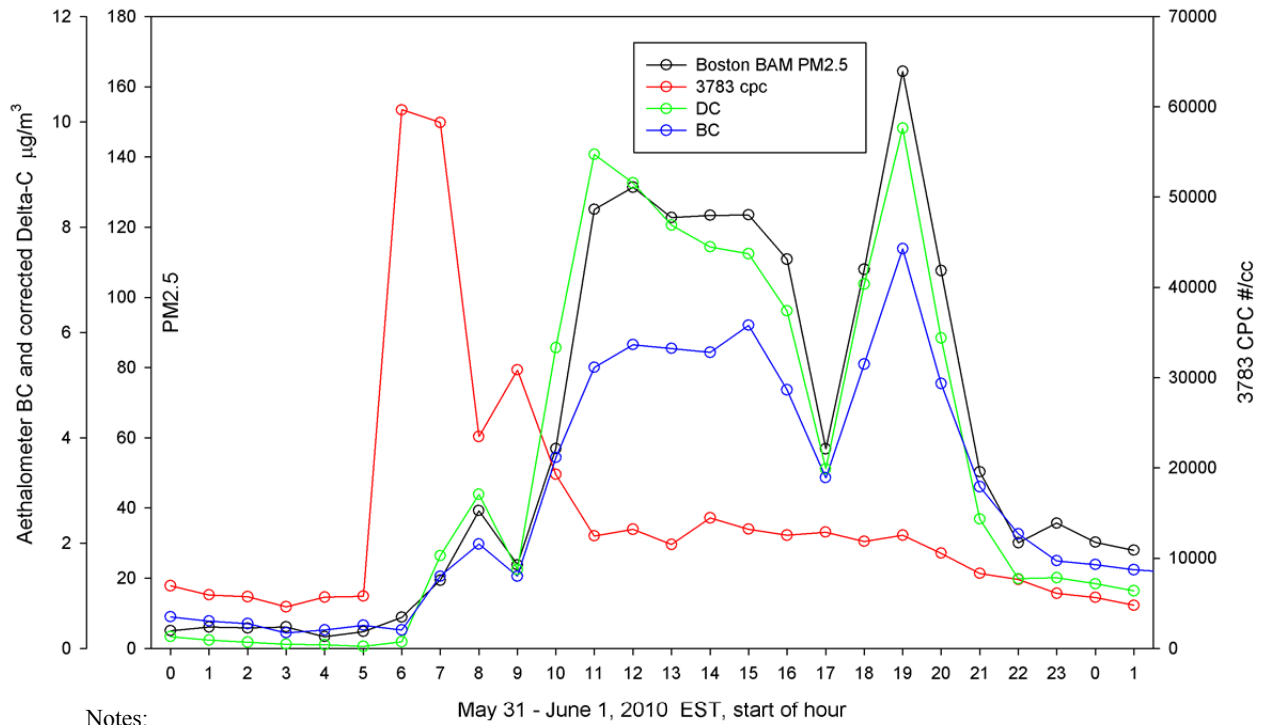


5.4. Wildfire Event of May 31, 2010

On the morning of May 31, 2010, smoke from a large wildfire in Quebec arrived in Boston, causing fine particulate ($PM_{2.5}$) to exceed $120 \mu\text{g}/\text{m}^3$ for several hours. BC concentration levels between 6 to $8 \mu\text{g}/\text{m}^3$ were recorded, consistent with woodsmoke BC being between 5% and 10% of the total woodsmoke PM (Naher et al., 2007). Figure 5-13 shows hourly data for $PM_{2.5}$, BC, and Delta-C, a woodsmoke indicator¹⁰ from South Street, and particle number concentration (PNC, or ultra-fine particles) from the HSPH site. PNC peaked early in the morning, before photochemistry could accelerate the particle aging process. BC, DC, and $PM_{2.5}$ tracked well from this common source, as expected.

¹⁰ Delta-C is UVC minus BC. UVC is the same measurement as BC but at a shorter wavelength of light. The shorter wavelength of UVC responds to organic compounds in fresh woodsmoke that the BC channel does not measure. BC is measured at 880 nm (near-IR), and UVC at 370 nm (near-UV).

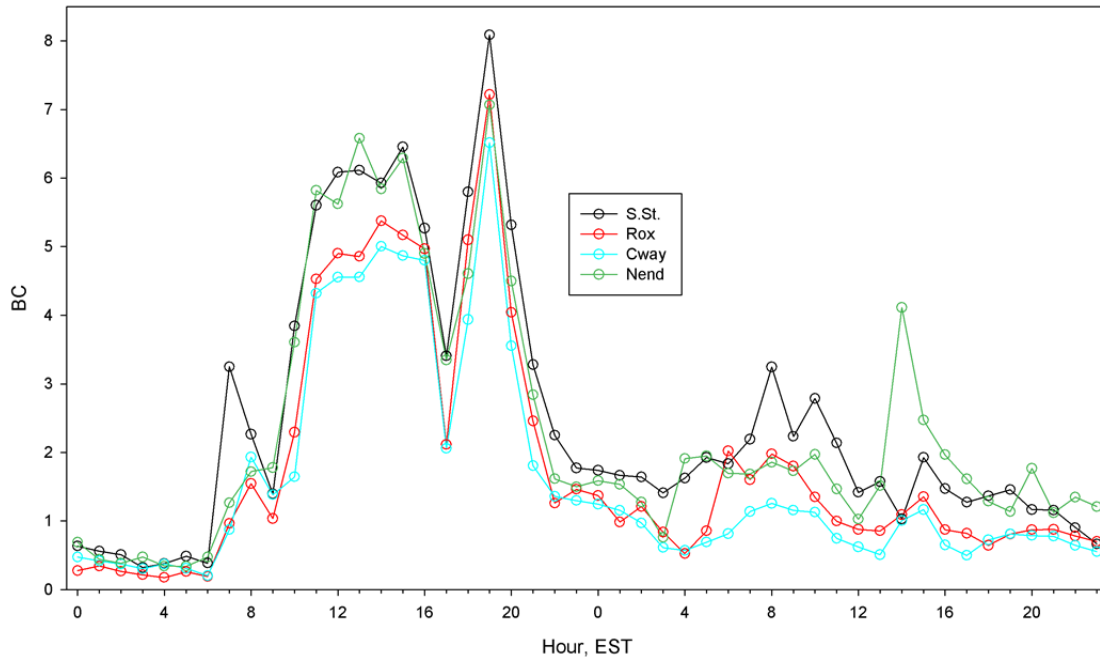
Figure 5-13. Hourly Event Data.



Notes:
 BAM is the beta-attenuation continuous PM monitor
 3783 CPC is the method for particle number concentration measurement.
 DC is "Delta-C", the 2-channel Aethalometer woodsmoke surrogate measurement.

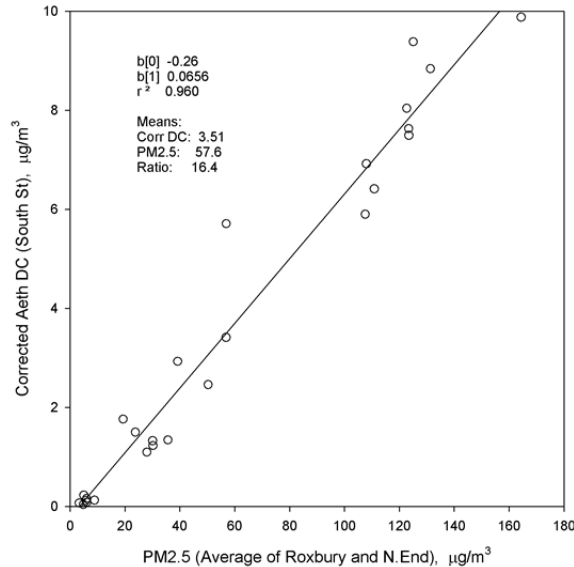
Figure 5-14 shows BC from the four Boston sites within the two-day time period of May 31 to June 1, 2010. Despite the source of the fire being several hundred kilometers away, there was some spatial difference across the sites. The only consistent pattern was that the HSPH Countway site was often the lowest. Roxbury generally tracked HSPH well. South Street or the North End site usually measured the highest BC levels.

Figure 5-14. Hourly BC for Four Boston Sites During the Woodsmoke Event.



Moreover, the wildfire event clearly shows the value of the Delta-C Aethalometer woodsmoke indicator. Figure 5-15 presents the regression of hourly Delta-C from South Street for PM_{2.5} (the average of the North End and Roxbury MassDEP continuous PM monitors). The R² value was 0.96. The ratio of PM_{2.5} to Delta-C was 16, which was in the expected range for woodsmoke that had undergone some photochemical aging.

Figure 5-15. Regression of Hourly Delta-C from South St. on PM2.5.



6. CONCLUSIONS.

6.1. Spatial Analysis.

Substantial gradients in BC existed over a 35 km scale, both in 2003 and for 2010 through 2012. Mean BC in 2003 varied by a factor of 3.5 from downtown Boston to the regional background site, and was much larger for sub-daily event periods (10 times or more). The data indicate that the neighborhood spatial scale of urban-excess PM_{2.5} for Boston was limited to approximately 10 miles from the downtown area. This is important from both an air toxics exposure and a control strategy perspective. In 2010, the ratio of North End to Stow mean BC was 2.8; when 2011 North End BC is used, the ratio increased to 3.2, which was similar to the ratio of 3.5 observed in 2003. The ratio between 2010 mean BC at South Street and Stow was 3.9. These data suggest that the relative gradient has decreased slightly since 2003. This is consistent with the majority of diesel PM controls implemented between 2002 and early 2005 being in place by the end of 2003.

For the core urban area, BC levels at all neighborhood-scale sites were elevated relative to the background site, but urban gradients were not distinct for most of these sites. Short-term (one-hour) correlations across these sites ranged from very good to poor. Some urban sites were much better indicators of BC in the general downtown area than others.

BC appeared to be a reasonable indicator of local mobile source aerosol. Winter oil space heating and woodsmoke did not appear to be significant BC interferences in the urban area.

A limited assessment of spatial trends from 2009 to 2012 at four sites showed results similar to the 2003 data. BC at the South Street site at the NESCAUM office was distinctly higher than other Boston sites, and was similar to BC measured at another South Street site in the summer of 2003. Based on pollutant-wind analysis, the proximity of the South Street site to the I-90 and I-93 interchange to the southwest as well as I-93 above ground to the south is the likely reason for the elevated BC at that site. The transportation complex at South Station did not appear to be a contributor.

Diurnal patterns for these four sites were similar in shape to 2003, with a distinct week-day morning peak and diurnal patterns only during weekend days. A notable exception to the weekend pattern was a large peak at 11 a.m. EST for South Street; this was due to a very local emergency diesel genset test every Saturday morning. One-minute BC has peaked to over 100 $\mu\text{g}/\text{m}^3$ during those tests.

6.2. Temporal Trends.

A substantial decrease in BC was observed between 2002 and 2005. At least two major factors likely drove this decrease: the drastic clean-up of PM emissions for the MBTA bus and the Boston school bus fleets, which occurred during this period. MassDEP's heavy-duty diesel Inspection and Maintenance program began in 2001, and was fully implemented by early 2003. This program likely contributed to the observed decrease in BC levels in 2003 and subsequent years. Thus the large decrease in BC between 2002 and 2004 is the result of specific intervention programs, and demonstrates the effectiveness of such efforts in reducing diesel PM. There are limited data from other urban areas that show a general downward trend in BC or

elemental carbon (EC) over the last decade, including data from the EPA chemical speciation network, but Boston is the only urban area where a rapid and substantial drop in BC can be attributed to intervention programs.

Trends after 2005 were present but not consistent across the three long-term Boston BC sites. Roxbury had a downward trend from 2007 to 2012, but it was not significant, with a p value of 0.11. When data from 2005 and 2006 data were included, there was no observed trend ($p = 0.8$). At the HSPH site, there was a significant downward trend from 2005 to 2012 ($p = 0.004$). The North End site showed no trend for any period after 2004. The Springfield BC data from July 2006 to 2012 also showed no trend for that period.

At all sites, there was a strong seasonal pattern for BC. In Boston, at all sites except for Roxbury, BC was substantially higher in the summer than in winter. This is consistent with seasonal wind speeds. The pattern was strongest at the North End and South Street sites. Roxbury BC was lower from February to May and constant during the remainder of the year. Springfield had a strong but different seasonal pattern; BC was lowest in April and highest in November.

6.3. Future Work.

BC will continue to be monitored at the four existing Boston sites, thus allowing assessment of future trends. The new MassDEP near-road monitoring site, located on the Southeast Expressway inbound, 2 km east of the Roxbury site, includes BC measurements starting November 1, 2013. This makes a total of five Boston BC monitoring sites.

A new version of the Aethalometer has been recently introduced (Model AE33 or TAPI-633). While its basic operating principles are unchanged, it is a radically different design. The new instrument attempts to correct for spot loading errors in real-time, but may produce data that are different from the older AE21 / AE22 Aethalometer. To ensure the integrity of ongoing trend analyses, it will be important to characterize any differences in BC data produced by the older and newer versions of the Aethalometer.

NESCAUM has been evaluating the performance of prototype and production versions of the AE33/633 compared with the AE21/AE22 (older models) Aethalometer performance over the last two years at the South Street site. The production version of the AE33 was evaluated from December 2012 to September 2013 using two Model 633 Aethalometers that MassDEP purchased. From August through October 2013, a comparison of the old and new Aethalometer instruments was performed at the Roxbury MassDEP site. Results show good correlation, but BC data from the new model 633 Aethalometer is substantially higher (~25 to 30%) than the existing AE22 Aethalometer at that site.

As mobile source BC continues to be reduced in a manner similar to the dramatic reduction of carbon monoxide over the last two decades, the utility of BC as an indicator is also likely to decrease. While this is desirable from a health and exposure perspective, there are other mobile source pollutants of concern that may not be reduced, and it may be difficult to find another easily measured marker of mobile sources.

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Appendix A: Preliminary Analysis for the Six-week Nine-site Data (January 11, 2003 to February 26, 2003).

Appendix A.

Spatial and Temporal Assessment of a Mobile Source Aerosol Indicator During Winter in Boston, MA: A Pilot Study

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Poster # 05-27, presented at “Particulate Matter: Atmospheric Sciences, Exposure and the Fourth Colloquium on PM and Human Health” in Pittsburgh, PA, March 31-April 4, 2003
An International Conference Managed by the American Association for Aerosol Research

Introduction. Many urban locations are expected to be near or over the annual U.S. EPA standard for PM_{2.5} of 15.0 µg/m³. Data from PM_{2.5} monitors in the same metropolitan area only a few miles apart can be substantially different, with some over and some under the standard. Variation on this spatial scale is often presumed to be driven by local mobile source particle emissions. It is important to define the spatial extent of elevated PM_{2.5} for compliance, air toxics assessment, and control purposes, as well as for health effects studies. One indicator of local mobile source aerosol in urban areas is black carbon soot (BC, associated with primary Diesel and automotive emissions), which has been shown to be well correlated with integrated elemental carbon (EC) filter samples. BC can be measured in real-time with a commercial instrument (Aethalometer) that is relatively simple to install and operate; the principle is light absorption through a quartz filter (optical density).

Study Design. A pilot study was performed during the winter of 2003 to assess the spatial and temporal variation in the local mobile-source aerosol over the greater Boston area, using BC as an indicator for that PM component. Given that other major mass components of PM_{2.5} (sulfate, organic carbon) in the northeast U.S. are secondary transported aerosols and tend to be uniform over this scale, the locally generated "tailpipe" component of PM should drive the shape of PM_{2.5} spatial gradients over the metro area. A series of nine monitoring sites were selected heading WNW from downtown Boston out 35 km (Figure 1), generally away from immediate large sources of local mobile-source emissions. This design avoids coastal influence and allows the pilot study to be more readily generalized to other large metro areas in the Northeast.

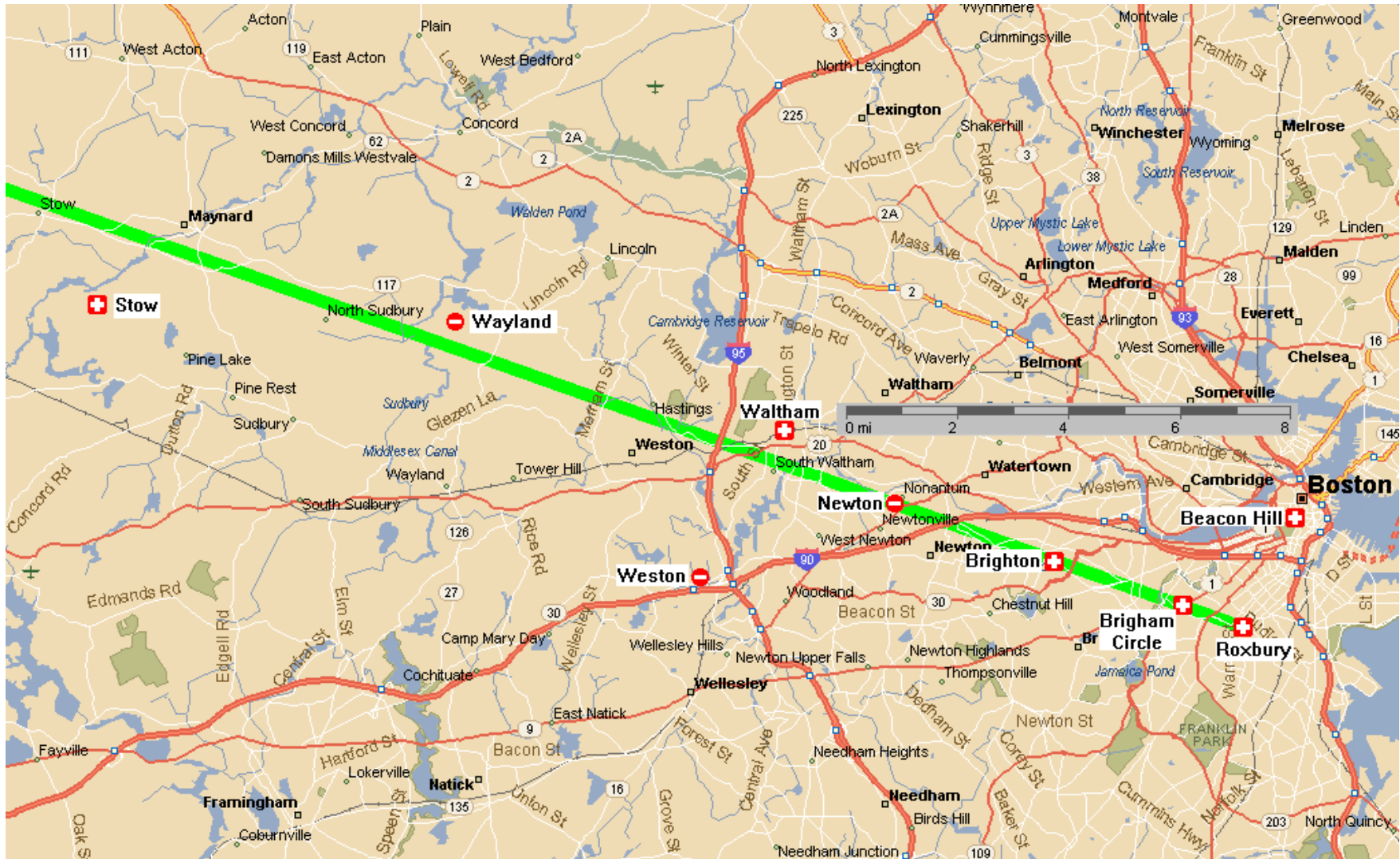
<u>Site Locations</u>	<u>Km</u>	<u>Site Description.</u>
Beacon Hill (Boston)	0.0	Urban Residential (adjacent to the State House)
Roxbury (Boston)*	3.5	Urban Residential/Commercial (Dudley Sq.)
Brigham Cir. (Boston)*	4.1	Urban Residential/Commercial (Harvard Medical Area)
Brighton (Boston)	7.0	Semi-Urban Residential
Newton (Nonantum)†	11.7	Suburban Residential/Light Commercial
Waltham	14.9	Suburban Residential/Light Commercial
Weston †	17.4	Suburban Residential, near I-90/I-95
Wayland †	25.1	Suburban Residential
Stow	35.3	Semi-rural open land; Regional Background site

* existing sites run by other organizations

† Non-core network sites, only run 11 Jan. to 26 Feb. or 6 Mar. 2003

Measurement Methods. BC was measured with Magee Scientific Inc. Aethalometers, models AE-16 and AE-42, with fine-mode size cut inlets. The default “sigma” value of 16.6 was used. All BC data are reported in $\mu\text{g}/\text{m}^3$ at STP. Leak tests and external flow calibrations were performed on all instruments. Data were collected at 5-minute intervals using the Aethalometer’s internal storage; 1-hour mean concentrations of BC were calculated with the Washington University-St. Louis Aethalometer data processor. Hourly data were screened for unusual local source influence, such as woodsmoke; a total of 31, 15, and 9 hours were removed from the Waltham, Beacon Hill, and Wayland sites respectively between 4 Jan. and 6 March 2003. An additional 11 hours were removed for all sites overnight on Saturday evening 18 January.

Figure 1. Site Locations



“+” indicates a core network site; “-” is a temporary site (11 Jan - 26 Feb 2003).

Population Density. Population densities at the nine monitoring sites were generated to assess whether population is a surrogate for local mobile-source aerosol concentrations. Population estimates were generated using LandView5 software, a spatial and demographic database application created by the U.S. EPA, Census Bureau, Geological Survey, and NOAA (<http://landview.census.gov>). The software tallies Census 2000 block data for those block centroids whose coordinates fall within a circle defined by a prescribed radius. Geographic coordinates for the monitoring sites were used as center points to estimate population density within a 0.5 mile radii or 0.8 square miles.

Data Analysis. The core data set used for most analysis was from the six longer-term “core” sites, from January 4 to March 6, 2003 (data from the three non-core sites were used only in the spatial frequency distribution analysis), since these core sites had more days of data and will continue for a full year. Average spatial and diurnal spatial patterns were segregated by workday vs. non-workday. Population densities at each monitoring site were compared to mean black carbon levels.

Results and Discussion. Data capture for all sites exceeds 97% for each site after editing for local woodsmoke influence. **Figure 2** shows the core six-site network data as 24-hour running averages for the Jan. 4 to Mar. 6, 2003 period; a wide range of spatial and temporal BC concentration is evident, with urban sites consistently higher than non-urban sites. **Figure 3** shows the frequency distributions and means of hourly BC concentrations by site for all nine sites; the order is downtown to background from left to right. As one would expect, the data are lognormally distributed, and there is a general trend of lower BC as sites become less urban. Some sites show clear woodsmoke influence during certain periods. Obvious cases were removed, but it is still likely that some woodsmoke influence remains for the suburban sites, especially Newton and Waltham, during this pilot study period. It is notable that Brighton, only 7.0 km from Beacon Hill and 5.8 km from Roxbury, has a mean BC of less than half that of either Roxbury or Beacon Hill. There is a factor of 3.5 between mean BC in

downtown Boston and the regional background site at Stow, which is important from an air toxics exposure point of view. Based on available annual mean PM2.5 data, the gradient over this range is much smaller (approximately a factor of 1.4), with sites in downtown Boston at or near the annual PM2.5 standard of 15.0 $\mu\text{g}/\text{m}^3$. The BC data from this pilot study imply that the size of the metro area near the annual PM2.5 standard may be limited to core urban areas.

Preliminary results do not confirm the original hypothesis of mobile source-related “hot-spots”, but point to a more homogeneous elevated concentration in the core urban area. The original study design had targeted the Roxbury (near a large city bus station and bus “barns”) and Weston (500 meters NW of a major interstate highway exchange and toll booths) sites as likely to have elevated BC from local traffic influence. Neither of these sites was elevated compared to surrounding sites; the mean BC at Roxbury was similar to Beacon Hill, and Weston was similar to the two sites further west. Earlier limited work showed a residential site in South Boston (2.9 km SE from Beacon Hill) to have similar levels of mean BC as Roxbury. This supports the possibility of a fairly uniform spatial pattern of elevated BC in the core urban area for residentially oriented monitoring locations. We plan to investigate this further with a micro-scale study in downtown Boston later this year.

Weather was colder than normal during the study period by approximately 3 °C. Two of the sites in this network have been running for 4 years; this provides some context for the effect of weather on data from this study. The following table compares mean BC for Roxbury and Brigham Circle over this period with three previous years:

Jan-Feb mean BC	Roxbury (range)	Brigham Circle (range)	Ratio of means (range)
2000-2002	1.42 (1.25-1.56)	0.83 (0.76 - 0.89)	1.71 (1.39 - 1.92)
2003	0.89	0.62	1.42

Compared to 2000-2002, 2003 Jan-Feb BC was 37% lower at Roxbury, and 25% at Brigham Circle, and was the cleanest of the 4 years by a distinct margin. The ratio of Roxbury to Brigham Circle BC (1.42) was also lower this year than the 3-year mean ratio; this is consistent with overall BC levels being lower, and suggests that the spatial gradients reported for this pilot study may be a lower-end estimate.

Figure 4 shows the diurnal pattern of BC for the six core sites, broken down by work and non-workdays. For workdays, the morning rush hour BC at Beacon Hill and Roxbury is five times higher than the Stow site; diurnal patterns become weaker as sites become less urban. The lack of any distinct workday diurnal pattern at the Stow site confirms that it represents the regional background BC concentration. Non-workday diurnal patterns show no clear time of day effects for most sites, in part because only 19 days of data were available.

Figure 5 shows mean BC across the six core sites, broken down by work and non-workdays. There is a distinct difference only for the three most urban sites, and no difference at Waltham or Stow; this is consistent with the less urbanized sites having more regional and fewer local sources of BC.

Figure 6 shows the relationship between population density (0.5 mile radius) and mean BC at the nine monitoring sites. With the exception of Brighton (whose BC levels were lower than adjacent sites), population estimates decrease linearly as mean BC decreases. The coefficient of determination (R^2) between population density and mean BC is 0.43 for all nine sites but increases to 0.81 without the Brighton site. This indicates that it may be possible to predict spatial gradients in mean BC concentrations across this scale by using local population density, a readily available metric.

Figure 7 is a single 24-hour period showing the effect of poor dispersion on BC levels at all nine sites. Cloud ceiling (from Logan Airport in Boston) is used as a surrogate for the minimum mixing height.

When the ceiling is less than 2000 feet, urban levels of BC increase dramatically, even though this happens after the morning rush hour. During hour 17, the cloud ceiling increases to 6000 feet, and BC levels drop rapidly. **Figure 8** is a five-day time series from the eight sites running during that period, and shows a more typical regional stagnation event. PM2.5 data from the Roxbury site is also shown (on the right-axis). A distinct morning rush hour peak for both BC and PM2.5 occurs on Thursday, but not on Friday. Late Friday evening shows another distinct multi-hour peak; the cause is unclear, but the Celtics were playing Toronto in Boston that evening (they won!). Both peaks show an EC to PM2.5 ratio of 10 at the Roxbury site.

Conclusions. Substantial gradients in BC exist over relatively small distances in the metropolitan Boston area, however there may be a core urban area where levels are elevated but gradients are not distinct. Mean BC varies by a factor of 3.5 from downtown to a regional background site; this factor is larger for sub-daily event periods. Based on spatial, hour-of-day, and day-of-week patterns, BC appears to be a reasonable indicator of local “tailpipe” aerosol despite potential for woodsmoke interference, but it is not highly specific to Diesel or on-road vs. off-road sources. These pilot data suggest that urban PM2.5 attainment areas may not always extend over the entire metropolitan region, but be limited to a somewhat smaller urban zone. Although PM2.5 gradients in greater Boston are no more than a factor of 1.5, the BC gradient of 3.5 and the relatively rapid drop-off within the urban area are important from an air toxics exposure and control strategy point of view.

Study Limitations. The single (and somewhat atypical) cold weather season and limited pilot study spatial scope and duration limit the precision of estimates of both spatial gradients and absolute BC concentrations. Woodsmoke may be an interference especially in suburban locations. The relative contributions of Diesel [on- and/or off- road] vs. automotive engines to observed BC concentrations is

uncertain. Population density data may be imprecise at the 0.5 mile radius level due to the spatial resolution of those data. Traffic density data were not available in a useful form to compare to observed BC patterns.

Future work. Six core BC sites will continue for a full year's operation to allow assessment of seasonal factors and the significance of woodsmoke interference. The question of "hot-spots" will be addressed with micro-scale studies in downtown Boston later this year, and possibly a different site near I-90/I-95 in Weston/Newton. A high elevation site near Boston (Blue Hill, 200 m elevation, 12.5 km south of Roxbury) will be added to the network to further assess regional transport of BC. Further analysis is required to determine whether local population density can be considered a surrogate for local traffic density, which might be a more appropriate metric for predicting BC concentrations.

Acknowledgments. The following organizations and companies have supported this work in kind:
Massachusetts Department of Environmental Protection
U.S. EPA, Region 1
Magee Scientific Company, Berkeley CA
Harvard School of Public Health/Boston EPA-PM Center
Washington University-St. Louis/ STL EPA Supersite
Appalachian Mountain Club, Boston MA
BGI Inc., Waltham MA.
ETA Associates, Newton MA

Figure 2. 24-hour running means of hourly BC, Six Core Sites, Jan 4 - Mar 6, 2003

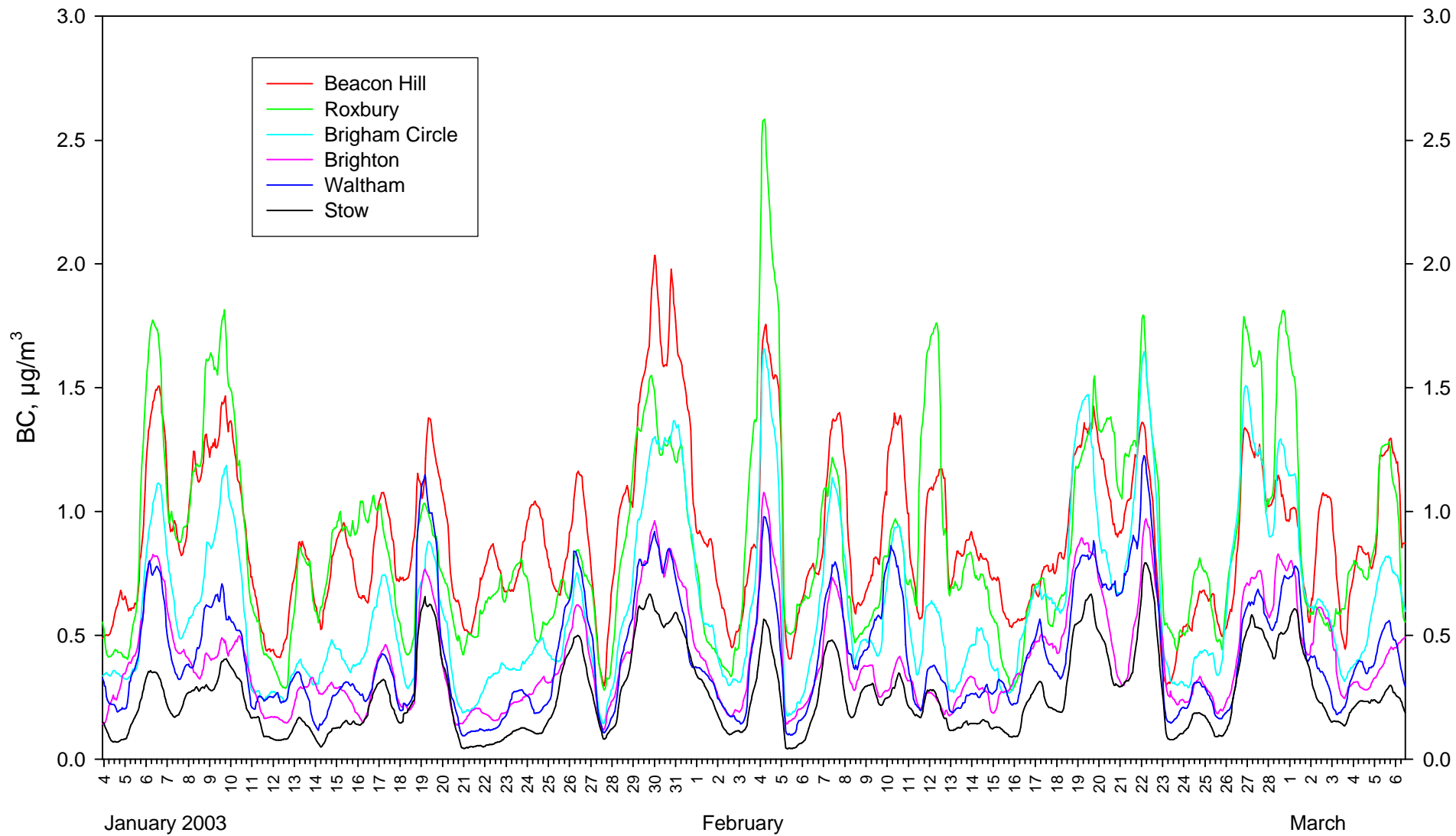


Figure 3. 11 Jan thru 26 Feb 2003 1-Hour Spatial BC Distributions

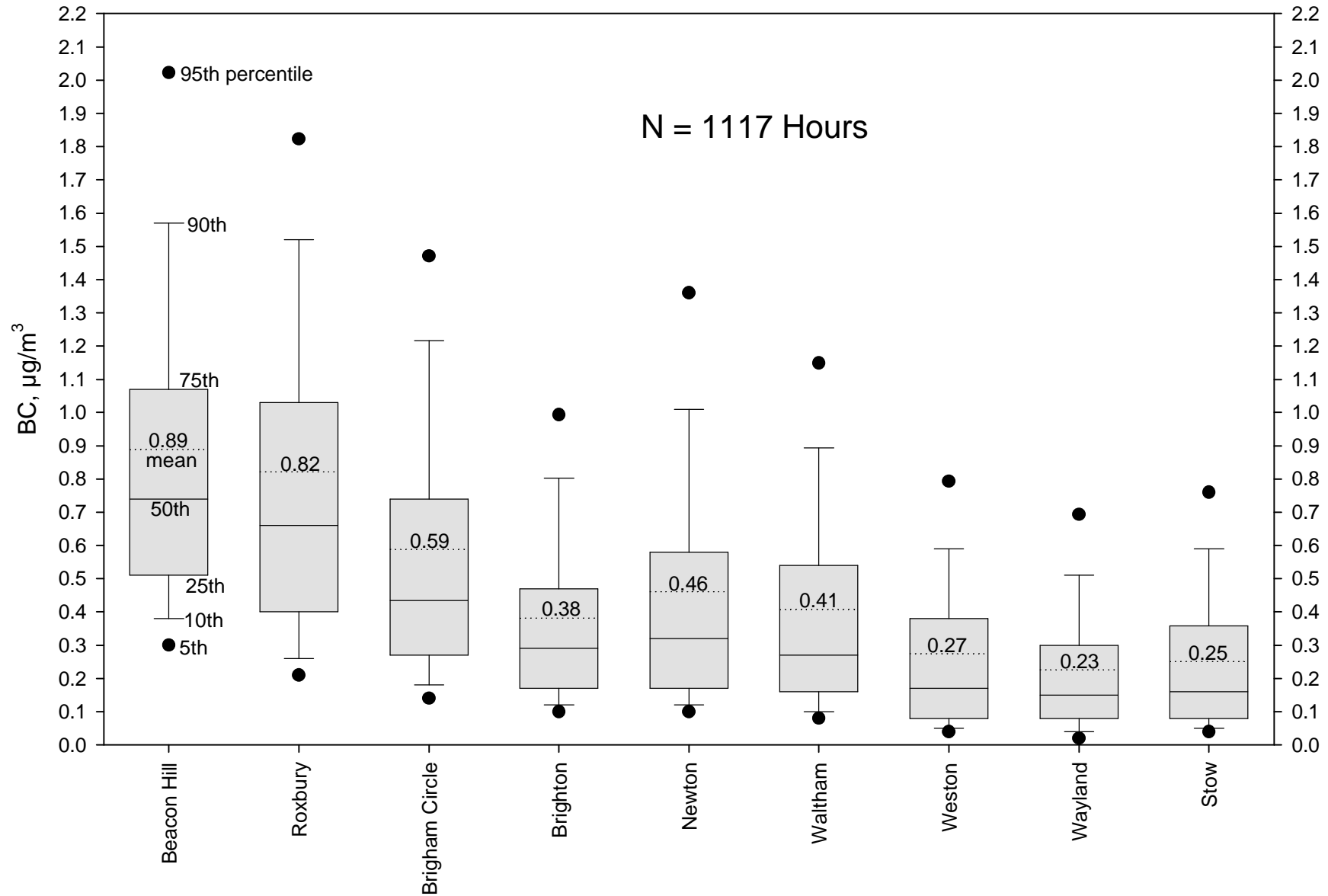


Figure 4. Diurnal BC Jan 04 - Mar 06, 2003

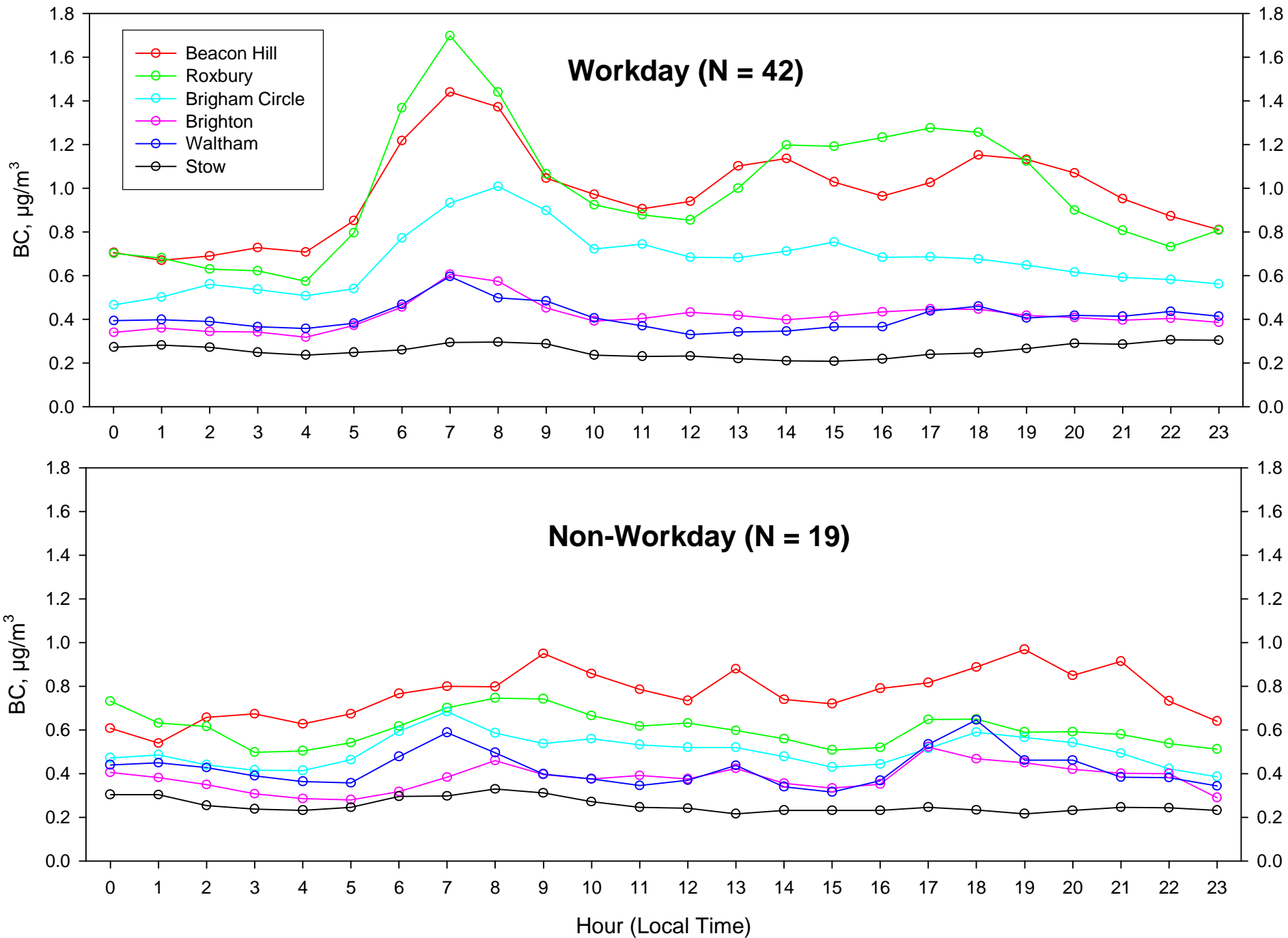


Figure 5.
Workday vs. Non-workday Mean BC
6 Core Sites, Jan 4 through Mar 6, 2003

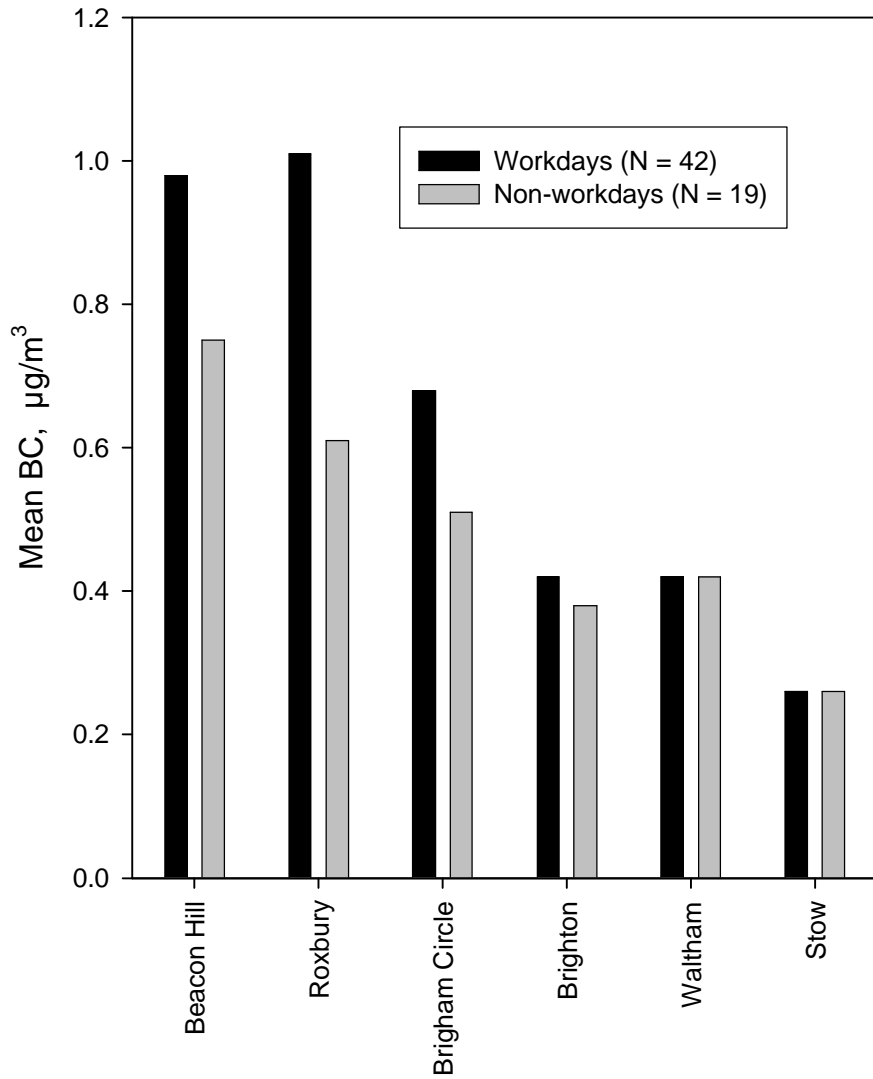


Figure 6.
Population Density (0.5 mile radius)
vs. Mean BC

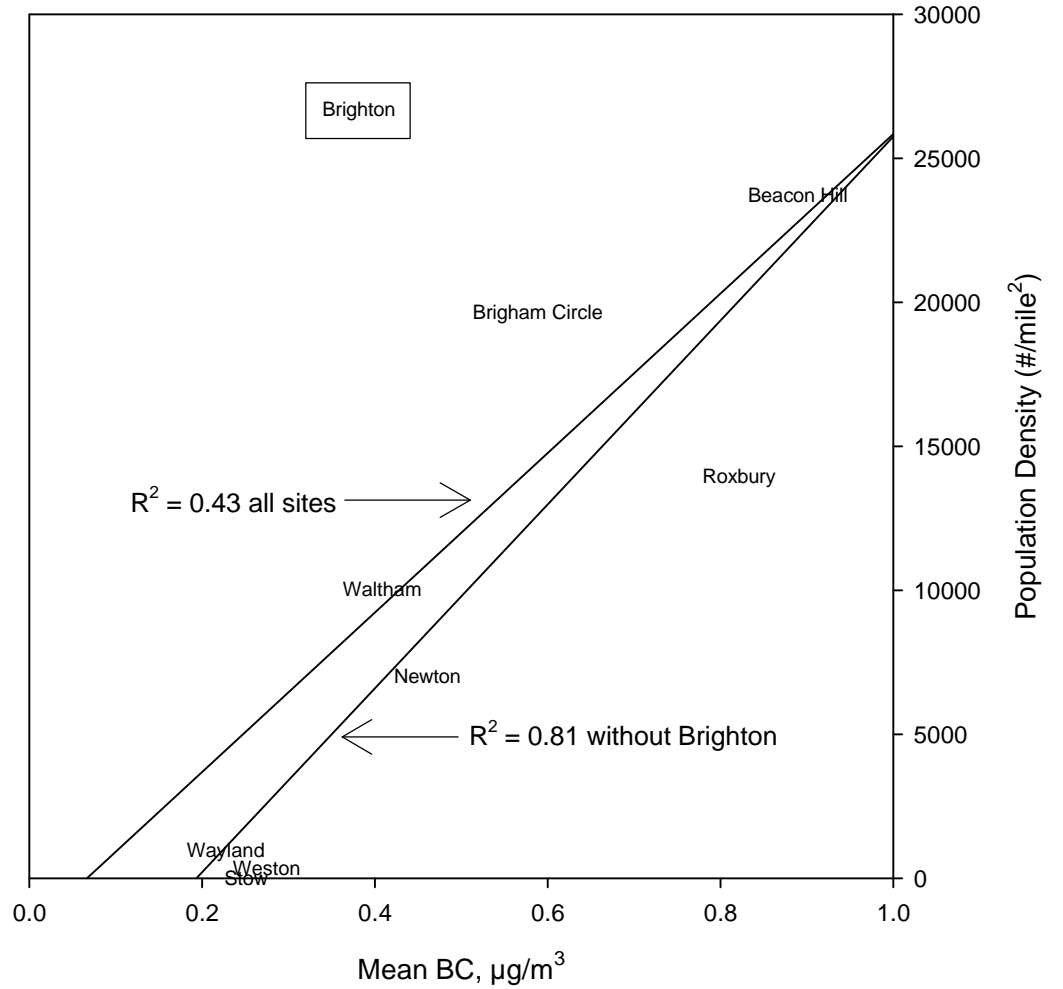
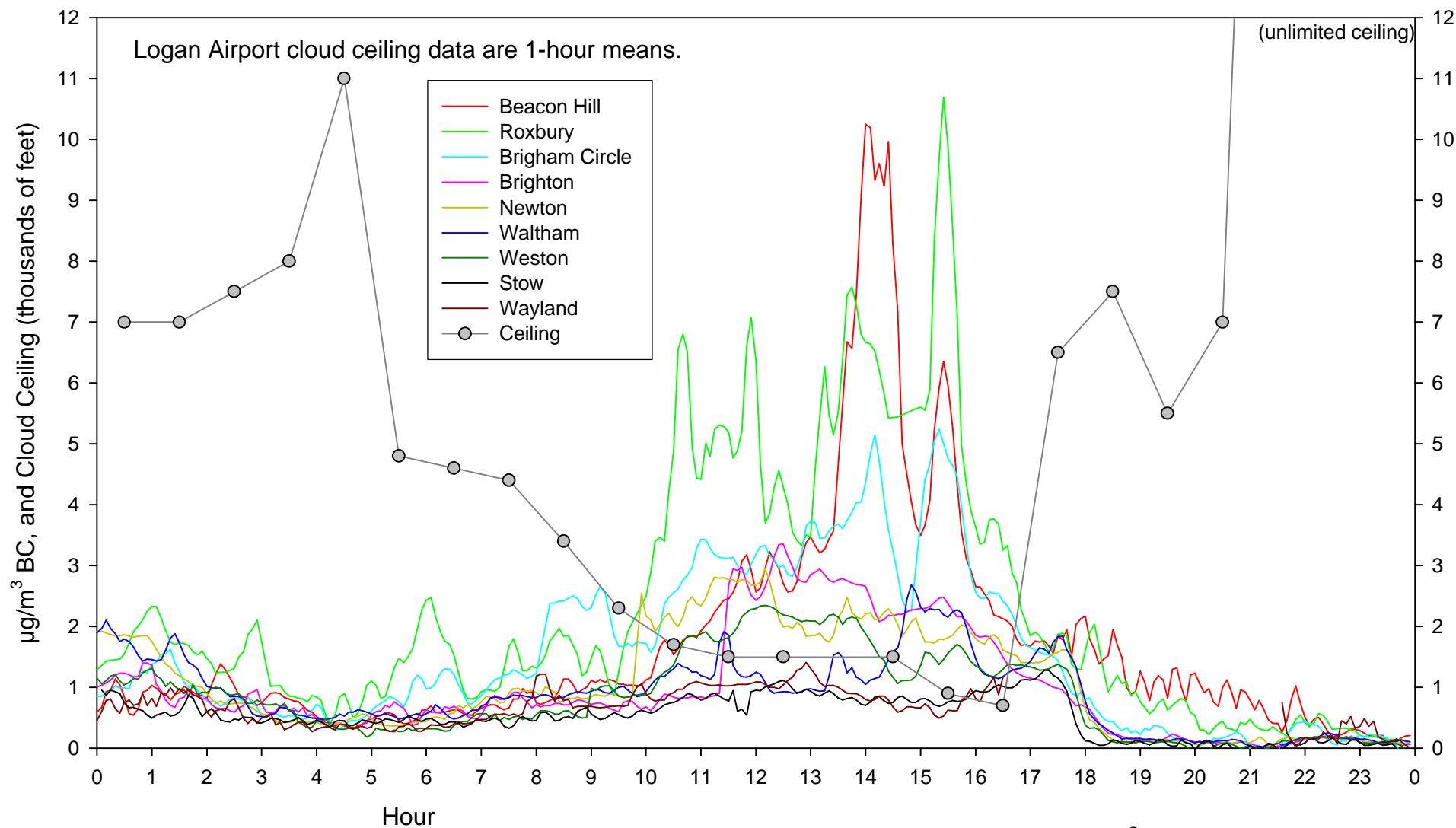


Figure 7. Feb 4 2003 BC Event
 15-minute running means of 5-minute data



Roxbury PM2.5 dropped from 41 to 5 $\mu\text{g}/\text{m}^3$ between 15:00 and 20:00

**Appendix B: Preliminary Analysis for the One-year
Six-site Data (December 20, 2002 to September 9,
2003), including the Summer Core Boston 10-site
Data.**

Appendix B.

Spatial and Temporal Aspects of Black Carbon Concentrations over the Boston Metro Area: An Update of Work in Progress

**George Allen and Philip R.S. Johnson
Northeast States for Coordinated Air Use Management**



**Based on material presented at the 22nd Annual Conference of the
American Association of Aerosol Research
Anaheim, CA October 22, 2003**

Rev. 16 Dec 2003

Background

- This study was designed to assess the spatial and temporal variation in "mobile source aerosol" as measured by black carbon in an urban area. Data collection is not yet complete, and all analysis presented here is preliminary and under development.
- By design, this is not a "hotspot" assessment, given that the monitoring sites were located >100 meters from the most intense "source" strength (adjacent major roadways) areas. Higher exposures than those reported here may exist at mobile-source "hotspots" both in the urban area and in non-urban areas as well.
- This study does assess "neighborhood-scale" (0.5 to 4 km) exposure gradients to black carbon across the metropolitan Boston area.
- Limitations of this study include semi-randomized monitor siting and monitor-to-monitor bias that are not accounted for at this time. Wind conditions that may result in dilution of source pollution at any given site have also not been taken into account in this preliminary analysis. The significance of observed gradients will be characterized using ANOVA analyses once the final year-long data set is available.
- This study has begun to characterize the urban mobile source aerosol gradient in greater Boston, has identified future areas for more refined microscale assessment, and has underscored the utility of black carbon as an indicator of fossil fuel combustion, showing that it can be reasonably specific for mobile source-related fine particulate in an urban area.

Introduction.

Need to define the spatial extent of elevated PM_{2.5} across urban areas:

- Compliance issues (PM_{2.5} attainment)
- Air toxics exposure assessment, Control assessment
- Health effects studies

Black Carbon (BC):

- Generally associated with fossil fuel combustion; in this study BC is shown to be a useful indicator of local [primary] mobile source aerosol in urban areas
- Not highly specific to diesel for the neighborhood scale siting (non-hotspot) used in this study

Method: Magee Scientific Aethalometer™

- Optical Density measurement, scaled to BC in $\mu\text{g}/\text{m}^3$
- Well correlated with DRI TOR-EC (less so with NIOSH/STN EC)
- Simple method, easy to deploy and operate
- Method bias across sites is typically 10% or less

Approach:

Previous work: Pilot study, winter of 2003 (AAAR PM conference poster, Spring 2003)

Assess spatial/temporal variation in “local mobile-source aerosol”

9 sites WNW from downtown

Scale: over the greater Boston area – out to 35 km (background)

Pilot results indicated that it was important to expand the work into a larger study:

Strong spatial gradients (>3 on average; more for events)

Look in detail at specific neighborhoods (Beacon Hill, S.Boston similar to Roxbury)

Run 6 sites for full year (address seasonal questions, woodsmoke and space heating interferences): all of 2003

“Neighborhood-Scale” study – Summer 2003: 10 sites in Boston

Detailed Study Description:

Many urban locations are expected to be near or over the annual U.S. EPA standard for PM_{2.5} of 15.0 µg/m³. Data from PM_{2.5} monitors in the same metropolitan area only a few miles apart can be substantially different, with some over and some under the standard. Variation on this urban spatial scale is often presumed to be driven by local mobile source particle emissions. It is important to define the spatial extent of elevated PM_{2.5} for compliance, air toxics exposure assessment, and control assessment purposes, as well as for health effects studies exposure estimates. One indicator of local mobile source aerosol in urban areas is black carbon soot (BC, associated with primary diesel and automotive emissions), which has been shown to be well correlated with integrated elemental carbon (EC) filter samples. BC can be measured in real-time with a commercial instrument (Aethalometer) that is relatively simple to install and operate; the principle is light absorption through a quartz filter (optical density).

This study assesses the spatial and temporal variation in the local mobile-source aerosol over the greater Boston area, using BC as an indicator for that PM component. Given that other major mass components of PM_{2.5} (sulfate, organic carbon) in the northeast U.S. are secondary transported aerosols and tend to be uniform over this scale, the locally generated "tailpipe" component of PM should drive the broad shape of PM_{2.5} spatial gradients over the metro area. A series of monitoring sites was selected heading WNW from downtown Boston out 35 km, using neighborhood-scale siting criteria (generally away from immediate large sources of local mobile-source emissions). The siting design avoids coastal influence; this and the neighborhood-scale siting allows the study to be more readily generalized to other large metro areas in the Northeast. More detail on the spatial scale of representation for monitor siting is in 40CFR58, appendix D: <http://www.epa.gov/ttn/amtic/files/cfr/pt58/40cfr58a.pdf>

6 Core Site Locations	Km	Site Description
Beacon Hill (Boston)	0.0	Urban Residential (near State House)
Roxbury (Boston)	3.5	Urban Residential/Commercial; EJ
Brigham Cir. (Boston)	4.1	Urban Residential/Commercial
Brighton (Boston)	7.0	Semi-Urban Residential
Waltham	14.9	Suburban Residential/Light Commercial
Stow	35.3	Semi-rural, open land; <u>Regional Background site</u> for Metro Boston

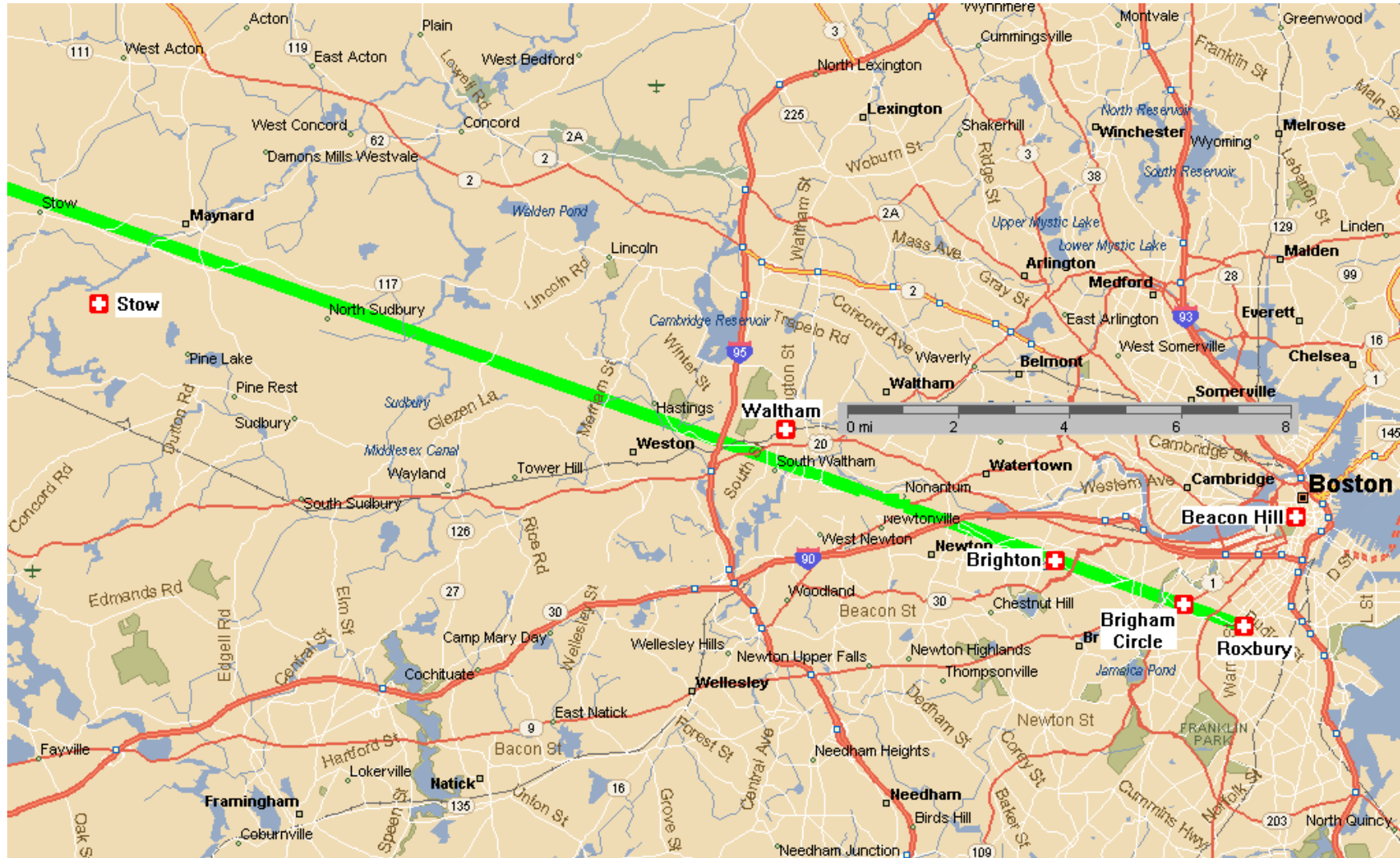


Figure: 6-Site Hourly BC Distributions, Dec. 20, 2002 to Sep. 09, 2003.

This set of boxplots updates the initial “pilot” analysis that was based on 2 months of winter 2003 data and shows the frequency distribution of hourly BC data.

Joy St. [Beacon Hill near State House] BC is essentially identical for all but perhaps the extreme hour values (95th percentile). The measurement methods used can not resolve concentration differences less than 10% between sites (site to site measurement bias).

The ratio between the mean or median BC from the highest urban sites and the Stow background site is between 3 and 3.5.

Brighton and Waltham BC remain essentially identical for these boxplot metrics, both about half of the highest urban sites.

Future statistical analysis will quantify the significance of this concentration gradient.

Figure: Diurnal BC, Six Greater Boston Sites Dec. 20, 2002 - Sep. 9, 2003.

This diurnal plot updates the initial “pilot” analysis that was based on 2 months of winter 2003 data and shows the temporal patterns of BC broken down by site and workday vs. non-workday. The much larger sample size and inclusion of both warm and cold weather seasons substantially decrease the uncertainty of in the initial pilot data interpretation.

The three “core” urban Boston sites show a distinct and strong rush-hour peak during workdays, and only a weak and indistinct peak during the same hours for non-workdays. This is consistent with expected traffic patterns and conclusions drawn by others previously. Note that Roxbury rush-hour peak BC is distinctly higher than Joy St. for both morning and afternoon, even though mean BC for these two sites is very similar.

Concentrations observed at Brighton and Waltham BC track remarkably well. The Stow background site shows no significant workday or non-workday diurnal pattern (errorbars are typically about 10% of the parameter value), confirming the lack of significant local traffic influence at that site.

This multi-season weekday/non-weekday diurnal analysis also provides increased confidence that BC is reasonably specific to local tailpipe aerosol, minimizing concerns related to potential interferences at these sites from other sources of BC such as oil-fired space heating and woodsmoke.

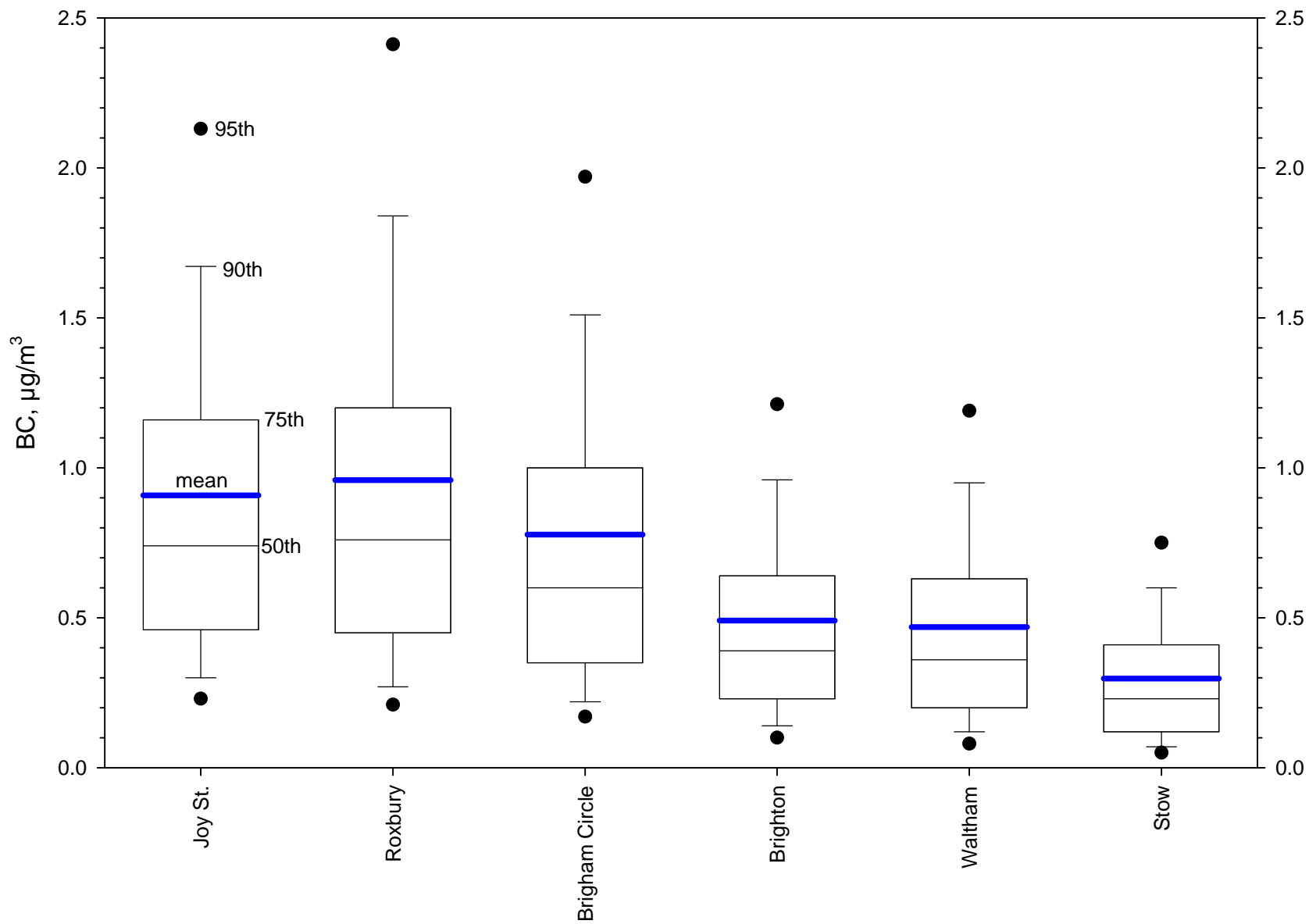
Figure: Workday/Non-Workday BC means across sites.

This plot shows the mean BC across sites by work/non-work day. As would be expected, the differences are highest (about 70%) at the sites with the most local traffic influence and highest BC levels, and decrease to about 10% at the Stow background site.

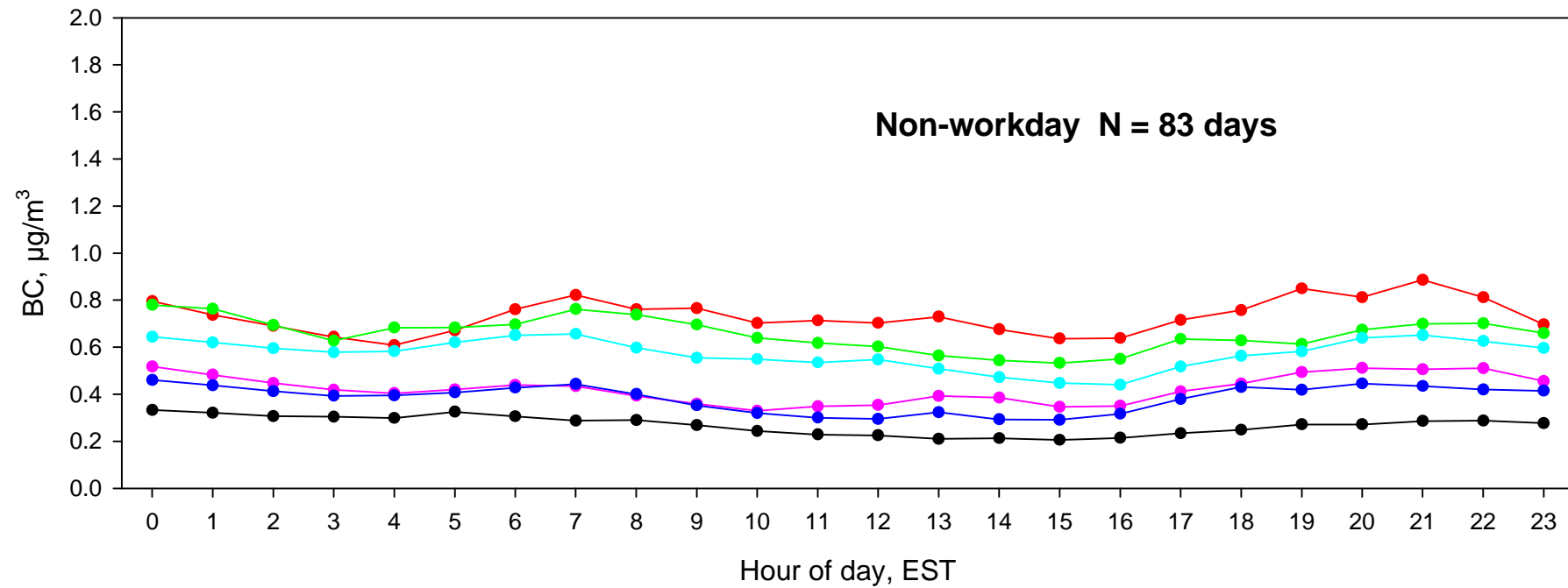
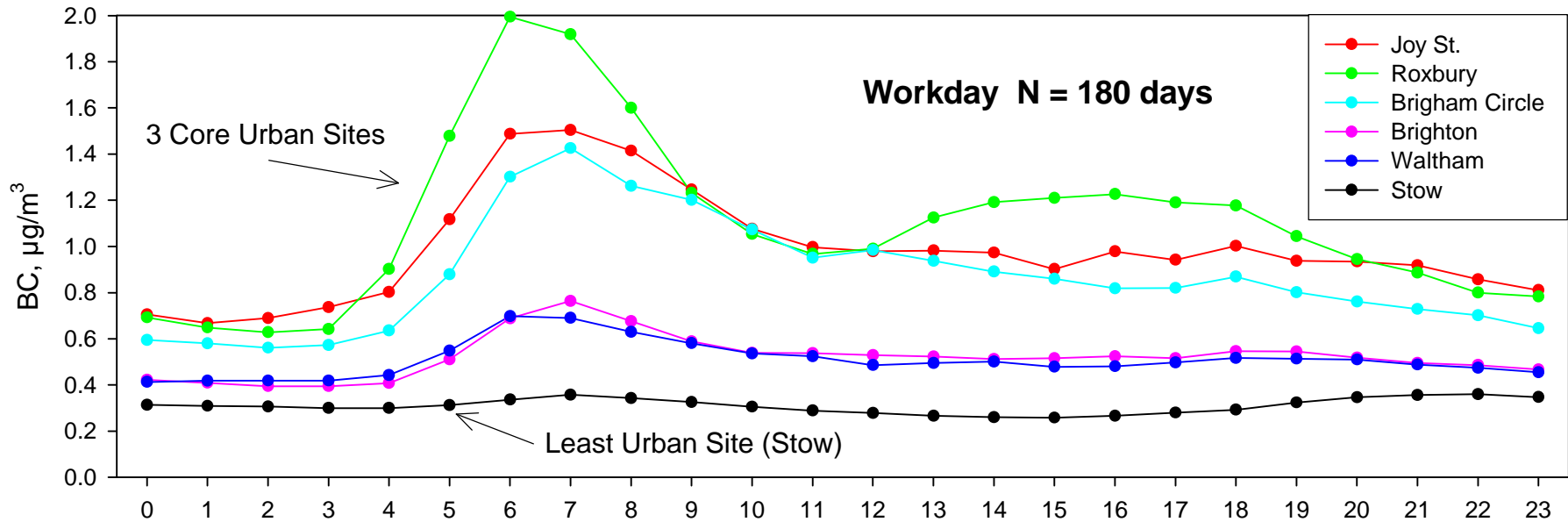
Figure: Cold vs. Warm season mean BC by site.

Although winter might be expected to have more of a local mobile source influence than summer (more and stronger inversions), these data show that all sites had substantially lower mean BC in the winter. This is most likely an artifact due to the unusually stormy weather in winter 2003; compared to Jan-Feb 2000-2002, Roxbury 2003 Jan-Feb mean BC was 37% lower.

6-Site Hourly BC Distributions, Dec 20, 2002 - Sep 09, 2003

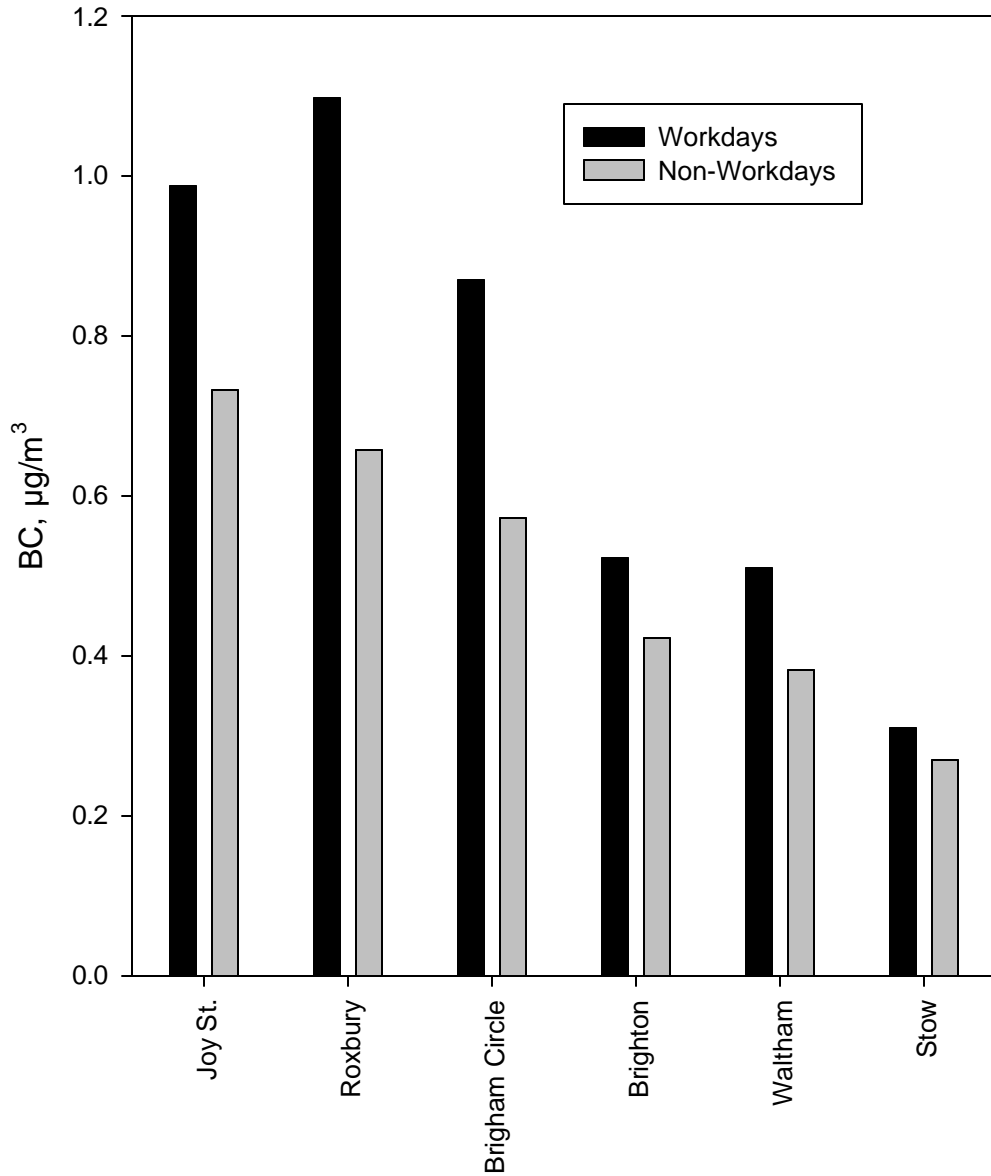


Diurnal BC, Six Greater Boston Sites Dec. 20, 2002 - Sep. 9, 2003

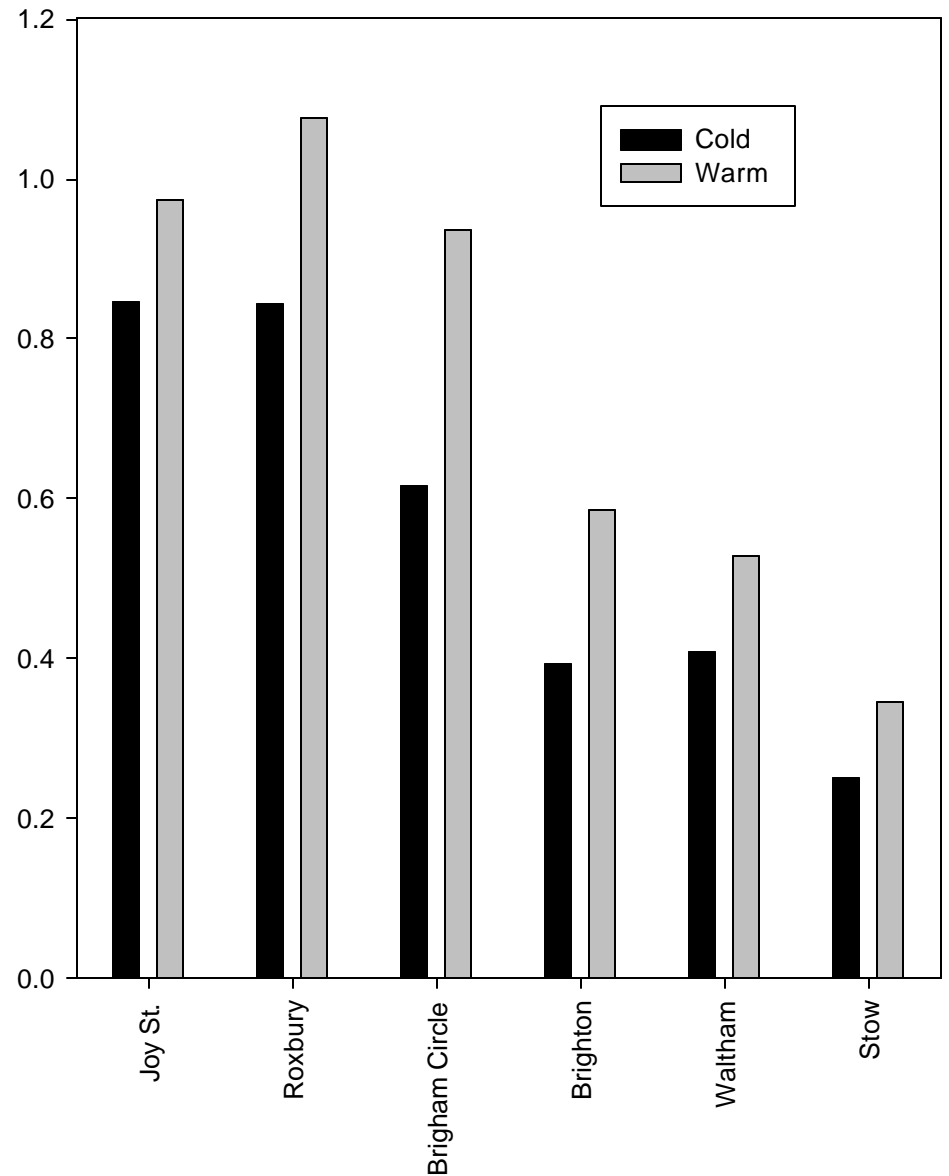


Note: This seasonal comparison is not valid due to a recently recognized strong seasonal bias in Aethalometer BC response

Workday / Non-Workday



Cold (Dec-Apr) and Warm (May-Sep)



“Neighborhood Scale” study: Summer 2003, for 2 months. 10 of 12 sites are in Boston; 9 are within a radius of 2.5 km; siting is representative of neighborhood scale (not hotspot/microscale) exposure. This table shows distance from the State House (Beacon Hill).

<u>Site Locations</u>	<u>Km</u>	<u>Site Description</u>
Joy St.	0.0	Urban Residential/Commercial. (Beacon Hill, near State House)
Pinckney St.	0.3	Urban Residential (Beacon Hill)
North End	1.1	Urban Residential/Commercial (near the I-93 Expressway)
South St.	1.0	Urban Commercial (near South Station bus and train terminals)
Hereford St.	1.9	Urban Residential (Back Bay)
Albany St.	2.4	Urban Commercial (BU School of Public Health)
South Boston	2.9	Urban Residential
Roxbury	3.5	Urban Residential/Commercial
Brigham Circle	4.0	Urban Residential/Commercial
Brighton	7.0	Semi-Urban Residential

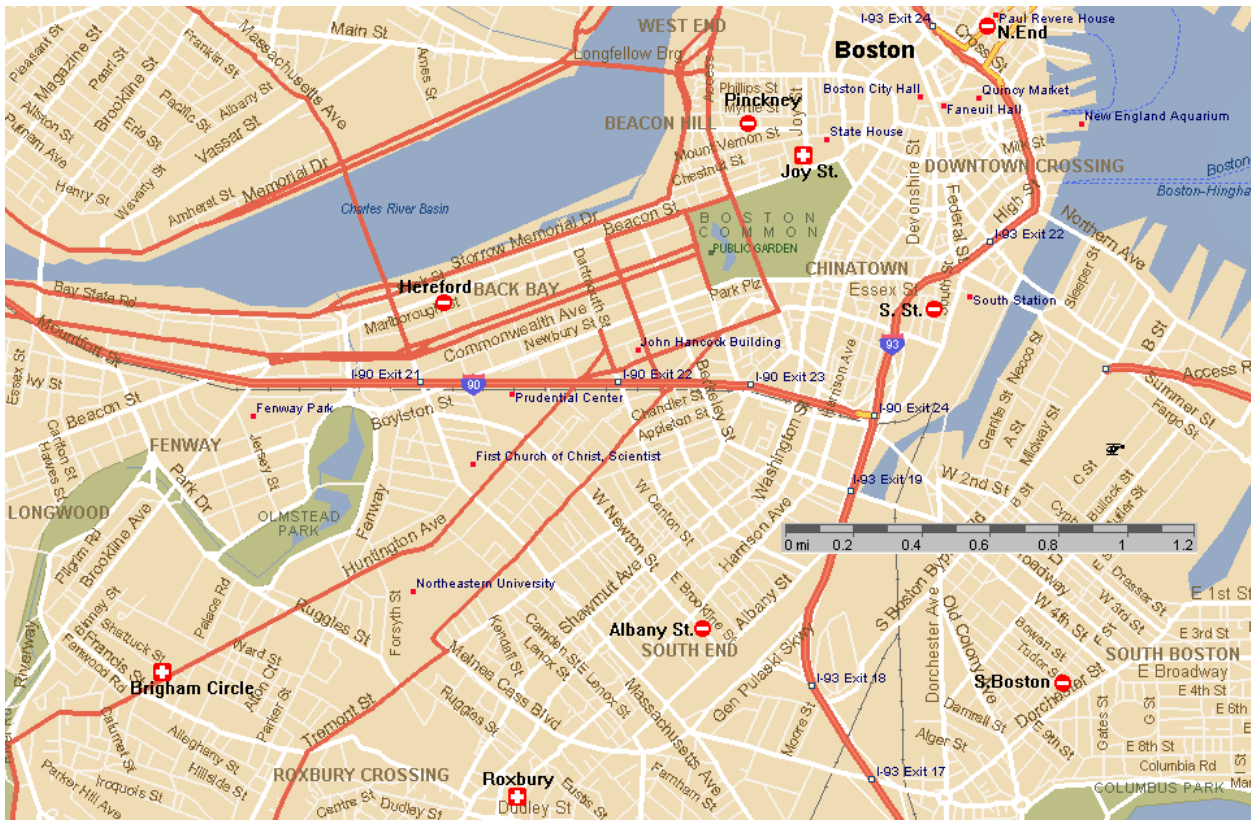


Figure: Summer 2003 1-hour Boston BC frequency distributions

This figure shows the distributions for all 12 BC monitoring sites, limited to days where all sites had data. Approximately 20 days are excluded due to two different sites that each had a 10-day period of missing data.

Mean BC for 8 of the 9 Downtown Boston sites during this study period was within 20% of $1.0 \mu\text{g}/\text{m}^3$, suggesting that with few exceptions, gradients for mean BC at neighborhood scale oriented sites in Boston are not substantial. The observed variation across sites could be influenced by variability in monitor siting, mobile source strength gradients, and microscale meteorology. Further data analyses will quantify the significance of these spatial BC concentration gradients.

The exception was the North End, with mean BC of $1.55 \mu\text{g}/\text{m}^3$, which might be due to proximity to the Expressway (southbound still above ground) and Callahan tunnel entrance, as well as Big Dig construction activity. This site is on the top of a 4-story building, 100 meters from the tunnel entrance and 200 meters from the southbound lane of the Expressway.

This question will eventually be answered, since this site is a permanent MA-DEP BC monitor; if post-Big Dig BC levels decline relative to the other two long-term BC sites in Boston (Roxbury and Brigham Circle) then it is likely that the local sources noted above were driving the observed elevated levels.

Note that the highest and lowest BC means for these 9 sites (North End and Pinckney St. on Beacon Hill) are only 1.3 km (0.8 miles) apart, with a ratio of 2.0.

Figure: Joy St., Pinckney St. and Roxbury, April - August 2003

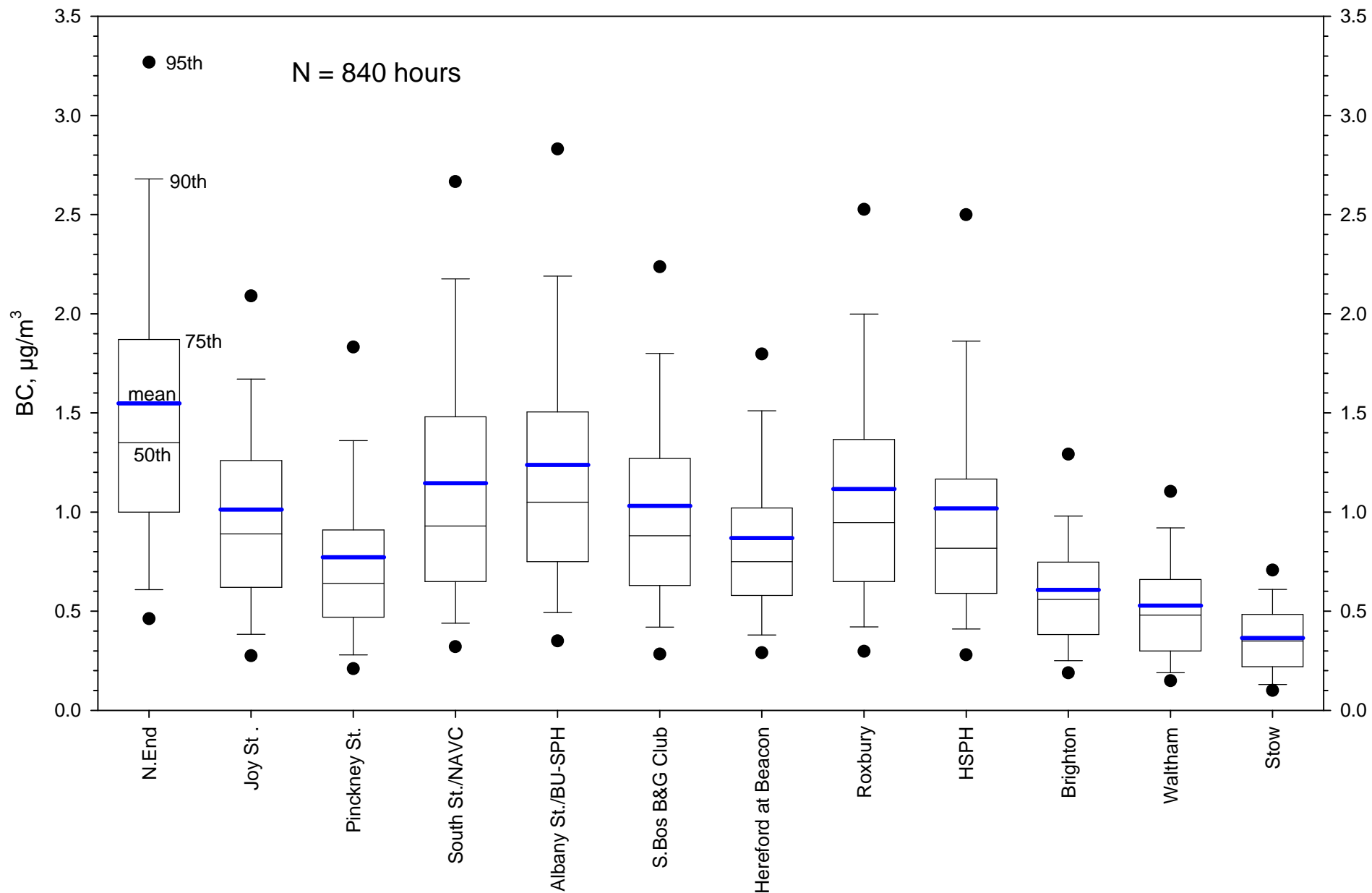
This box plot of frequency distributions examines in more detail (more sample days and thus a more stable relationship) the differences between the two sites on Beacon Hill (0.3 km apart) and Roxbury. One question raised by the pilot work last winter (and the rationale for the Pinckney St. site) was “If Joy St. is similar to Roxbury BC on average, what’s the cause and scale of the elevated BC on Beacon Hill?”. Pinckney St. is about as far removed as possible from through-traffic streets on Beacon Hill.

For this longer period, mean BC for Joy, Pinckney, and Roxbury are 0.94, 0.74, and $1.04 \mu\text{g}/\text{m}^3$ respectively. The ratio of Pinckney to Joy St. is 0.79 for both the mean and the 95th percentile values. This suggests that part of Joy St.’s BC is traffic that is very local (micro-scale), but gradients on this scale are not substantial.

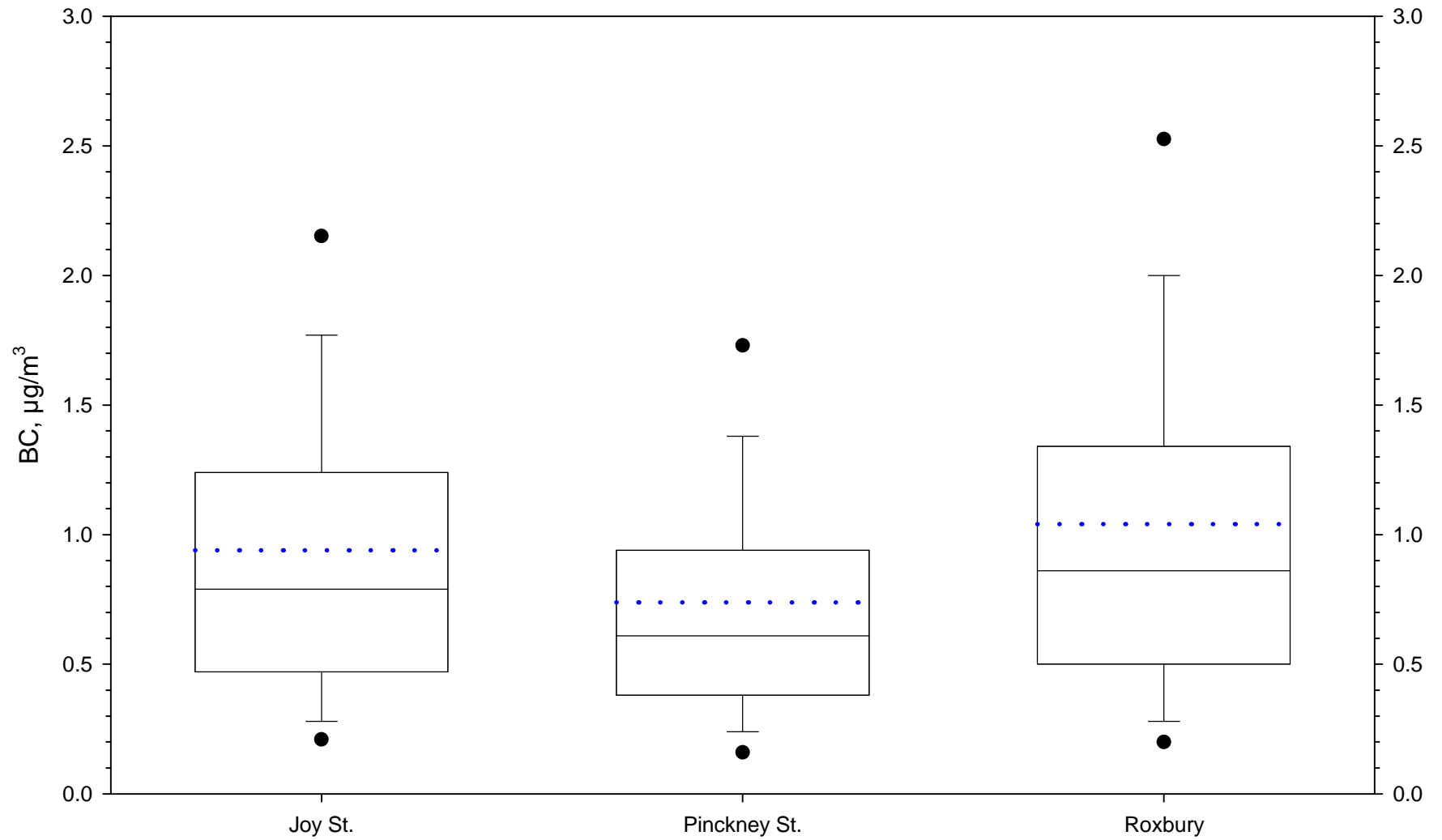
Figures: BC vs. Population Density, Winter 9-site and Summer Boston 10-site Regressions

The first plot shows data from the winter 2003 pilot study that suggested that population density might be a useful surrogate for average BC concentrations over a large spatial scale and with a large range of BC concentrations (R^2 of 0.81 if Brighton removed). The same analysis was performed on the 10 Boston sites for the summer neighborhood scale study. The second plot shows that for a smaller spatial scale with smaller dynamic range, population density can not be used to predict BC - the slope is actually reversed with higher BC associated with lower population density ($R^2 = 0.52$). This might be explained by core commercial and transit corridor areas such as the South St. site near South Station having lower population density but high traffic activity.

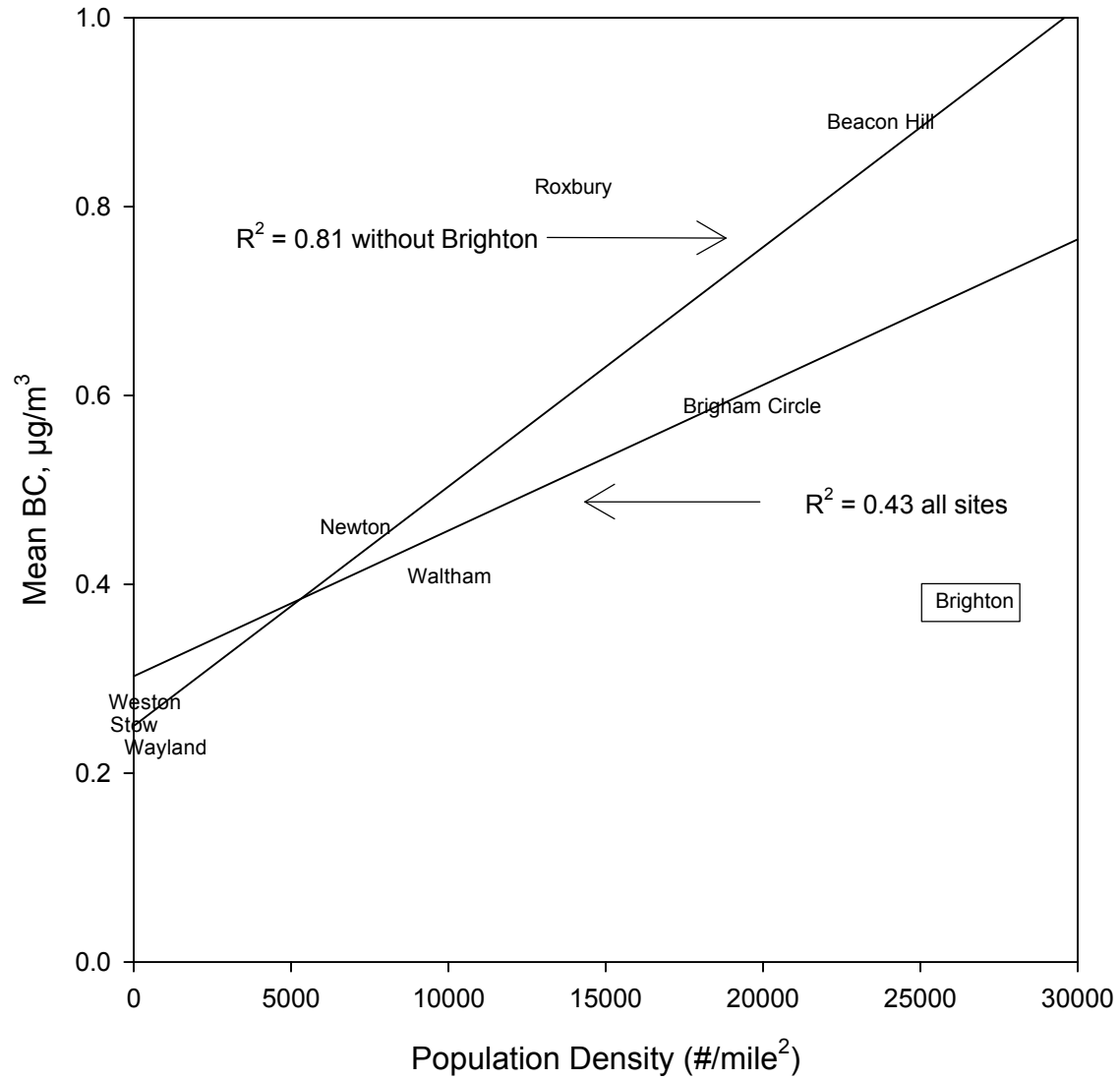
Summer 2003 1-hour Boston BC percentiles Limited to days with data from all sites



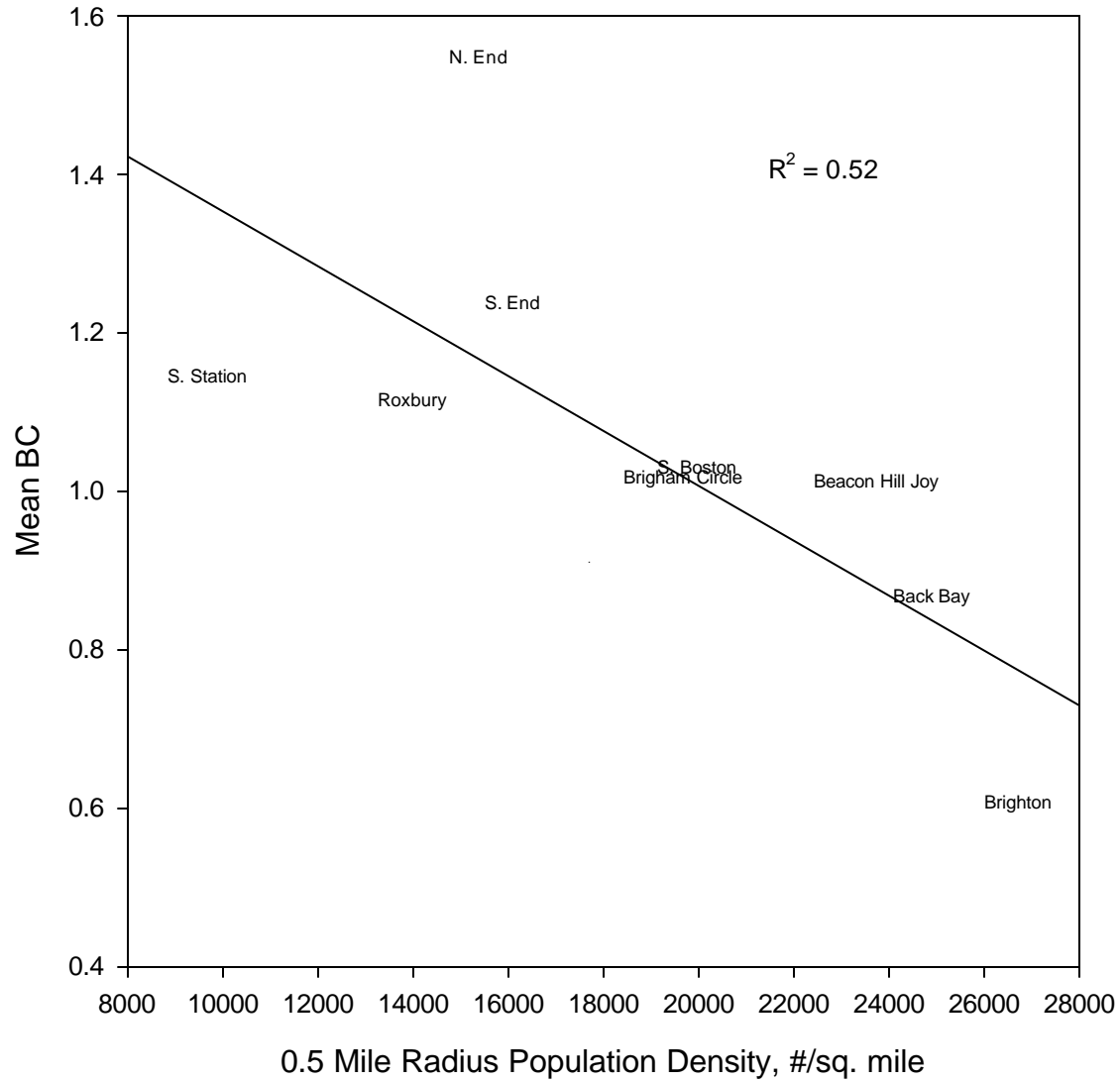
Hourly BC Distributions April 17 - August 30, 2003



Winter 2003 9-Site Pilot:
Mean BC vs. Population Density (1/2 mile radius)



Boston Neighborhood Scale BC vs. Population Density Summer 2003



Figures: Two time series “case study” examples

These two time series plots show examples of short term patterns and gradients of hourly BC across the Boston area. The first, July 13-15, is the 9 core Boston sites and the Stow background site. Tuesday July 15 was one of the dirtier days of the summer with several sites exceeding $4 \mu\text{g}/\text{m}^3$ BC for several morning hours. The ratio of these sites to the Stow background site for this peak period is approximately 10, similar to that observed during the winter pilot project.

The second time series is a 5-day period covering August 6 to 11. All 12 sites are included in this plot. Thursday the 7th shows a distinct evening rush-hour peak, not a common feature. The very high peak in South Boston on Friday the 8th at hour 07 EST is substantially higher than other sites, although the other urban sites peak at the same hour. This site could have been influenced by local marine diesel sources, since major Boston Harbor piers are about 1 mile away to the NNE. Winds at Logan Airport were NNE to NE at a few miles/hour during this time. That peak hour was influenced by two contiguous very high 5-minute BC values (22 and $13 \mu\text{g}/\text{m}^3$); without those values the mean for this hour is $6 \mu\text{g}/\text{m}^3$, more similar to the other sites.

Both of these time-series plots show a very distinct “clean Sunday and dirty work-day” effect.

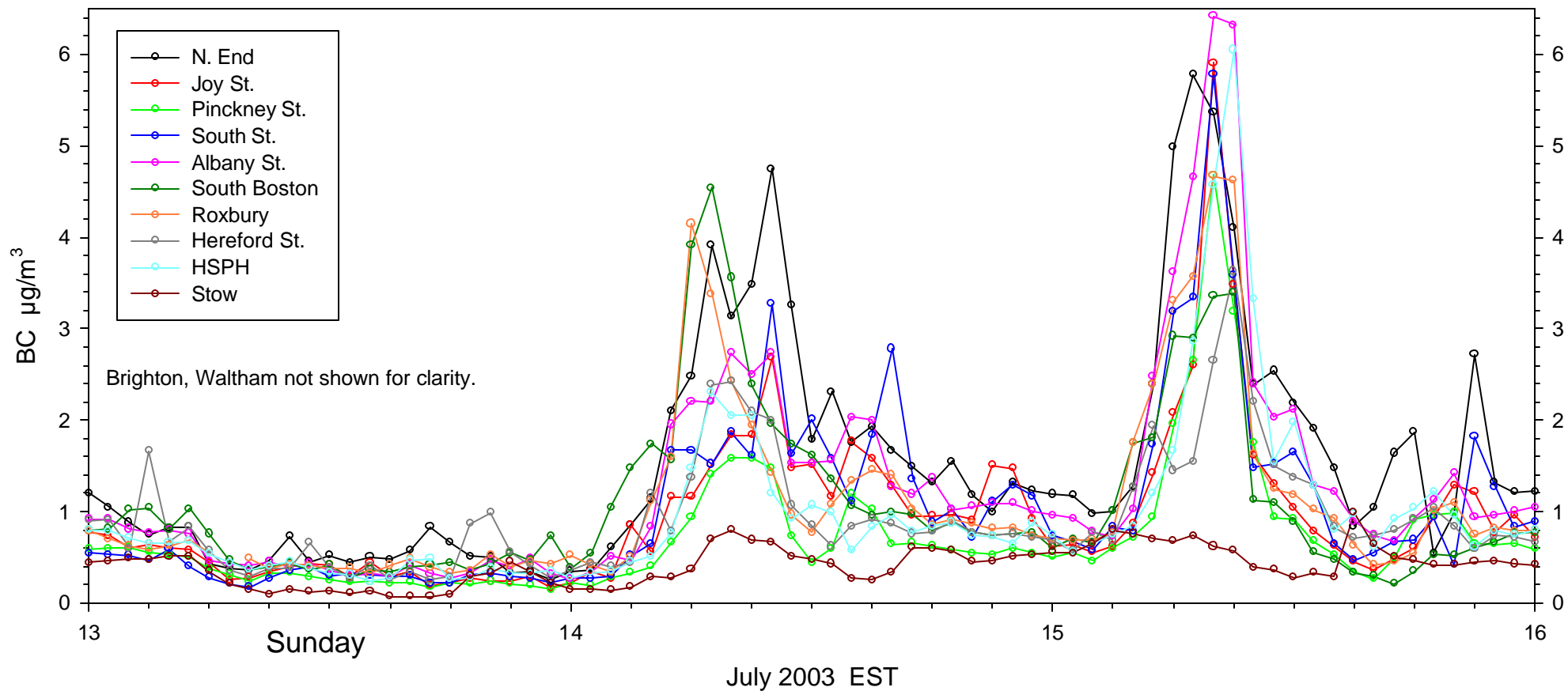
Figure and Table: August 6-11 case study: hourly scatter plots and correlation matrix

Scatter plots for hourly BC for six site-pairs and a table for all site pairs during this August 5-day period are shown as examples of the short-term relationships across different spatial scales. There is a wide range of correlation, from reasonably high ($R^2 = 0.85$ for the two Beacon Hill sites) to very low (0.08 for Stow and South St.).

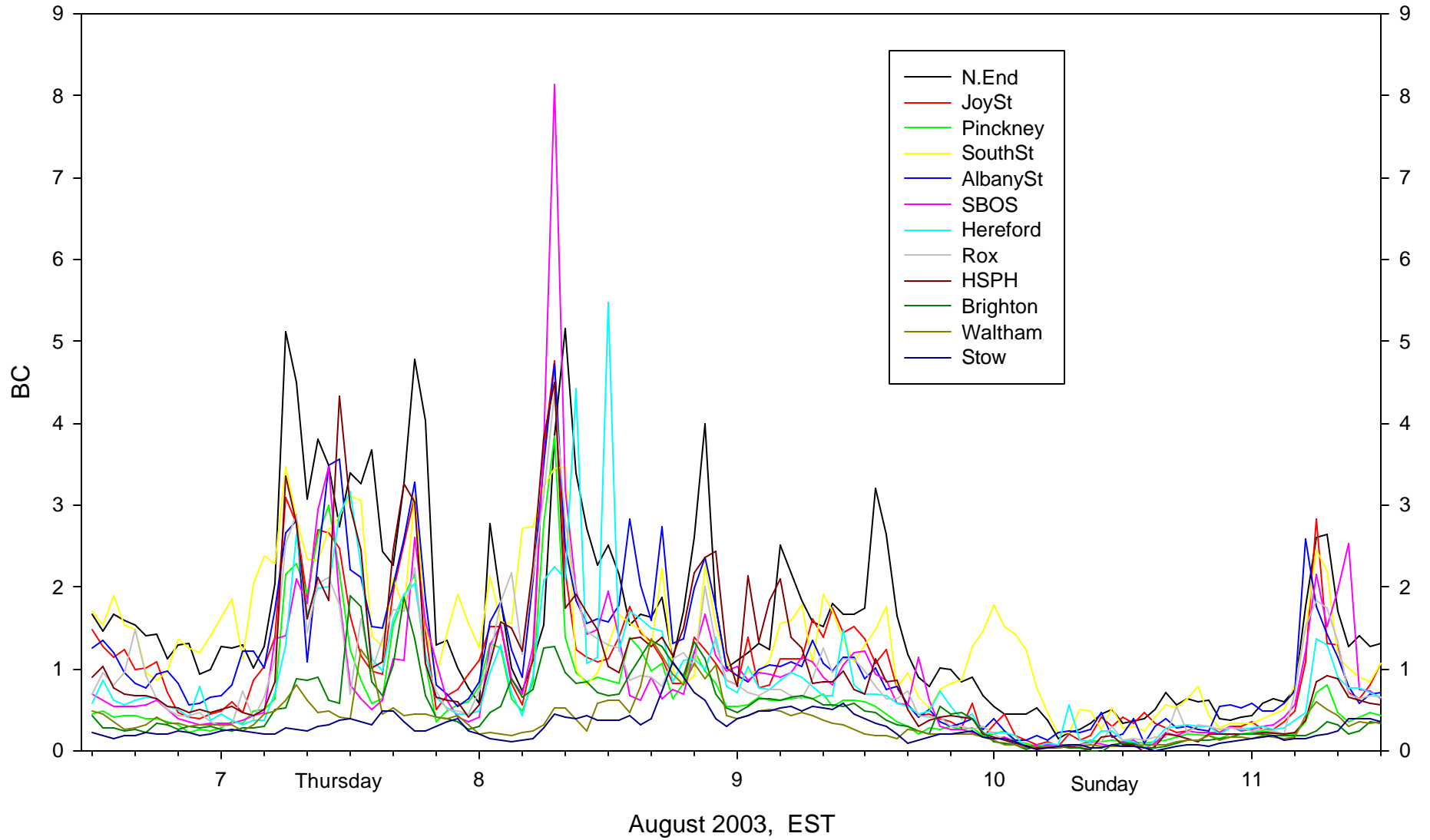
Distance between sites is not always a predictor of how well they are correlated. Joy St. and Roxbury (3.5 km apart) have an R^2 of 0.72, while Hereford St. and South St. (2.4 km apart) R^2 is 0.28 for the same time period. Some sites, such as South St. and especially Hereford St. are not well correlated with other urban sites. Others (Roxbury, Joy St., Albany St.) seem to be reasonably well correlated with most urban sites; these three sites also have means that are very similar (within a few percent).

The scatter plots show some interesting patterns for some site-pairs. The two downtown Boston sites with the lowest mean BC (Hereford St. and Pinckney St.) are well correlated when levels are below about $1 \mu\text{g}/\text{m}^3$ BC. But when levels are high at either site, they tend to be decoupled temporally. South Boston and South St. are clearly influenced by different sources. North End and Stow, the highest and lowest sites in the study, are pretty well decoupled at this time scale; note that here the axes are not scaled the same; the bottom line is the 1:1 line. Essentially all hours at North End are at or above the Stow BC levels.

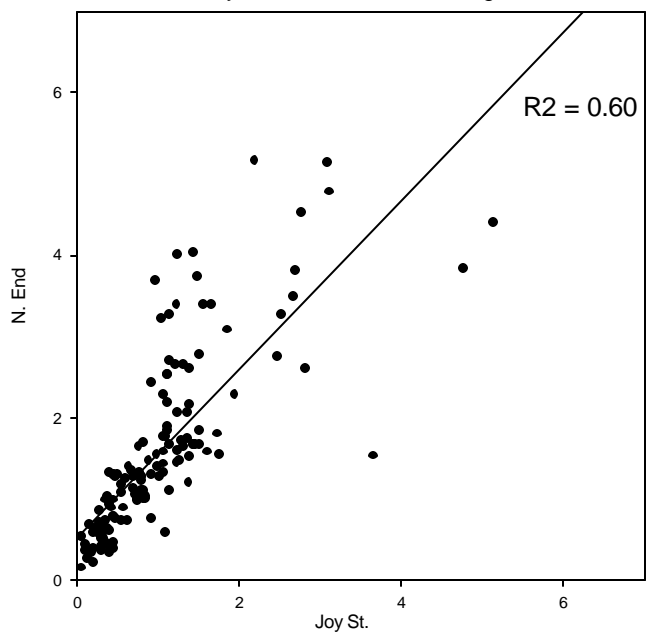
Boston BC event case study July 13-15 2003



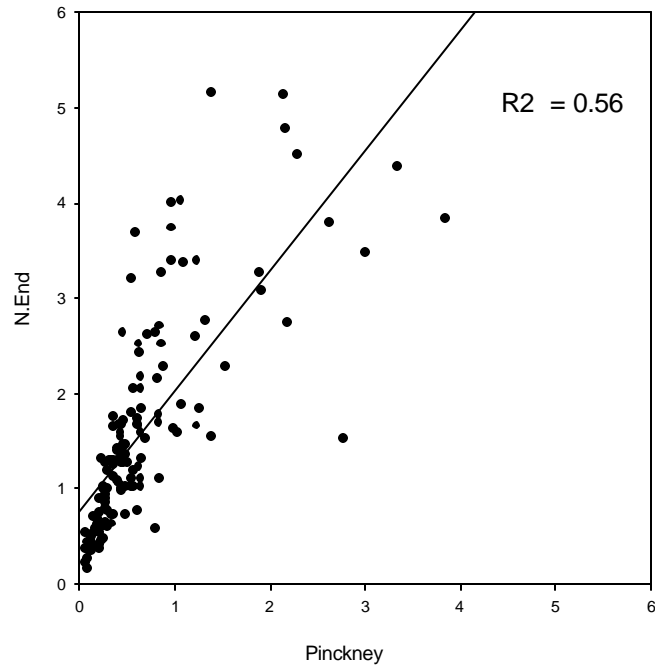
12-Site BC, 1-hour means
Aug 6 - Aug 11, 2003



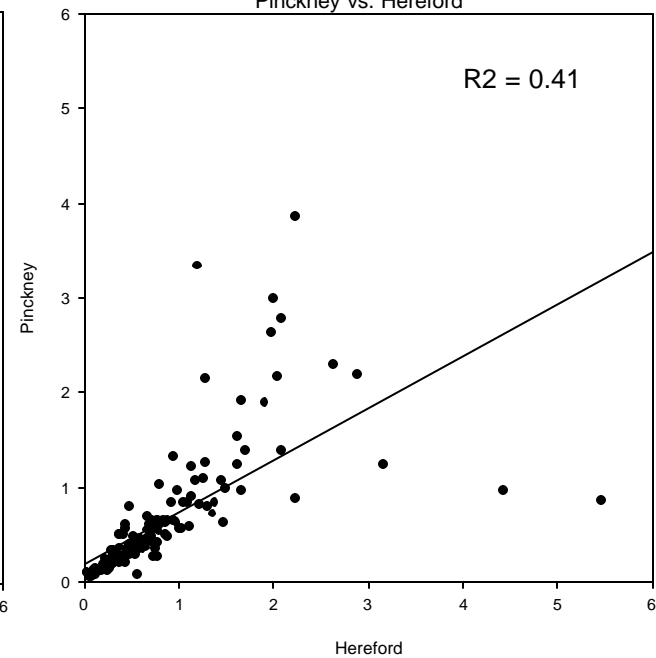
N.End vs. Joy St. 1-hour mean BC, Aug 6-11 2003



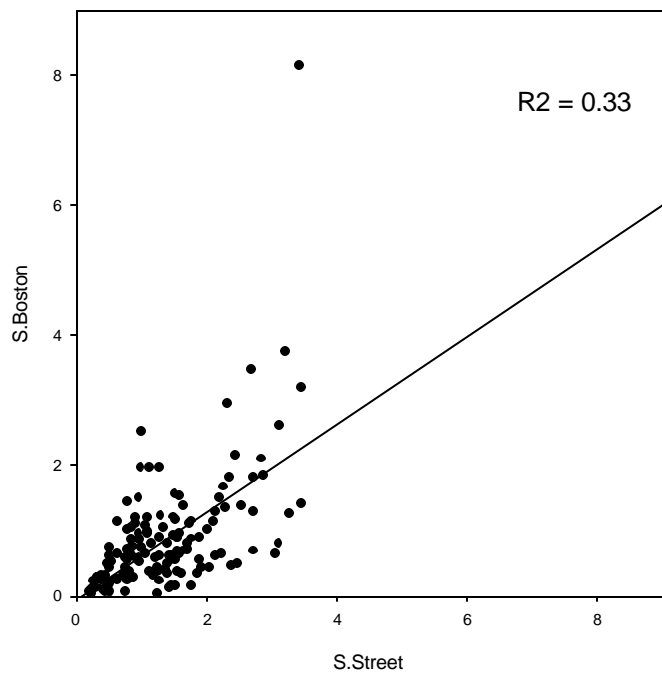
N.End vs. Pinckney



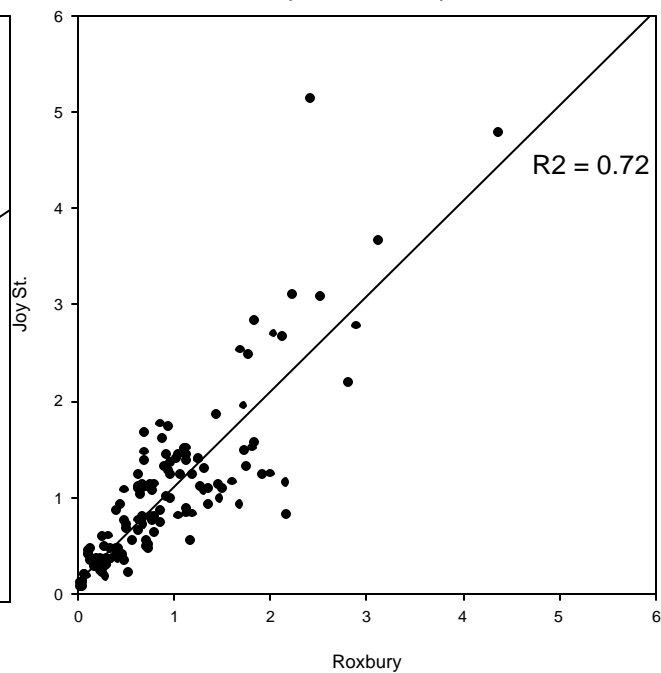
Pinckney vs. Hereford



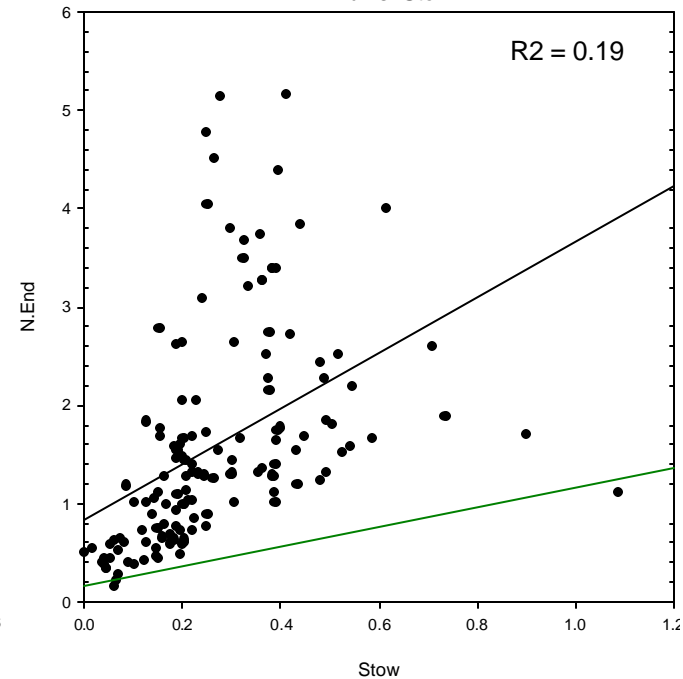
S.Boston vs. S. Street



Joy St. vs. Roxbury



N.End vs. Stow



1-hour R² Matrix, August 6-11 2003

R2 .70 or higher

R2 .50 to .69

	N.End	Joy St.	Pinckney St.	S.St	Albany St.	S.Bos.	Hereford St.	Rox	BrigCir	Brighton	Waltham	Stow
N.End	x	.60	.56	.54	.63	.40	.46	.59	.51	.46	.30	.19
Joy	.60	x	<u>.85</u>	.54	<u>.79</u>	.62	.36	<u>.72</u>	.59	.38	.18	.14
Pinckney	.56	<u>.85</u>	x	.51	<u>.79</u>	.64	.41	.69	.67	.44	.19	.13
S. St.	.54	.54	.51	x	.57	.33	.28	.53	.48	.35	.21	.08
Albany	.63	<u>.79</u>	<u>.79</u>	.57	x	.55	.46	<u>.73</u>	.66	.51	.31	.19
S.Bos	.40	.62	.64	.33	.55	x	.34	<u>.70</u>	.48	.24	.09	.10
Hereford	.46	.36	.41	.28	.46	.34	x	.41	.48	.45	.27	.17
Rox	.59	<u>.72</u>	.69	.53	<u>.73</u>	<u>.70</u>	.41	x	.63	.40	.20	.14
Brig.Cir	.51	.59	.67	.48	.66	.48	.48	.63	x	.59	.22	.22
Brighton	.46	.38	.44	.35	.51	.24	.45	.40	.59	x	.44	.39
Waltham	.30	.18	.19	.21	.31	.09	.27	.20	.22	.44	x	.38
Stow	.19	.14	.13	.08	.19	.10	.17	.14	.22	.39	.38	x

Conclusions and Preliminary Findings

Substantial gradients in BC exist over a 35km scale

Mean BC varies by a factor of 3.5 from downtown Boston to the regional background site

==> Much larger factor for sub-daily event periods: 10x or more

These data indicate that the neighborhood spatial scale of “urban excess” PM_{2.5} for Boston is limited to approximately 10 miles from downtown. This is important from both an air toxics exposure and control strategy perspective.

Core urban area: BC levels at all neighborhood-scale sites are elevated relative to the background site, but urban gradients are not distinct for most of these sites.

Short-term (1-hour) correlations across these sites range from very good to poor; some urban sites are much better indicators of the general downtown area than others.

BC appears to be a reasonable indicator of local “tailpipe” aerosol, not highly specific to diesel or on-road vs. off-road sources. Winter space heating and woodsmoke do not appear to be significant interferences in the urban area.

Limitations of this preliminary report

This study does not assess worst-case “hotspot” exposure scenarios in either urban or non-urban settings, since it is based on neighborhood scale, not mid-scale or micro-scale (“roadway”) monitor siting. As such, these findings should not be construed to suggest that meaningful exposures do not occur outside of urban areas. Potential instrument bias has not yet been removed that could effect gradient assessments. ANOVA analysis to assess the significance of gradients has not yet been performed, and may result in modification of these preliminary conclusions.

Limitations of this study include semi-randomized monitor siting and monitor-to-monitor bias that are not accounted for at this time. Meteorological conditions that may influence observed BC concentrations at any given site have also not been taken into account in this preliminary analysis. The statistical significance of observed gradients will be characterized using ANOVA analyses once the final year-long data set is available.

Appendix C: Change in Reported BC Without and With Spot Loading Correction for Two Sites in 2003-2004 (Scatter and time-series plots for HSPH and North End).

Appendix C.

Change in reported BC without and with spot loading correction for two sites in 2003-2004.

The Aethalometer BC has an artifact that results in under-reporting of BC concentrations that varies over time on the scale of hours to seasons. This artifact was examined for the HSPH and N.End sites during 2003 and 2004 using both scatter and time-series plots. Figures 1 and 2 are regressions of correct vs. original (raw) 1-hour BC data for these two sites. There is a bias of approximately 20 percent at both sites, with errors approaching a factor of two for some hours.

Figure 1. HSPH Corrected vs. Raw 1-hour BC, 2003

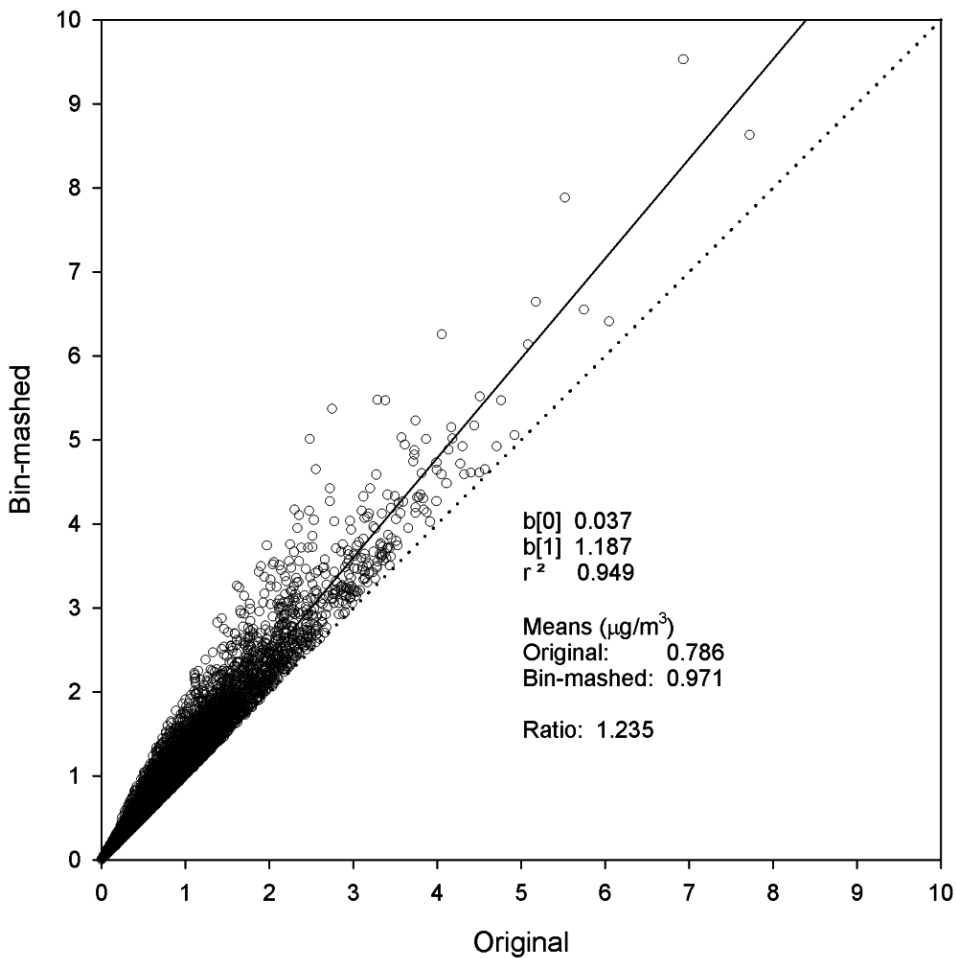
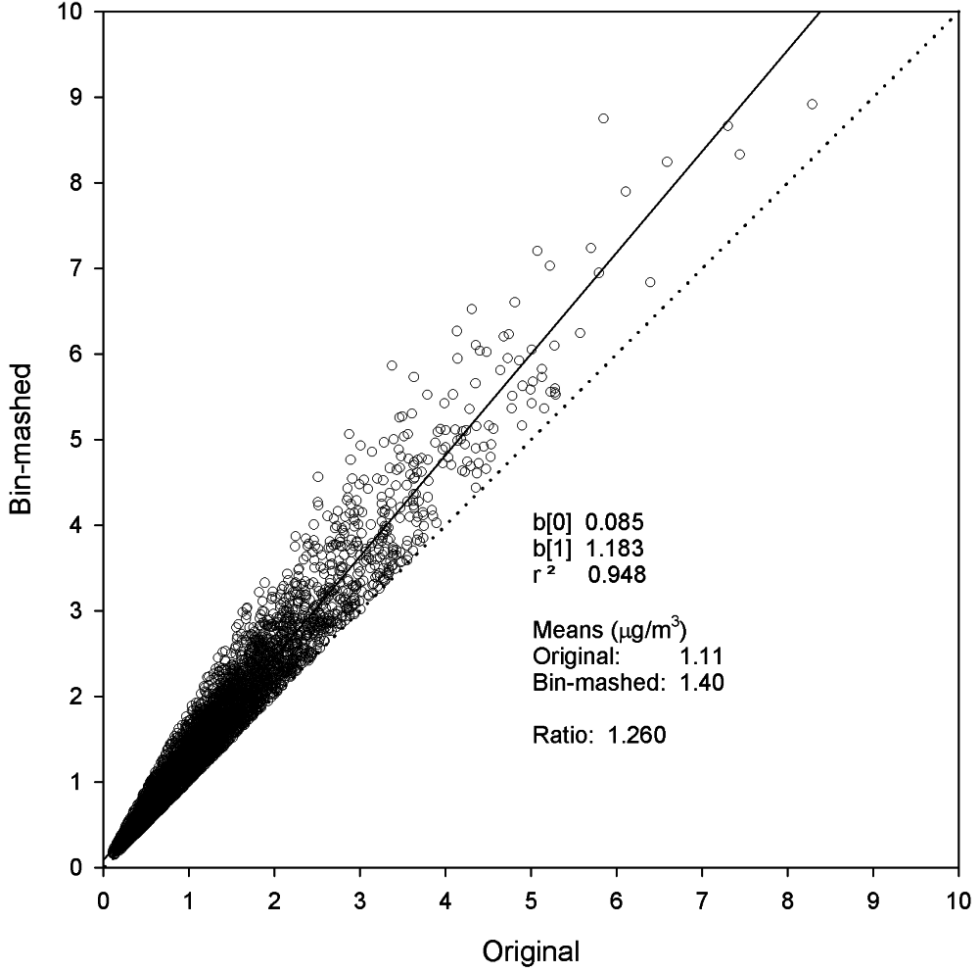


Figure 2. North End Corrected vs. Raw 1-hour BC, 2003-2004



Figures 3 and 4 show this artifact error as a function of time over the same periods. The red line is the ratio of hourly corrected to original BC concentrations, and changes both at hourly time-scales (from aerosol loading during measurements on a single spot on the sample tape) and seasonal scales (from changes in the aerosol composition).

Figure 4. HSPH 1-hour BC and Ratio of Corrected to Original BC Concentration

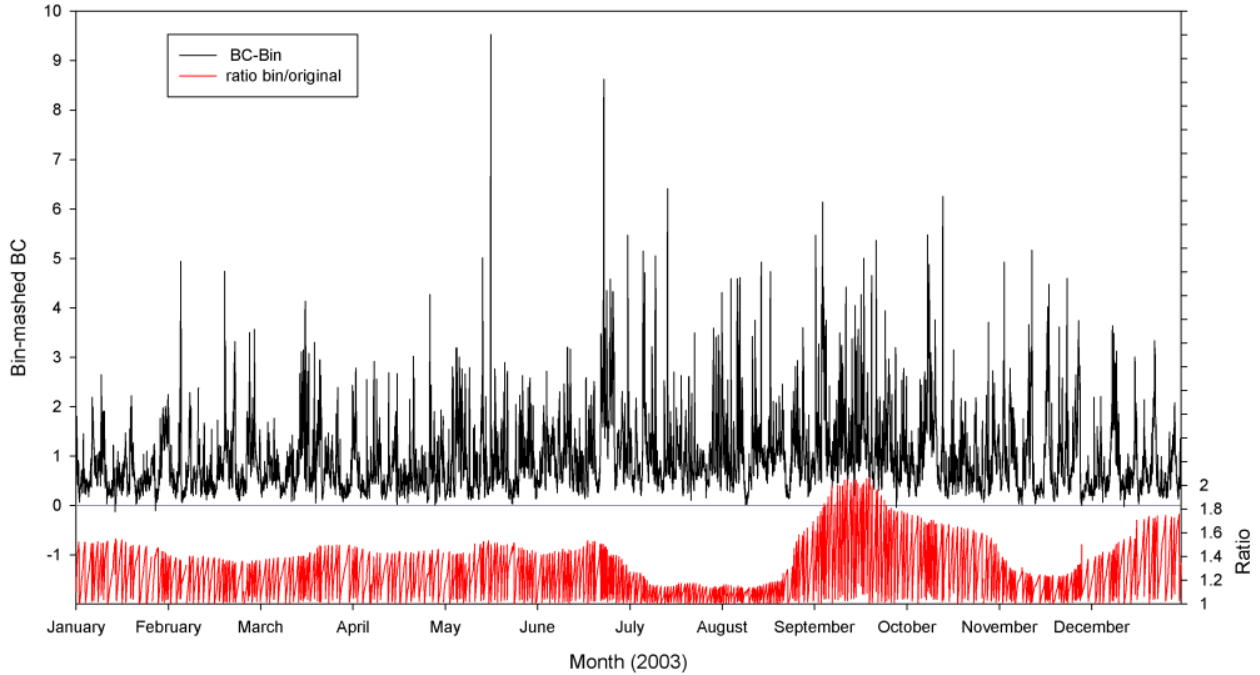
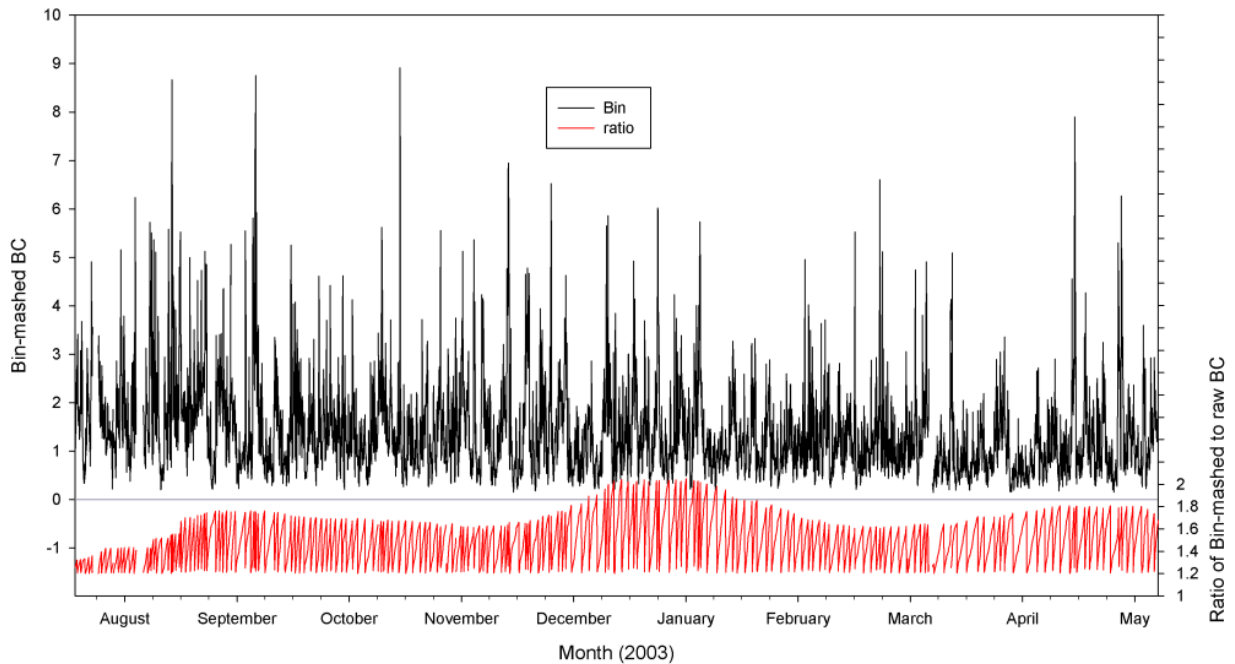


Figure 3. N.End 1-hour BC and Ratio of Corrected to Original BC Concentration



Appendix D: Sonoma Technologies' September 30, 2011 Draft Memo to U.S. Environmental Protection Agency on the Aethalometer Spot Loading Artifact, Including Examples of Difference Between Raw and Bin-corrected BC Data, and Trends Analysis of the HSPH Countway BC Data.

****DRAFT** Technical Memorandum**

September 30, 2011

STI-910212-4217

To: Dave Shelow, Neil Frank, EPA OAQPS

From: Steve Brown, Jay Turner (Washington University in St. Louis), George Allen (NESCAUM)

Re: Development and implementation of an updated validation tool for Aethalometer measurements: data acquisition and processing

This draft technical memorandum is part of the deliverable for AIRNow Work Assignment (WA) 2-12, Task 6. It includes key findings on differences between raw and processed (adjusted) Aethalometer data and a summary of the raw data acquired and processed as part of this project. Raw Aethalometer data are biased by optical saturation effects. In an effort to understand these effects and to correct them, the Washington University Air Quality Laboratory (WUAQL) Aethalometer Data Masher was modified as part of this WA and used to process raw data from sites throughout the U.S. Along with the Data Masher software and its user's guide, which provides the technical details of the correction algorithms in the Data Masher, this technical memorandum summarizes the current correction methodology and data changes as a result of this correction.

Key Findings

Key findings from this work include the following:

- There are two major factors that influence the bias caused by the optical saturation (particle loading) effect. First, the effect is influenced by aerosol composition with a highly scattering aerosol reducing the effect. At many locations, the aerosol composition exhibits strong seasonality and in general the adjustments will be higher in locations and during periods with low sulfate, such as the western U.S. and in wintertime.. Second, the bias increases as the particle deposit accumulates on the Aethalometer filter tape. Thus, changes in the maximum attenuation (ATN) setting influence the extent of error. The maximum ATN is a user-controlled parameter; sometimes it is intentionally changed by the user while other times it is unintentionally changed (e.g., following instrument repair when the instrument is returned with default settings, or by turning on or off the ultra-violet (UVC) channel on a 2-channel instrument).
- For default maximum ATN settings, the magnitude of Data Masher adjustments to the Aethalometer black carbon (BC, measured at 880 nm) data are greater for 1-channel instruments (BC only) than for 2-channel instruments (BC and UVC). The majority of data sets collected in this project from state and local air monitoring agencies are for

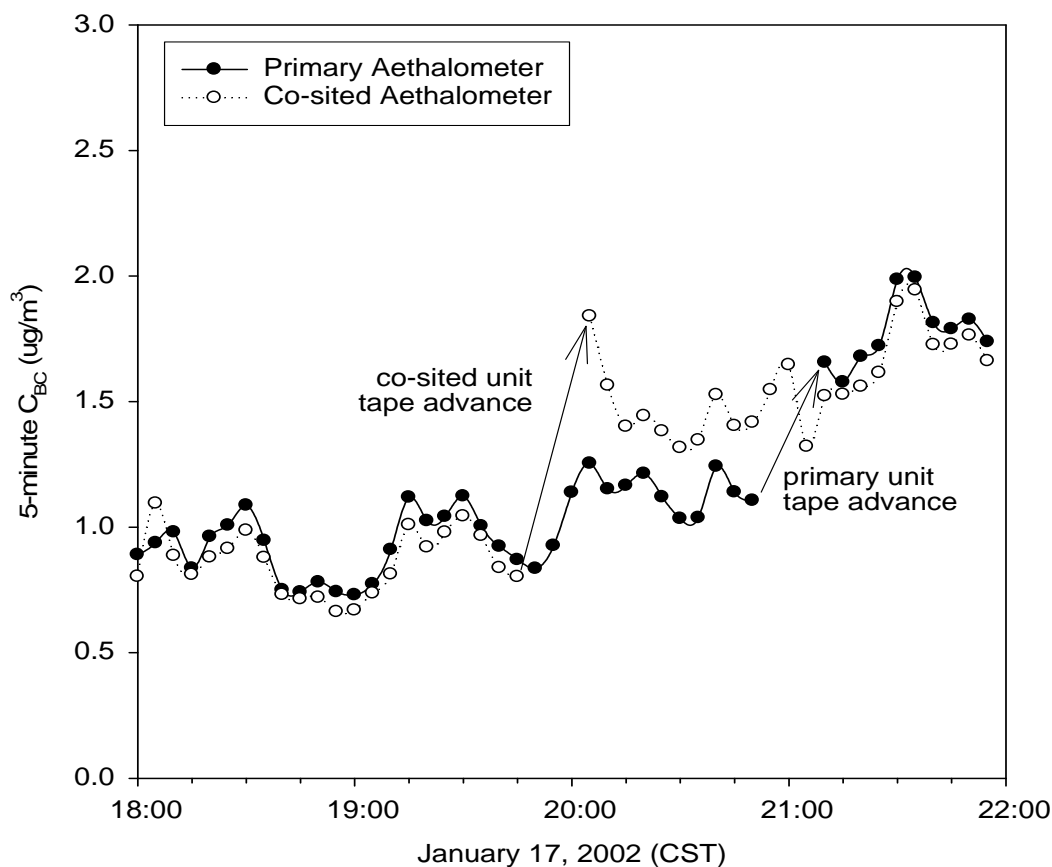
2-channel instruments. The adjustments to the UVC channel data are larger relative to the BC data.

- The algorithm implemented for this project applies smoothing over several tape advances (“spots”). While most analyses were performed using 30-spot smoothing, larger smoothing ranges may be necessary when the concentration adjustment is small (e.g., low maximum ATN and small optical saturation parameters).
- The algorithm implemented for this project (the “bin” method, described below) typically produces results similar to the legacy “gap” method. However, the bin method has the advantage of providing quantitative metrics of the quality of the adjustments. Also, the time series of adjustments sometimes significantly differs between the two methods when data are very noisy (real noise or instrument noise). In such cases, there is anecdotal evidence that the bin method outperforms the gap method. Data from older Aethalometers (AE16 or AE21) tend to be noisier than the current model (AE22).

Introduction

Filter-based optical methods for estimating ambient particulate matter BC concentrations suffer from a mass loading effect whereby the instrument response decreases with increased BC loading. One such instrument, the Aethalometer, continuously collects particles onto a filter and measures the wavelength-dependent transmission of light through the deposit. The mass loading effect is most commonly observed as a step discontinuity in the reported concentration when advancing the Aethalometer filter tape to deposit aerosol onto a clean filter spot instead of a particle-laden filter spot. **Figure 1** shows an example of this effect using collocated Aethalometers. Initially there is good agreement between the reported BC values, but after the tape advances for the co-sited unit, it reports concentrations that are systemically higher than the primary unit. These higher values after the tape advance are actually closer to the true BC concentration, while the data prior to the tape advance are biased low. Later in the time series, the tape advances for the primary unit and subsequent data again exhibit good agreement. The mass loading effect is not governed solely by deposited absorbing (e.g., soot) aerosol. It is also influenced by the abundance of internally- or externally-mixed scattering aerosol (e.g., sulfate) in the ambient air that co-deposits with the absorbing black carbon particles.

Figure 1. Four-hour time series of 5-minute black carbon concentration data for collocated Aethalometers, East St. Louis. Each instrument exhibited one tape advance during this period. The collocated unit data stream was adjusted for instrument-to-instrument bias.



Several approaches have been proposed to adjust the data for these artifacts. For the Aethalometer, one approach uses the difference in reported concentration before and after a filter tape advance to estimate the artifact. Another approach is to perform a regression of the reported concentration on the attenuation using all of the data over a specified time period. Both of these approaches assume the true BC concentration is not changing over the time period of interest (across the tape advance for the first approach, for the entire time period used for the regression in the second approach). Thus, additional temporal aggregation of the data or smoothing of the artifact estimates is needed to dampen the effect of this limiting assumption. As part of this project, the Aethalometer data post-processing program developed at Washington University at St. Louis (the WUAQL Data Masher) that is publicly available to the air quality measurement community has been revised to include an algorithm for adjusting the raw data using both of these approaches. The gap-based algorithm was implemented for prior projects whereas this project focused on implementing a regression-based algorithm that uses all of the data. This latter algorithm has been implemented in the current version of the program and was used throughout this project.

Methods

The loading effect leads to the Aethalometer-reported concentration decreasing with increased ATN (a measure of the light absorption by the deposited aerosol) even when the measured aerosol has a constant BC concentration. The goal of the algorithm is to correct for the ATN effect so that, on average, concentration is independent of ATN. The loading effect is assumed to follow the form:

$$BC_t = BC_r (1 + k \cdot ATN) \quad (1)$$

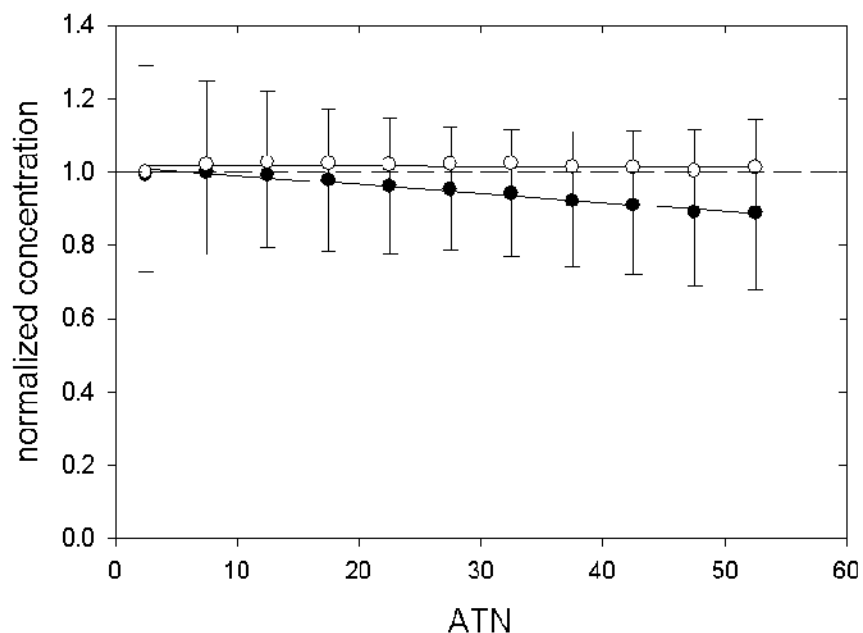
where BC_t is the true (adjusted) concentration, BC_r is the Aethalometer-reported concentration, and k is an empirical parameter that, in this algorithm, is obtained from the regression of concentration on ATN. The parameter k is termed the “optical saturation parameter” and is zero for no adjustment to the raw data. For example, at $ATN = 50$, a k -value of 0.010 would adjust the BC concentration upward by 50%. The remainder of this section summarizes the methodology. More details are provided in the Data Masher User Guide.

Considerable effort was invested to develop a methodology that would stabilize the analysis across a range of data sets. Simply binning the data by ATN was insufficient because some high-concentration extreme values can have considerable influence on the regression. After testing several approaches, the following methodology was adopted. The first step is to bin the raw concentration data (typically collected on a time base of 5 minutes but sometimes 1 minute) by attenuation. The algorithm allows the user to specify either a fixed bin width (e.g., 5 ATN units) or an approximate number of bins to be equally distributed over the attenuation range observed for each channel (Aethalometers can have 1, 2, or 7 channels corresponding to the number of wavelengths for which absorption is measured). The default is a fixed bin width of 5 ATN units. Tape advances are followed by an instrument stabilization period with no data reported. The time duration of all data gaps are identified and the mode gap size (e.g., three consecutive 5-minute missing records) is used to flag the tape advances in the time series. If the second mode gap occurs at a frequency of at least 5% of the mode tape advance gap frequency, the user is given the option to also consider the second mode gap as tape advance events.

Data recorded between each tape advance are stratified into the ATN bins and the mean concentration is calculated for each bin. These concentration values are then normalized to the average concentration over all the bins. These steps ensure that the time series data between each tape advance is given similar weight in the regression. Next, these normalized, binned concentrations are aggregated over a user-specified number of tape advances w (default $w = 30$) and the median normalized value is calculated for each bin. Bin-specific median values are regressed on ATN to determine the empirical data adjustment parameter, k . **Figure 2** shows an example of the data used for the regression of normalized concentration on ATN (solid circles). Error bars denote the interquartile ranges about the median normalized concentrations. The open circles show the median normalized concentrations estimated from Equation (1) using the fitted k -value. Ideally these values should be tightly clustered around the horizontal line at

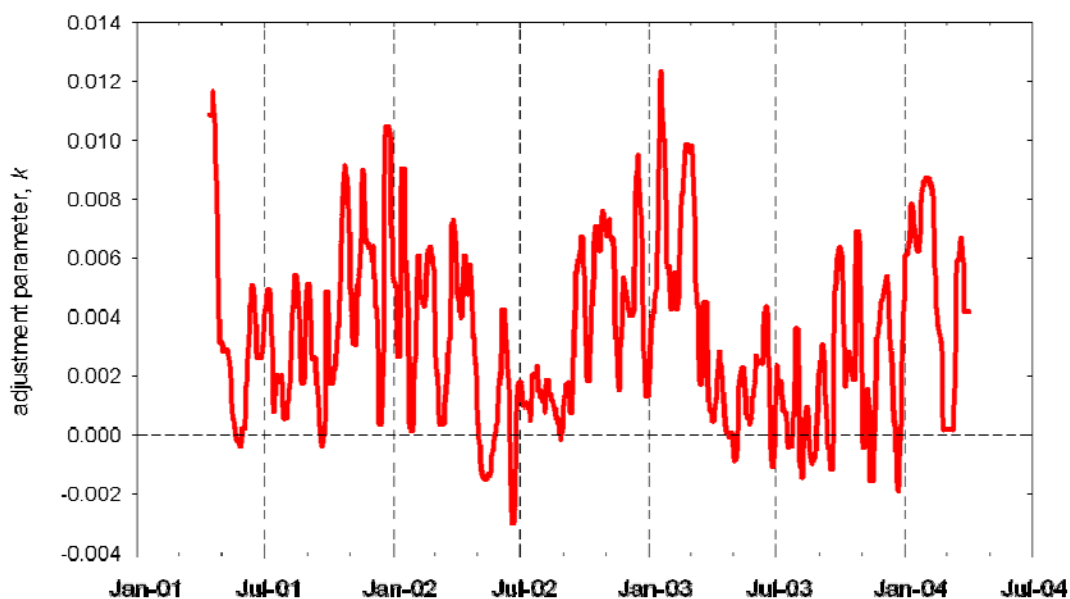
normalized concentration of unity (i.e., the ATN dependence of concentration has been removed); in this example they are biased high by less than 2%, which is a minimal bias.

Figure 2. Aethalometer BC concentration for April 2001 through March 2004 at East St. Louis, binned by ATN and normalized within each tape advance. Solid circles are the median normalized concentrations; error bars denote the interquartile ranges. Open circles are the estimated concentration values using Equation (1) and the regression-estimated k -value of 0.0025.



The example shown in Figure 2 is based on all tape advances over the three-year period of April 2001 to March 2004 for BC data collected at East St. Louis with a 2-channel Aethalometer (880 and 370 nm for BC and UVC, respectively). As previously mentioned, the data corrections are not made using a single adjustment for all data but rather the analysis is performed on a user-specified centered moving window of width w tape advances. A regression to determine k is performed for each tape advance (except the first $w/2$ and last $w/2$ tape advances) using data within the centered window of w tape advances. For each regression, the data point with the maximum residual is removed and the regression is repeated. If the leave-one-out regression k -value differs from the initial k -value by more than 0.001, the data point with maximum residual is deemed to exert too much influence and the leave-one-out slope is used for the k -value. **Figure 3** shows the k -value time series for the East St. Louis BC data set after smoothing with a centered median smoother of window size $w=30$ tape advances. The k -values for the spin-up period (first $w/2$ tape advances) and spin-down period (last $w/2$ tape advances) are imputed using the first- and last-resmoothed k -values, respectively.

Figure 3. Aethalometer BC data adjustment parameter time series for April 2001 through March 2004 at East St. Louis. Adjustment parameters from the regressions of median normalized concentration on ATN with a window of 30 tape advances; the time series was smoothed using a centered median smoother with a window of 30 tape advances.



Given the smoothed time series of k -values, the raw data are adjusted using Equation (1) with the smoothed k -value for tape advance j used to adjust all data between tape advances j and $j+1$.

The overall process is repeated for each channel of data in addition to the 880 nm BC data (i.e., 370 nm UVC data for a 2-channel Aethalometer or the remaining 6 channels for a 7-channel Aethalometer). Up to three years of data can be processed in a single batch when the raw data is collected with a 5-minute time base.

Aethalometer Black Carbon Data Assembled as Part of this Project

Data were received from local, state, and regional agencies as well as the U.S. Environmental Protection Agency (EPA). Based on available site identifier codes associated with the delivered data, 146 sets of files were received; generally each set is for a single site, but there may be multiple sets of data for a given site if operations or instruments were changed. A summary of data is displayed in **Table 1**. In addition, we paired daily averaged processed data with data reported in the Air Quality System (AQS) provided by EPA. A summary of these data sets is provided in **Table 2**. A comparison of official, AQS data and the processed data for an example site is provided in a later section of this document.

Summary of Aethalometer Data Processing Steps and QC Criteria

The Aethalometer Data Masher program can process data in a single run that spans approximately one year for 1-minute data or three years for 5-minute data. In this work, raw Aethalometer data files were combined from subdirectories into one- or three-year data sets for efficient processing. Multiple subdirectories often contain partial data from the same day and have the same file name. These files were renamed to ensure no data were lost; the Data Masher automatically removes any duplicate data. Each data set was processed using the default Data Masher settings and a site name was added to each file.

The hourly Data Masher output was used for calculation of 24-hr and monthly averages in Microsoft Access. A 75% completeness criterion was required for all averages. 1- and 5-minute Data Masher output was not processed further. Hourly data were used to produce seasonal and diurnal box plots with the requirement that BC-corrected mass concentrations were greater than or equal to $-0.5 \mu\text{g}/\text{m}^3$. Hourly data were also used to produce monthly box plots with no data screening. These plots are provided electronically with this memo.

Example of Differences between Raw and Processed (Adjusted) Data

As an example, the relationship between raw and adjusted BC concentrations at the Harvard School of Public Health (HSPH) EPA PM-Center Countway site in Boston, Massachusetts, is shown in **Figures 4 and 5** for October 1999 through September 2009. The Aethalometer was a 1-channel unit with a BC maximum ATN setting of 75 used during this period. The average adjusted/raw ratio for this time period is 1.19. In winter the average ratio was 1.30; in summer the average ratio was 1.08. The time series reveals that even within a given season, this ratio can vary. In particular, September time periods show different patterns, likely due to differences in BC sources and/or meteorology.

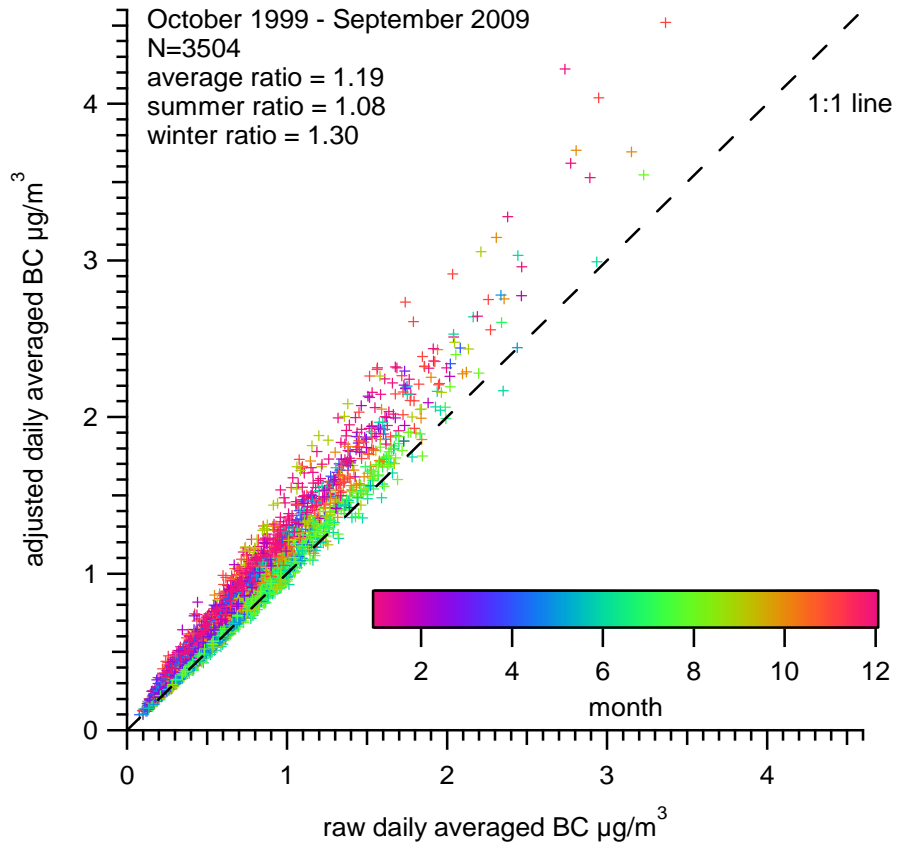


Figure 4. Daily averaged adjusted versus raw BC concentrations ($\mu\text{g}/\text{m}^3$) during October 1999 through September 2009 at the Harvard Boston site, colored by month. Maximum BC ATN = 75.

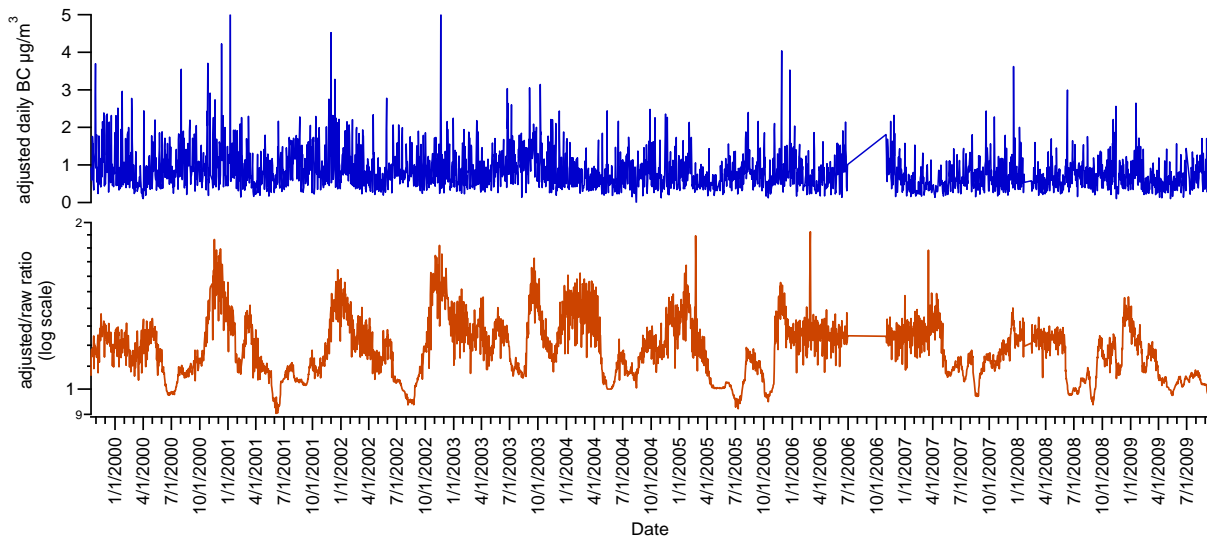


Figure 5. Time series of daily averaged BC concentrations $\mu\text{g}/\text{m}^3$ and daily averaged adjusted/raw ratio at the Harvard Boston site. Maximum BC ATN = 75.

Example of Differences between HSPH Official Data and Processed (Adjusted) Data

As an example, the processed data were compared to the official data as reported in the AQS by HSPH for the Harvard Boston PM-Center site in **Figures 6 through 8**. The HSPH official data set included HSPH validation of the raw data to void certain records but did not include adjustments for the mass loading effect. Raw data, as obtained from the instrument, are used to generate the adjustments; thus, it is necessary to screen the adjusted data to remove all records that were voided in the official data. The average adjusted/official ratio was 1.19 and the average difference between adjusted and official data was 124 ng/m^3 . The largest differences between adjusted and official data were seen in wintertime when there is less sulfate to suppress the mass loading effect.

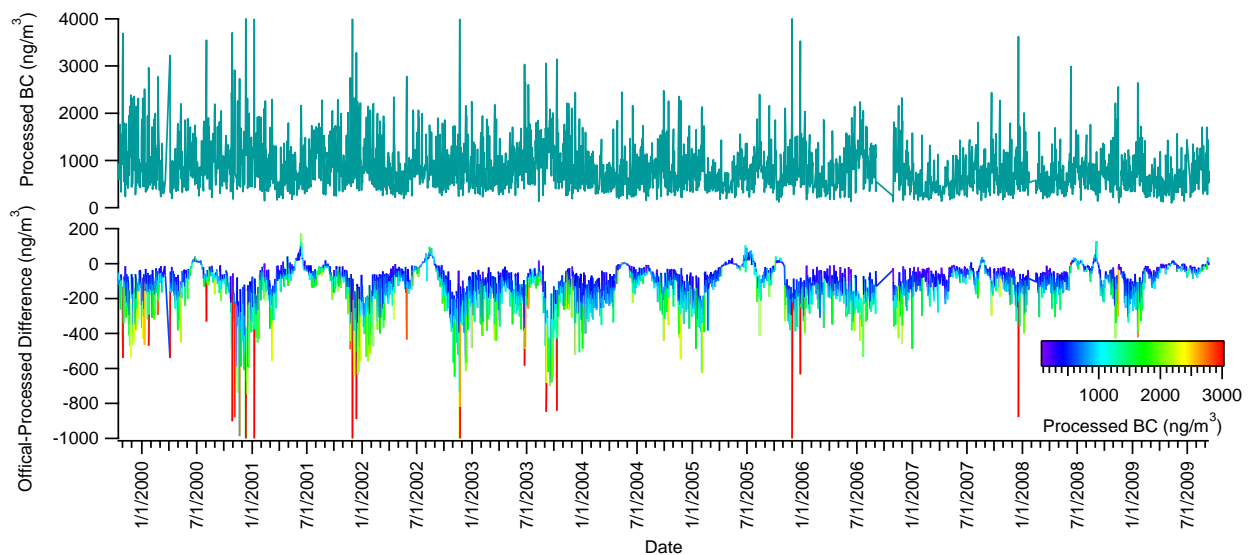


Figure 6. Time series of daily averaged processed (adjusted) BC concentrations and the difference in daily averaged processed BC with HSPH reported BC concentrations (ng/m^3) at the Harvard Boston site in October 1999 through September 2009.

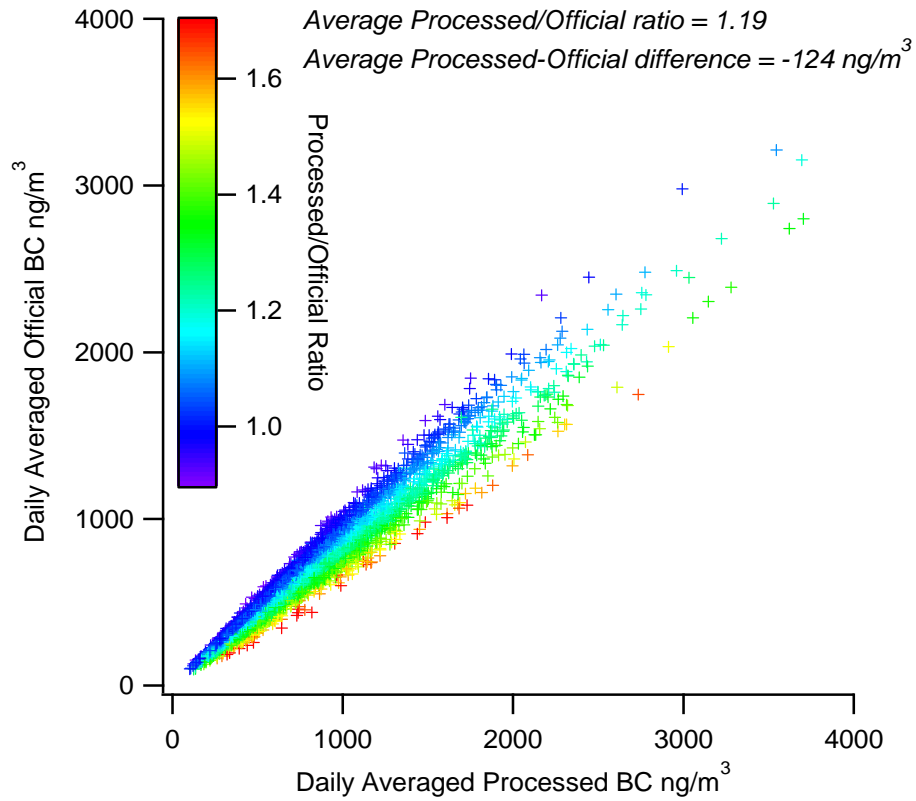


Figure 7. Comparison of daily averaged processed (adjusted) BC concentrations with daily averaged HSPH reported (“official”) BC concentrations (ng/m^3) at the Harvard Boston site from October 1999 through September 2009.

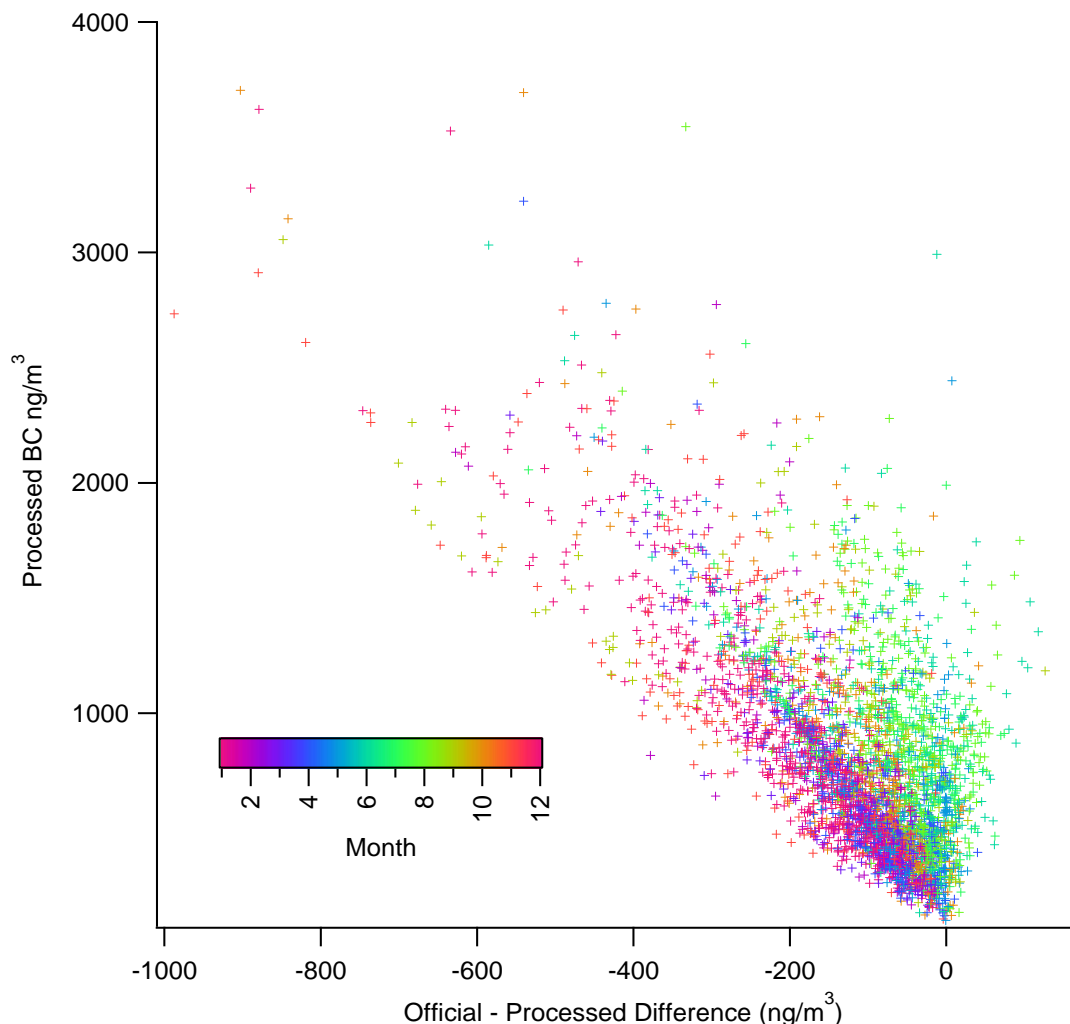


Figure 8. Comparison of daily averaged processed (adjusted) BC concentrations with the difference between daily averaged HSPH reported (“official”) BC concentrations and processed BC concentrations (ng/m^3) at the Harvard Boston site from October 1999 through September 2009.

Example of Black Carbon Climatology

Processed (adjusted) data from the HSPH EPA PM-Center Countway site in Boston, Massachusetts, are also used to demonstrate BC climatology. **Figure 9a** shows the day-of-week pattern for daily-average adjusted BC over the entire data set (October 1999 through September 2009). Box plots were constructed for the distributions of daily-average BC divided by the centered seven-day day average BC; this normalization stabilizes the BC concentration with respect to episodic behavior. BC concentrations are consistently high on weekdays, lower on Saturdays, and lowest on Sundays. **Figure 9b** shows the annual distributions of daily-average adjusted BC for 2000-2008. Box plots are omitted for 1999 and 2009 because data for these years were incomplete. While the annual trend is not strictly monotonic, BC concentrations have clearly decreased over the past decade, consistent with

Boston BC measurements made by the Massachusetts Department of Environmental Protection at two sites.

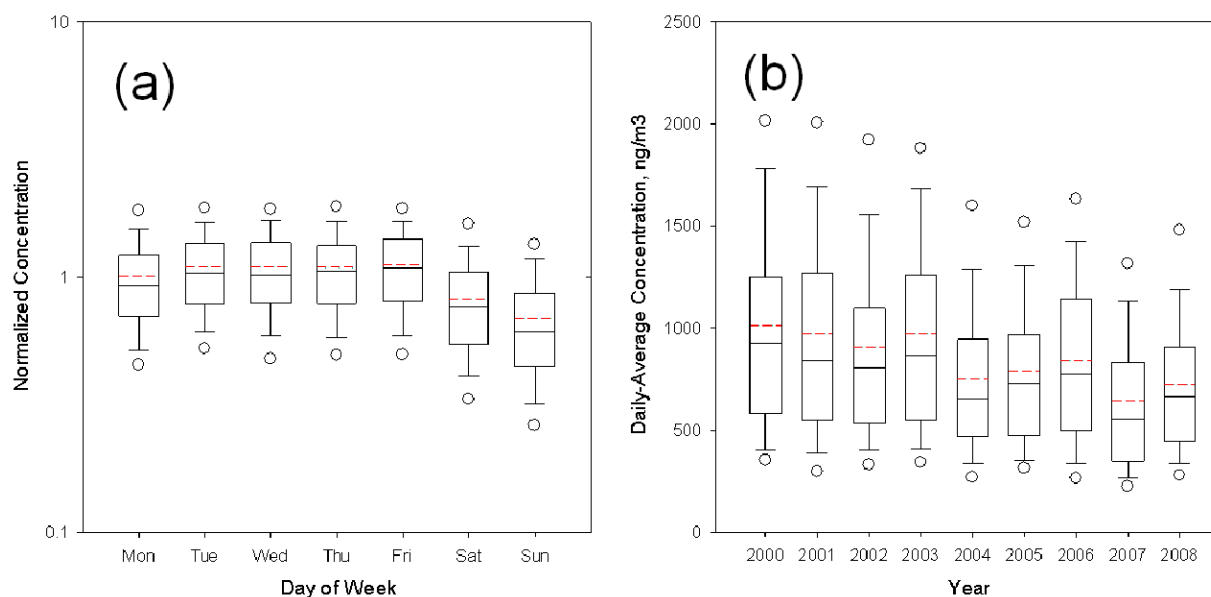


Figure 9. (a) Day-of-week patterns and (b) annual patterns for daily-average processed (adjusted) BC concentrations collected at the Harvard Boston site from October 1999 through September 2009. For (a), normalized concentration was calculated as the daily-average BC divided by the centered seven-day BC. Open circles are 5th and 95th percentiles, whiskers are 10th and 90th percentiles, box boundaries are 25th and 75th percentiles, the interior black line is the median and the interior, and the dashed red line is the arithmetic mean.

Figure 10 shows the long-term trend for seasonal BC concentration distributions. Seasonal median BC concentrations are typically highest in the summer (red boxes) and lowest in the spring (green boxes). Summer season and fall season BC was elevated in 2003 and 2006 compared to the other years. Winter exhibits the largest inter-annual variability with broad distributions during 2000-2002 and narrower distributions and lower median BC in the subsequent years.

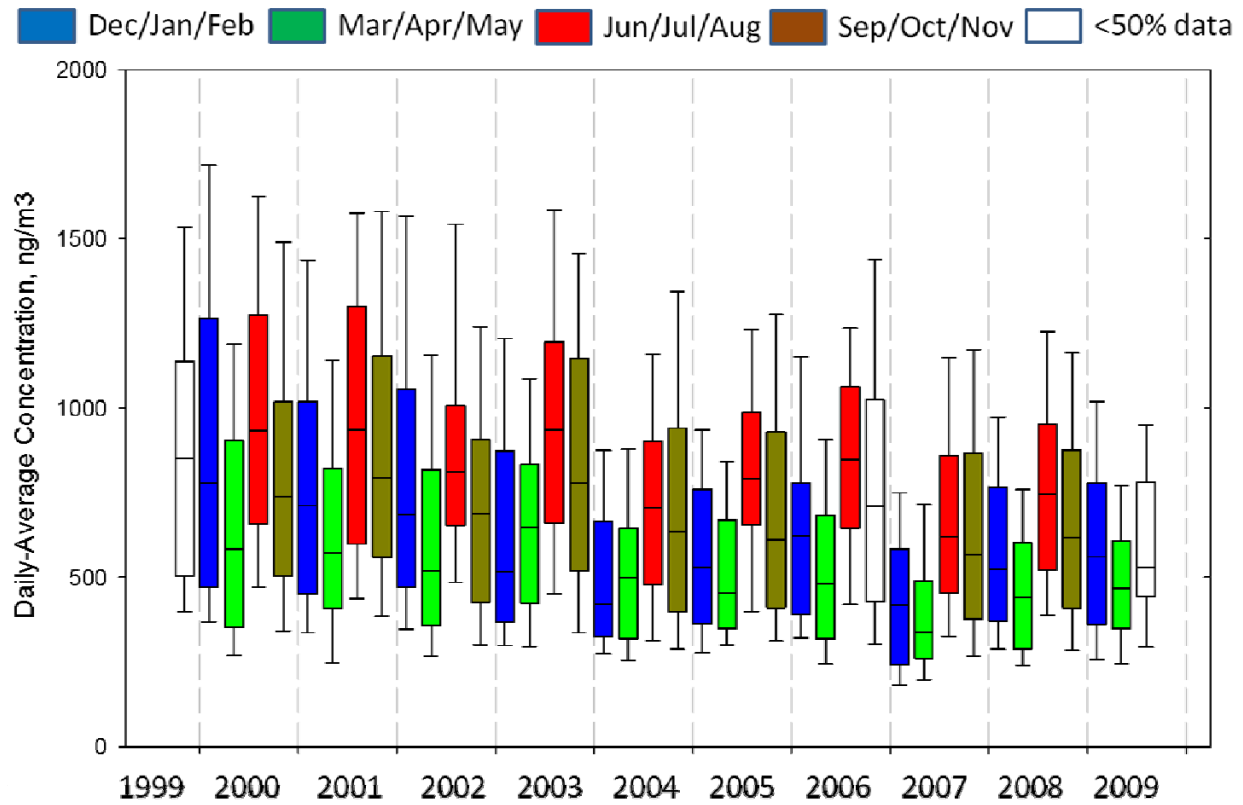


Figure 10. Seasonal distributions of adjusted BC concentration for data collected at the Harvard Boston site from October 1999 through September 2009. Whiskers are 10th and 90th percentiles, box boundaries are 25th and 75th percentiles, and the interior black line is the median.

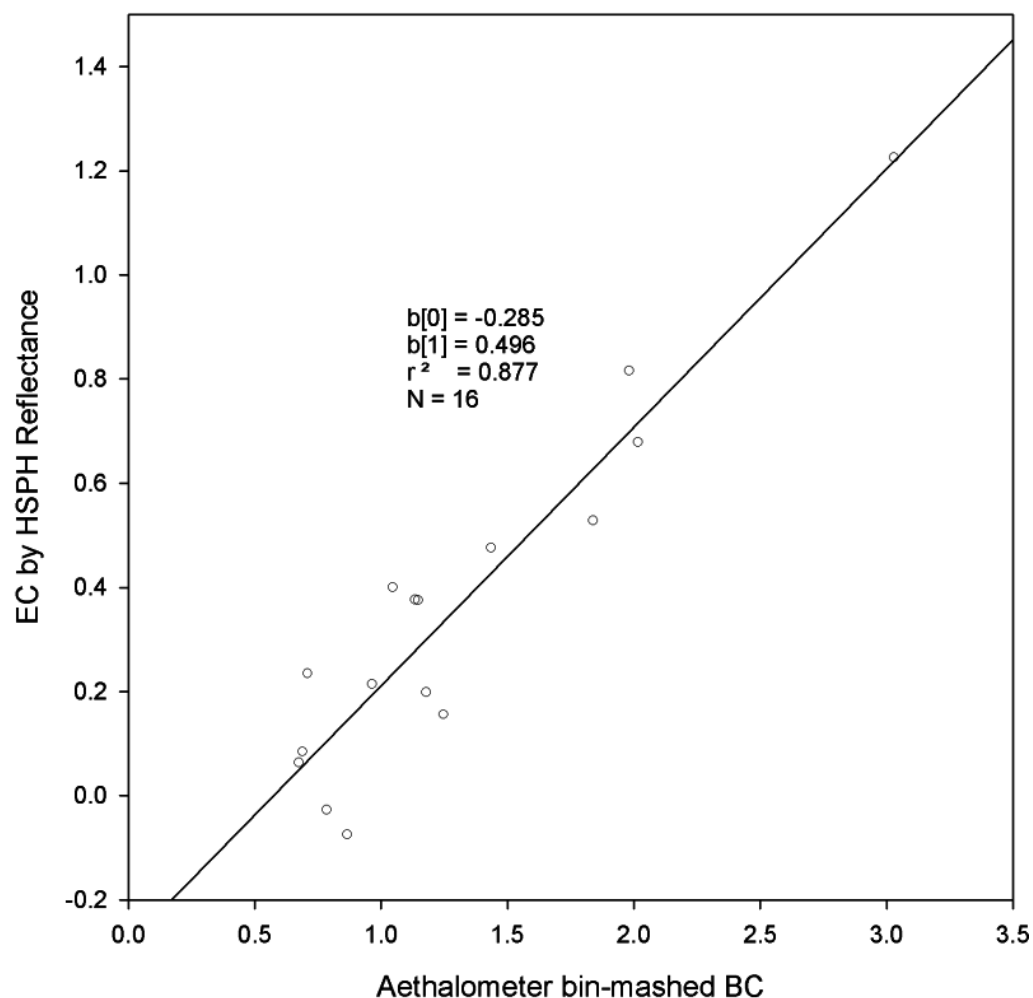
**Appendix E: Reflectance Analysis Method for
Estimating BC at the North End Site, February to
April 2003.**

Appendix E.

Reflectance analysis method for estimating BC at the N.End site for February-April 2003.

The North End site BC measurements started July 1, 2003. For trend purposes, 2003 was an important year. The Harvard School of Public Health (HSPH) has been determining BC from Teflon filters using a EEL Model M43D Smokestain Reflectometer, and analyzed all available Federal Reference Method (FRM) N.End filters for 2003 (there were no FRM filters available before 2003). Figure 1 shows the relationship between reflectance EC reported by HSPH and collocated Aethalometer 24-hour BC for sixteen days between August and November 2003.

Figure 1. Reflectometer EC vs. Aethalometer BC, August-November 2003



Twenty-one Teflon filters from the every 3rd-day FRM sampler were available for February through April 2003, and analyzed by HSPH. The Reflectometer EC data were converted into equivalent BC data using the regression shown here. The mean estimated BC for these data was 1.35 $\mu\text{g}/\text{m}^3$, compared to 1.32 $\mu\text{g}/\text{m}^3$ for the mean Aethalometer BC from July to December 2003.

**Appendix F: Turner (2011): Description of the
Binned Aethalometer Artifact Correction Method.**

Appendix F.

A Refined Methodology to Adjust Aethalometer Black Carbon Data for Measurement Artifacts

Extended Abstract 2011-A-676-AWMA

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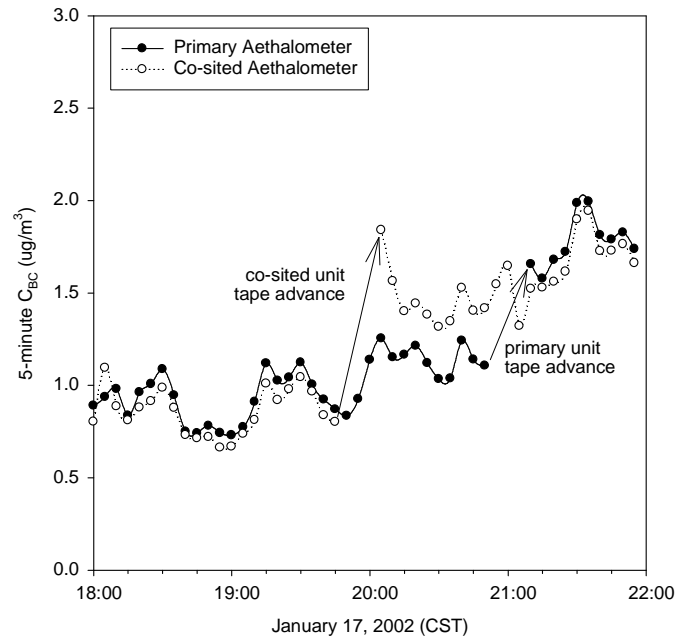
INTRODUCTION

Filter-based optical methods for estimating ambient particulate matter black carbon (BC) concentrations suffer from a mass loading effect whereby the instrument response decreases with increased BC loading. One such instrument, the AethalometerTM, continuously collects particles onto a filter and measures the wavelength-dependent transmission of light through the deposit. The mass loading effect is most-commonly observed as a step discontinuity in the reported concentration upon advancing the Aethalometer filter tape to deposit aerosol onto a clean filter spot instead of a particle-laden filter spot. **Figure 1** shows an example of this effect using collocated Aethalometers. Initially there is good agreement between the reported BC values but after the tape advances for the co-sited unit, it reports concentrations that are systemically higher than the primary unit. These higher values are actually closer to the true BC concentration with the data prior to the tape advance being biased low. Later in the time series the tape advances for the primary unit and subsequent data again exhibit good agreement. The mass loading effect is not governed solely by deposited absorbing (e.g. soot) aerosol. It is also influenced by the abundance of internally- or externally-mixed scattering aerosol (e.g. sulfate) in the ambient air that co-deposits with the absorbing black carbon particles.

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Extended Abstract #2011-A-676-AWMA. "A Refined Methodology to Adjust Aethalometer Black Carbon Data for Measurement Artifacts", Jay Turner, George Allen, Steven Brown, and Neil Frank. Proceedings of the 104th Annual Meeting of the Air & Waste Management Association, Orlando, FL, June 21-24, 2011.

Figure 1. Four hour time series of 5-minute black concentration carbon data for co-sited Aethalometers, East St. Louis. Each instrument exhibited one tape advance during this period. The co-sited unit data stream has been adjusted for instrument-to-instrument bias.



Several approaches have been proposed to adjust the data for these artifacts. For the Aethalometer, one approach uses the difference in reported concentration before and after a filter tape advance to estimate the artifact.¹ Another approach is to perform a regression of the reported concentration on the attenuation using all of the data over a specified time period.² Both of these approaches assume the true BC concentration is not changing over the time period of interest (across the tape advance for the first approach, for the entire time period used for the regression in the second approach). Thus, additional temporal aggregation of the data or smoothing of the artifact estimates is needed to damp the effect of this limiting assumption. The Aethalometer data post-processing program developed at Washington University that is publicly available to the air quality measurement community is being revised to include an algorithm for adjusting the raw data using both of these approaches. The gap-based algorithm was implemented for prior projects³ whereas the current work focuses on implementing a regression-based algorithm that uses all of the data. The latter algorithm is the focus of this extended abstract.

METHODS

The loading effect leads to the Aethalometer-reported concentration decreasing with increased attenuation (ATN, a measure of the light absorption by the deposited aerosol) even when the measured aerosol has a constant BC concentration. The goal of the

algorithm is to regress out this decreasing trend so that, on average, concentration is independent of ATN. The loading effect is assumed to follow the form:^{1,2}

$$BC_t = BC_r (1 + k \cdot ATN) \quad (\text{eqn 1})$$

where BC_t is the true concentration, BC_r is the Aethalometer-reported concentration, and k is an empirical parameter that, in this algorithm, is obtained from the regression of concentration on ATN.

The first step is to bin the raw concentration data (typically collected on a time base of 5-minutes but sometimes 1-minute) by attenuation. The algorithm allows the user to specify either a fixed bin width (e.g. 5 ATN units) or an approximate number of bins to be equally distributed over the attenuation range observed for each channel (Aethalometers can have 1-, 2- or 7-channels corresponding to the number of wavelengths for which absorption is measured). The default is a fixed bin width of 5 ATN units. Tape advances are followed by an instrument stabilization period with no data reported. The time duration of all data gaps are identified and the mode gap size (e.g. three consecutive five-minute missing records) is used to flag the tape advances in the time series. Data recorded between each tape advance are stratified into the ATN bins and the mean concentration is calculated for each bin. These concentration values are then normalized to the average concentration over all the bins. This step ensures that the data time series between each tape advance is given similar weight in the regression. Next, these normalized, binned concentrations are aggregated over a user-specified number of tape advances w (default $w = 30$) and the median value is calculated for each bin. Bin-specific median values are regressed on ATN to determine the empirical data adjustment parameter, k . **Figure 2** shows an example of the data used for the regression of normalized concentration on ATN (solid circles). Error bars denote the interquartile ranges about the median normalized concentrations. The open circles show the median normalized concentrations estimated from equation (1) using the fitted k -value. Ideally these values should be tightly clustered about the horizontal line at normalized concentration of unity (i.e. the ATN dependence of concentration has been removed) and in this example they are biased high by up to 2%.

The example shown in **Figure 2** is based on all tape advances over the three year period April 2001 to March 2004 for BC data collected at East St. Louis with a two-channel Aethalometer (880 and 370 nm for BC and UV-C, respectively). As previously mentioned, the analysis is actually performed using a user-specified centered window for the number of data traces between tape advances (w) used to calculate the median normalized concentrations. A regression to determine k is performed for each tape advance except the first $w/2$ and last $w/2$ records. These spin-up and spin-down periods are imputed with the first- and last-calculated k -values, respectively. **Figure 3** shows the k -value time series for the East St. Louis BC data set (black line). This time series is further smoothed using a centered median smoother of window size w (red line).

Figure 2. Aethalometer BC concentration for April 2001 – March 2004 at East St. Louis, binned by ATN and normalized within each tape advance. Solid circles are the median normalized concentrations; error bars denote the interquartile ranges. Open circles are the estimated concentration values using equation (1) and the regression-estimated k -value of 0.0025.

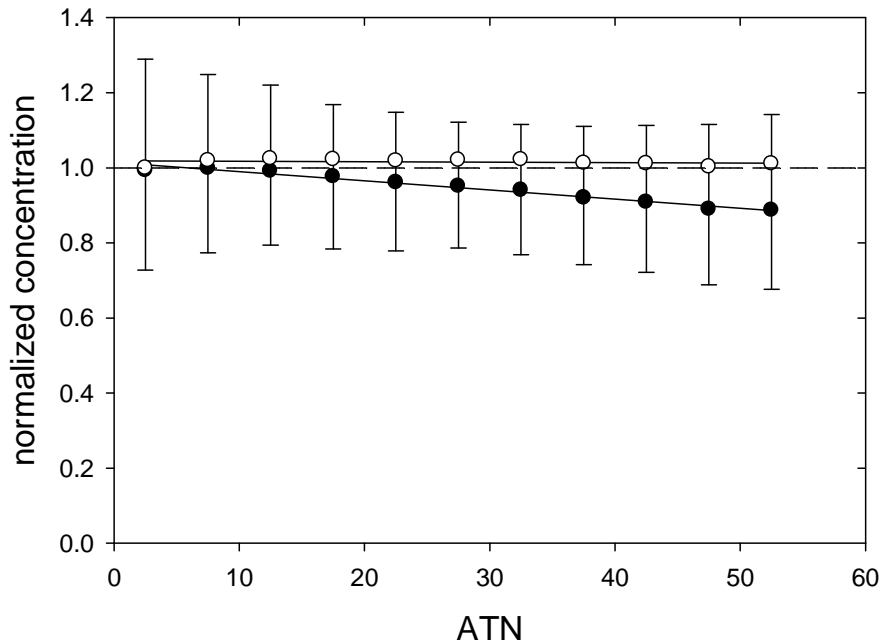
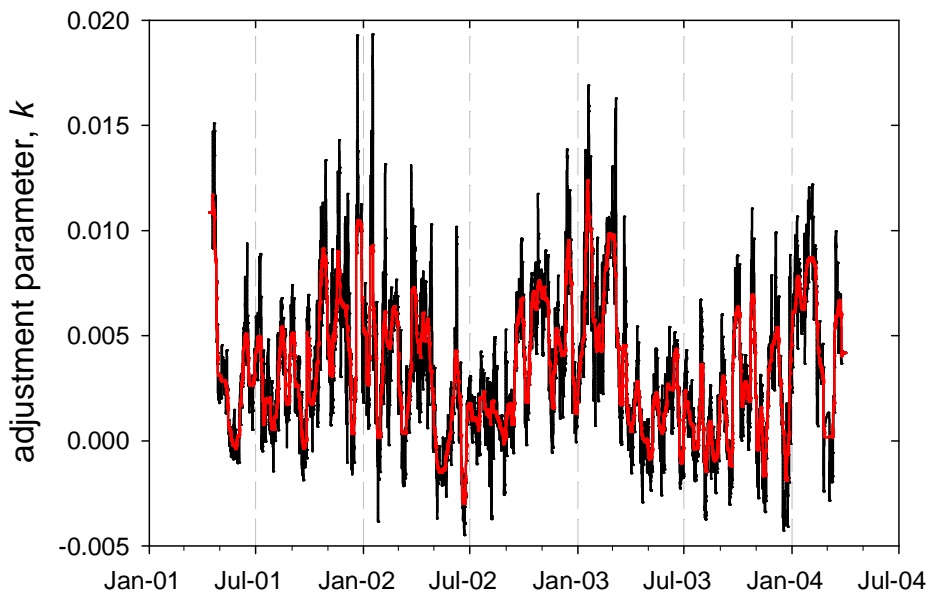


Figure 3. Aethalometer BC data adjustment parameter time series for April 2001 – Mach 2004 at East St. Louis: adjusted parameters from the regressions of median normalized concentration on ATN (black line); and smoothed parameters using a centered median smoother with a window of 30 tape advances (red line).



Given the smoothed time series of k -values, the raw data are adjusted using equation (1) with the smoothed k -value for tape advance j used to adjust all data between tape advances j and $j+1$.

The overall process is repeated for each channel of data in addition to the 880 nm BC data (i.e. 370 nm UV-C data for a two channel Aethalometer or the remaining six channels for a seven-channel Aethalometer). Up to about three years of data can be processed in a single batch if the raw data were collected at five-minute time base.

RESULTS

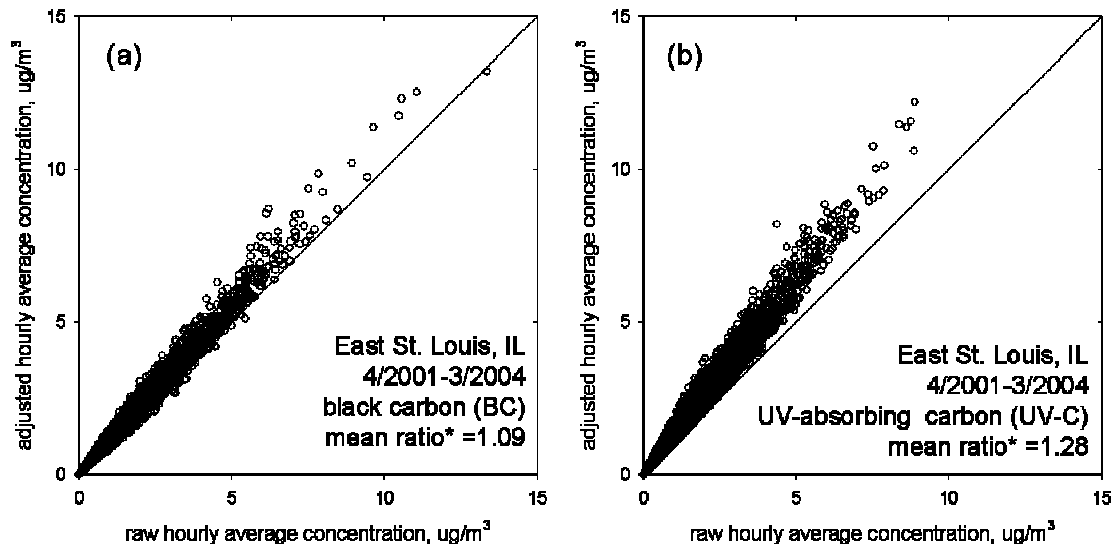
The adjustment parameter time series shown in Figure 3 is consistent with the pattern obtained using the tape advance gap method to estimate the parameter.¹ It exhibits a local maximum each winter and local minimum each summer because in St. Louis the concentration of scattering aerosol reaching the Aethalometer filter tape is higher in the summer than the winter and this aerosol partially offsets the BC mass loading effect.⁴

Figure 4 shows scatter plots of the adjusted hourly concentrations on the raw hourly concentrations for black carbon (BC, fig 4a) and UV-absorbing carbon (UV-C, fig 4b) using the April 2001 – March 2004 East St. Louis data set. The mean ratio of the adjusted-to-raw hourly concentrations is 1.09 for BC and 1.28 for UV-C. UV-C is measured at 370 nm and since absorption increases with decreasing wavelength the UV-C channel reaches the user-specified maximum attenuation (which triggers a tape advance) before the BC channel. For the two-channel Aethalometer deployed in East St. Louis the maximum attenuation was set to 125 which is reached for the UV-C channel when the BC channel attenuation is about 50. Thus, the UV-C data have a much larger artifact than BC data as demonstrated by the magnitude of the mean adjustment (9% and 28%, respectively). Note that a one-channel Aethalometer (BC only, 880 nm) with a maximum attenuation set to 125 would exhibit about the same percentage adjustment as the UV-C channel data in this example.

SUMMARY

The Aethalometer data post-processing software developed at Washington University has been revised to include a refined algorithm for adjusting the data for mass loading effects. For well-behaved data such as the example used in this extended abstract, the adjustments are generally consistent with an algorithm that derives adjustment parameters from the concentration change across each tape advance (not shown). The refined algorithm is expected to be superior for noisy data because it uses all the data and thus is more likely to damp the noise. While changes in the mass loading effect can occur on time scales shorter than the time over which this algorithm windows the data and smooths the adjustment parameter time series, the adjustments do capture changes in the mass loading effect that occur on longer times scales (weeks-to-months) and thus it is an improvement over using the raw data. For example, for the East St. Louis deployment there were on average three tape advances per day so the windowing and smoothing with $w = 30$

Figure 4. Raw and adjusted BC (a) and UV-C (b) hourly concentration values for the April 2001 – March 2004 data collected at East St. Louis. The mean ratio of the hourly adjusted-to-raw concentrations includes only those hours with adjusted concentrations greater than 1 $\mu\text{g}/\text{m}^3$.



corresponds to about ten days. The software is currently being beta tested and will be publicly available by mid-2011. It is currently being used to reprocess data collected from several sites across the United States towards developing a more robust description of aerosol black carbon climatology.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge David Shelow (USEPA), Natalie LaGuardia (STI) and Theresa O'Brien (STI) for their contributions to this project.

Although the research described in this presentation has been funded wholly or in part by EPA through a contract to the Sonoma Technology, Inc., it has not been subjected to the Agency's required peer and policy review and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

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KEYWORDS

Aethalometer, black carbon, optical saturation

**Appendix G: Turner (2008): Source Apportionment
for the North End Site.**

Appendix G.

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7 **TEMPORAL PATTERNS AND SOURCE APPORTIONMENT OF AMBIENT FINE**
8 **PARTICLE AETHOLOMETER BLACK CARBON IN BOSTON, MASSACHUSETTS**
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34 submitted August 1, 2008
35 for presentation at the *TRB 2009 Annual Meeting*
36 and publication in the *Transportation Research Record: Journal of the TRB*
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39 word count 7,263 (4,513 words and 11 figures @ 250 words per figure)
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47 **ABSTRACT**

48

49 Five years of ambient fine particle Aethalometer™ black carbon (BC) data from Boston,
50 Massachusetts, was analyzed for temporal patterns exerted on varying scales. The data were
51 collected by Massachusetts Department of Environmental Protection at the “North End” site
52 which is near the western terminus of the Sumner Tunnel. Diurnal profiles for hourly-average
53 BC on weekdays exhibited a strong mobile source signature with maximum concentration during
54 rush hour. In contrast, on weekends the morning rush hour BC enhancement was quite small and
55 maximum BC was observed in the early evenings. Diurnal profiles stratified by season revealed
56 a local maximum for the winter BC in the evenings for both weekdays and weekends. For nearly
57 three years of data collection at this site, the Aethalometer also collected data for UV-absorbing
58 carbon (UVC). UVC concentrations similar to BC are an indicator for fossil fuel combustion
59 while UVC enhancement above BC is an indicator for biomass combustion. An apportionment
60 of BC to traffic (more generally, fossil fuel) and wood smoke (more generally, biomass
61 combustion) sources yielded 15% contribution from wood smoke on an annual basis; wood
62 smoke was 5% of the summer BC and 28% of the winter BC, and accounted for more than 50%
63 of the BC on weekend nights in the winter. This analysis provides evidence that wintertime
64 biomass combustion, likely including both space heating and recreational fireplace use, is a
65 significant contributor to Aethalometer black carbon concentrations in Boston and it would be
66 erroneous to attribute all of the BC to mobile sources.

67 **INTRODUCTION**

68

69 This is substantial interest in mobile source contributions to ambient fine particle burdens and
 70 their associated adverse health effects. Elemental carbon (EC) has been used in some studies as
 71 a surrogate for estimating diesel particulate matter concentrations and exposures, and the
 72 potential flaws in this approach have been summarized by Schauer (1). For example, fine
 73 particle EC in urban environments can originate from a variety of emission sources including,
 74 but not limited to, mobile sources and biomass combustion. EC is conventionally measured by
 75 thermal-optical methods with the term EC suggesting carbon that is refractory in nature.

76 Another commonly-measured parameter is black carbon (BC) which is based, as its name
 77 suggests, on a measurement of light absorption. While BC and EC are inherently different
 78 measurements and both are operationally defined, these parameters are often highly correlated
 79 and BC is often used synonymously with EC in an urban air quality and health effects context.
 80 One method for measuring BC is the Aethalometer™ (Magee Scientific, Berkeley, CA) which
 81 quantifies the absorption of light by aerosol particles continuously deposited onto a quartz fiber
 82 filter tape. Certain versions of this instrument measure light absorption at two-or-more
 83 wavelengths and it has been shown that the wavelength dependence of absorption is quite
 84 different for emissions from fossil fuel combustion and biomass combustion (2). Allen *et al.* (3)
 85 and Sandradewi *et al.* (4-6) presented and applied methodologies which use multi-wavelength
 86 Aethalometer data and additional fine particle measurements to quantitatively apportion ambient
 87 fine particulate matter burdens to wood smoke and traffic emission sources. In this study, we
 88 examine a five year time series of ambient fine particle Aethalometer data for evidence of
 89 different source types contributing to the BC burden at a site in Boston, Massachusetts.

90

91 As previously stated, the Aethalometer measures the absorption of light by aerosol particles
 92 continuously deposited onto a quartz fiber filter tape. Wavelength-specific light transmission
 93 through the particle-laden filter, T , is read at a user defined time interval (typically 5-minutes).
 94 The attenuation, ATN, is –

$$95 \quad ATN = -100 \times \ln(T) = -100 \times \ln\left(\frac{S_B - S_Z}{R_B - R_Z}\right)$$

96 where S_B and S_Z are the light intensities measured downstream of the particle-laden deposit with
 97 the upstream lamp on and off, respectively; and R_B and R_Z are the light intensities measured
 98 downstream of the clean section of the filter tape with the upstream lamp on and off, respectively.
 99 The effective absorption coefficient, b_{ATN} [L^{-1}], which corresponds to absorption by particles
 100 deposited in the quartz fiber filter tape is –

$$101 \quad b_{ATN} = \frac{A \cdot \Delta ATN}{Q \cdot \Delta t}$$

102 where A is the particle deposit area [L^2], Q the air sample flow rate [$L^3 t^{-1}$], Δt the time interval
 103 between measurements [t], and ΔATN the change in attenuation between measurements. The
 104 aerosol absorption coefficient, b_{abs} [L^{-1}] is then given by -

$$105 \quad b_{abs} = \frac{b_{ATN}}{H(\lambda) \cdot R(\lambda, ATN)}$$

106 where $H(\lambda)$ is the absorption enhancement in the massively scattering environment of the quartz
 107 fiber filter and $R(\lambda, ATN)$ accounts for the decrease in the absorption enhancement with particle
 108 loading onto the filter. Finally, the ambient concentration of absorbing material, $C(\lambda)$ [$M L^{-3}$], is
 109 given by –

$$110 \quad C(\lambda) = \frac{b_{abs}}{E_{abs}} = \frac{b_{ATN}}{E_{ATN}}$$

111 where E_{abs} [$L^2 M^{-1}$] is the aerosol mass absorption efficiency and E_{abs} [$L^2 M^{-1}$] is the effective
 112 particle mass absorption efficiency for particles deposited in the quartz fiber filter. Black carbon
 113 (BC) is the term used for $C(880 \text{ nm})$ while ultraviolet absorbing carbon (UVC or UVPM) is the
 114 term used for $C(370 \text{ nm})$. The user programs into the Aethalometer for each wavelength an
 115 “absorption cross-section”, $\sigma(\lambda)$ [$L^2 M^{-1}$], which has been determined from laboratory
 116 experiments and corresponds to –

$$117 \quad \sigma(\lambda) = [E_{abs} H](\lambda) = E_{ATN}(\lambda) \quad \text{with} \quad R = 1$$

118 The Aethalometer does not account for optical saturation (i.e. $R=1$ is assumed) and
 119 compensation for this phenomenon must be handled by the user during data post-processing.
 120 When the light transmission drops below a user-defined threshold (that is, when ATN exceeds a
 121 threshold value) the filter tape is advanced and particles are collected on a clean section of the
 122 filter tape.

123
 124 While it has long been known that the absorption per unit mass of deposited absorbing aerosol
 125 decreases with increased loading (i.e. $R \neq 1$), only recently have there been extensive efforts to
 126 compensate the data for such effects. Arnott *et al.* (7) developed a first-principles model for light
 127 attenuation in the Aethalometer which showed that the loading-dependent negative bias for the
 128 Aethalometer-reported concentration is worst for an aerosol with a low single scattering albedo
 129 which is the ratio of the light removed by scattering to the total light extinction. For an aerosol
 130 with a high single scattering albedo, the optical saturation phenomenon is further complicated by
 131 a matrix effect from co-deposited scattering aerosol which can partially or even wholly offset the
 132 optical saturation effect. Turner *et al.* (8) have reviewed various equations proposed to
 133 compensate Aethalometer data for these effects, including comparisons to the model of Arnott *et*
 134 *al.* (7). Some equations perform better at low single scattering albedo (9-11) with others perform
 135 better at high single scattering albedo (12). For sites such as Boston with significant seasonal
 136 variations in the single scattering albedo, driven to a large extent by seasonal variations in the
 137 aerosol sulfate concentration, no single equation is preferred for all conditions and the equation
 138 of Virkkula *et al.* (11) has been implemented as described in the Methods section of this paper.

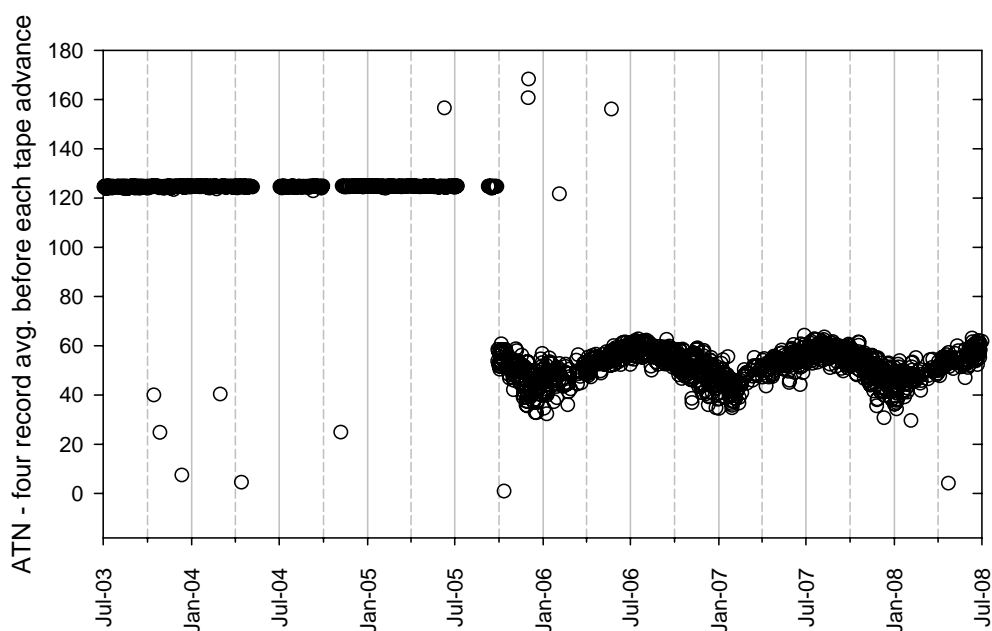
139
 140 After compensating the data for optical saturation effects, the conditioned data set was mined for
 141 insights into the emission sources contributing to BC at the North End site by examining BC
 142 diurnal profiles and applying the model of Sandradewi *et al.* (4-6) to apportion BC to traffic and
 143 wood smoke sources. This approach assumes that traffic (or more broadly, mobile sources) is
 144 the only significant contributor to BC from fossil fuel combustion and wood smoke is the only
 145 significant contributor to biomass combustion. No effort is made to determine the total
 146 particulate matter burdens from these emission sources since this would require additional air
 147 quality parameters that were not measured.

148

149 **METHODS**

150
151 An AE-21 Aethalometer (SN #413) has been operated since July 2003 at the Massachusetts
152 Department of Environmental Protection (MADEP) "North End" monitoring site in Boston
153 (174 North Street, AIRS Site ID 25-025-0043), 42.363N, -71.054E. The site is located on the
154 roof of a four-story building with the inlet approximately 20 meters above ground and 25 meters
155 above sea level. The immediate area is a mix of commercial and residential use with substantial
156 traffic activity. The Sumner Tunnel exit (Rt. 1A south) is at the opposite edge of the building
157 from the monitor. The inner harbor of Boston is ~ 500 meters east of the site.
158

159 Five years of raw Aethalometer data, from July 2003 through June 2008, were obtained from
160 MADEP for this analysis. While validated hourly-average BC concentration data is uploaded by
161 MADEP to the USEPA Air Quality System (AQS) and is publicly available, this analysis
162 required not only the BC concentration data but also the attenuation data (ATN) to compensate
163 for optical saturation effects; thus, the raw data files were obtained from MADEP and were not
164 subjected to MADEP's validation protocols. Data were collected at five minute time resolution
165 and reported at standard conditions of 1013 mbar and 25°C. A cyclone was placed on the
166 Aethalometer inlet to achieve a particle size cutpoint of 2.5 μm aerodynamic diameter ($\text{PM}_{2.5}$).
167 Initially the Aethalometer was programmed to collected data at 880 nm (BC) only; starting in
168 September 2005 the instrument was reprogrammed to also collected data at 370 nm (UVC). The
169 maximum attenuation was initially programmed to 125; thus, tape advances were triggered when
170 the BC channel attenuation exceeded 125. Starting in September 2005 tape advances were
171 triggered when the UVC channel attenuation exceeded 125. The later case typically corresponds
172 to BC attenuation in the range from 30 to 60 (**Figure 1**).
173
174



175 1.
176 **FIGURE 1** Time series for the BC channel (880 nm) attenuation just prior to each tape
177 advance.

178 The Aethalometer data were compensated for optical saturation using the equation of Virkkula
 179 *et al.* (11) as implemented by Turner *et al.* (8). The governing equation is –

$$180 \quad R(\lambda) = (1 + k(\lambda) \cdot ATN)^{-1}$$

181 or simply -

$$182 \quad C(\lambda) = C_{AETH}(\lambda) \times (1 + k(\lambda) \cdot ATN)$$

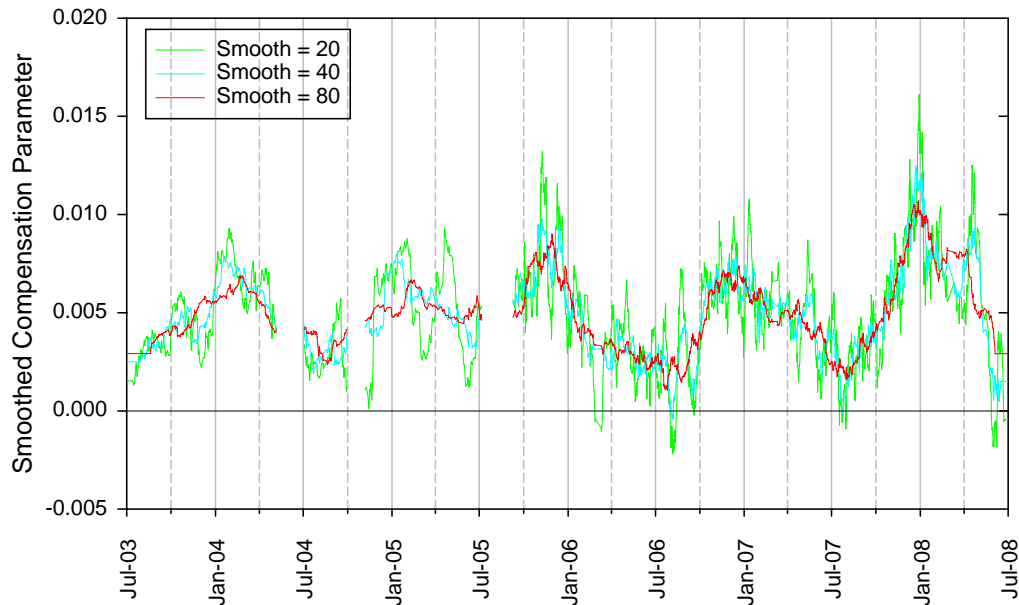
183 where $C_{AETH}(\lambda)$ is the concentration reported by the Aethalometer with $\sigma(370 \text{ nm})$ and
 184 $\sigma(880 \text{ nm})$ set to 39.5 and 16.6 m^2/g , respectively, and k is the compensation parameter. The
 185 negative bias from optical saturation causes a step increase in the reported concentration across a
 186 tape advance if the aerosol being sampled has a constant composition and concentration. The
 187 compensation parameter k is calculated for each tape advance assuming the ambient
 188 concentration of absorbing material is constant for the 15-minute tape advance-induced
 189 instrument stabilization period as well as the averaging times used to estimate the concentrations
 190 before and after the tape advance. In reality, the ambient concentration is often changing over
 191 this period and thus the individual estimates of k are smoothed over numerous tape advances to
 192 stabilize the estimates. In this study, a centered rolling median of the tape advance-specific k
 193 values was used to generate a time series of smoothed k which was applied to the raw (C_{AETH})
 194 data.

195 RESULTS

196 Aethalometer Data Conditioning

197
 198 Aethalometer data was compensated for optical saturation effects by smoothing over 20, 40, and
 199 80 tape advances. **Figure 2** shows the smoothed compensation parameter, k , for the entire time
 200 series. The compensation parameter exhibits a seasonal pattern with maximum values in the
 201 winter (relatively large negative bias in the reported concentration values) and minimum values
 202 in the summer (relatively small negative bias in the reported concentration values). This pattern
 203 is consistent with the role of scattering aerosol on partially offsetting the optical saturation effect.
 204 In Boston, ammonium sulfate is the largest contributor to aerosol scattering and exhibits a
 205 seasonal pattern with highest concentrations in the summer and lowest concentrations in the
 206 winter.
 207
 208

209
 210 The September 2005 change in the wavelengths measured (from BC only to BC plus UVC)
 211 affected the characteristic BC channel attenuation when a tape advanced was triggered (Figure 1).
 212 The average time between tape advances was 40- and 20-hours before and after this change,
 213 respectively. The tape advance frequency affects the time period corresponding to a given
 214 smoothing parameter. Figure 2 shows that smoothing over 80 tape advances yields a similar
 215 seasonal pattern for each year of the five year measurement period. It is recognized that this
 216 degree of smoothing can only account for seasonal variations in the optical saturation effect and
 217 it will not account for changes occurring on finer time scales (e.g. as synoptic weather patterns
 218 move through the area on three-to-five day time scales, often altering the aerosol composition).
 219 The analyses and interpretation of data presented in this paper are consistent with this limitation.
 220 While conditioned data was obtained using a smoothing parameter of 80, the sensitivity of
 221 certain results to the smoothing parameter was investigated to ensure the robustness of the results.
 222

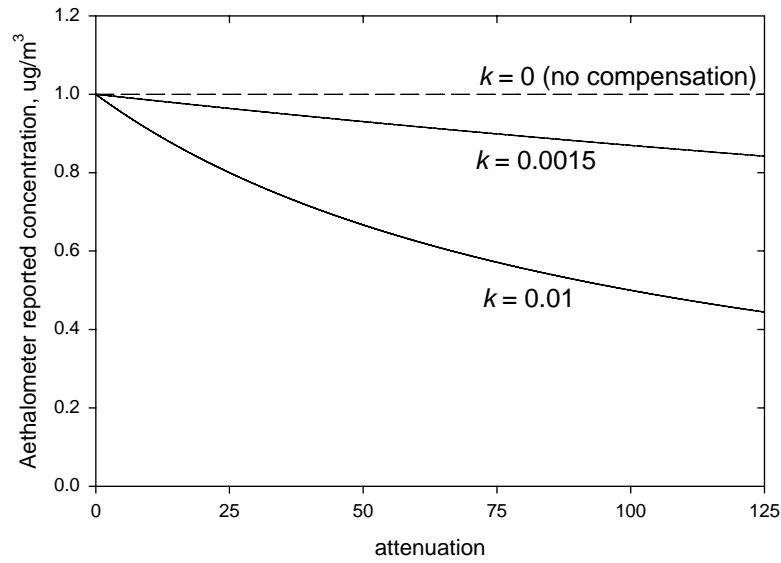


223
224 **FIGURE 2** Time series for the BC optical saturation compensation parameter, k , for
225 various smoothing parameters.
226

227
228 Record-specific adjustments to the data are affected by the compensation parameter and the
229 record-specific attenuation. **Figure 3** shows modeled traces for the Aethalometer-reported
230 concentration as a function of attenuation for a constant $1 \mu\text{g}/\text{m}^3$ BC aerosol at the 1st and 99th
231 percentiles of the smoothed compensation parameter ($k = 0.0015$ and 0.010 , respectively).
232 Virtually all of the ambient data falls between the two solid curves, corresponding to record-
233 specific concentration adjustments of up to $\sim 50\%$. Compensated 5-minute data were rolled up to
234 hourly averages. **Figure 4** shows scattergrams for the raw and compensated hourly data for the
235 time periods before and after the change in instrument configuration. The largest adjustments
236 were in the winter prior to the change in the instrument configuration.
237

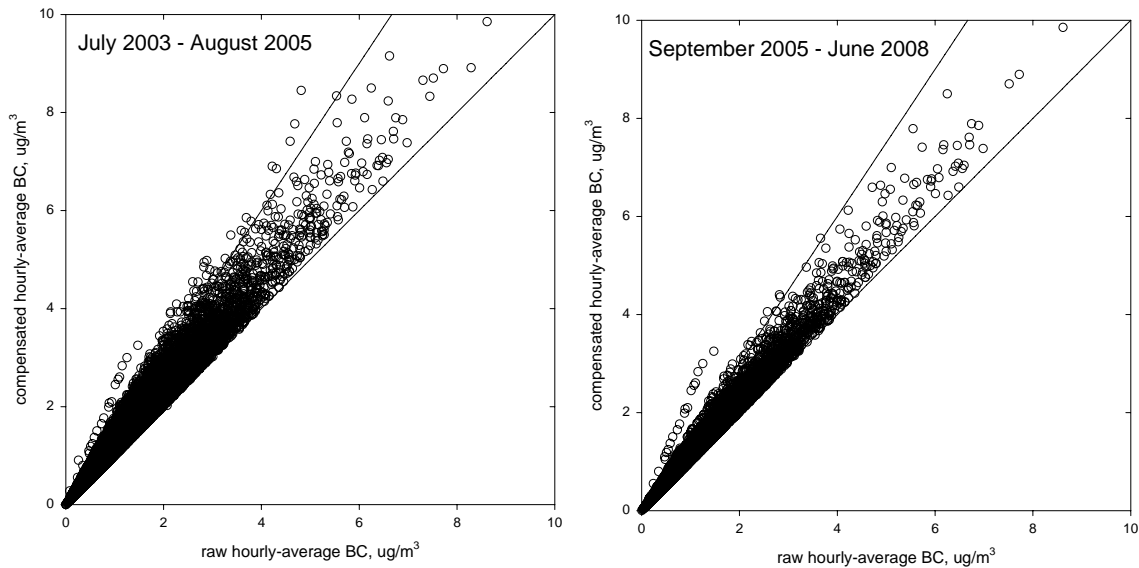
238 Year-to-Year Trends

239
240 **Figure 5** shows box plots for the hourly BC concentration distributions by year (defined as the
241 period from July through the following June) for the five year data set. The first year (July 2003
242 through June 2004) exhibits higher concentrations than the subsequent years. This difference is
243 more dramatic for the compensated data compared to the raw data because the higher attenuation
244 values of 125 reached during the first two years corresponds to larger adjustments for optical
245 saturation effects. Compensation is most significant for the winter periods and thus the right-
246 hand plot shows the distributions by year for the period from October through March only. The
247 raw data suggests that the first year was no different from more-recent years and the median
248 concentration was lowest in the second year. In contrast, the compensated data shows the
249 highest median was observed in the first year and the second year no longer exhibits the lowest
250 median. This analysis demonstrates the importance of compensating the data for seasonal trends
251 in the optical saturation artifact, as it leads to different interpretation of year-to-year patterns in



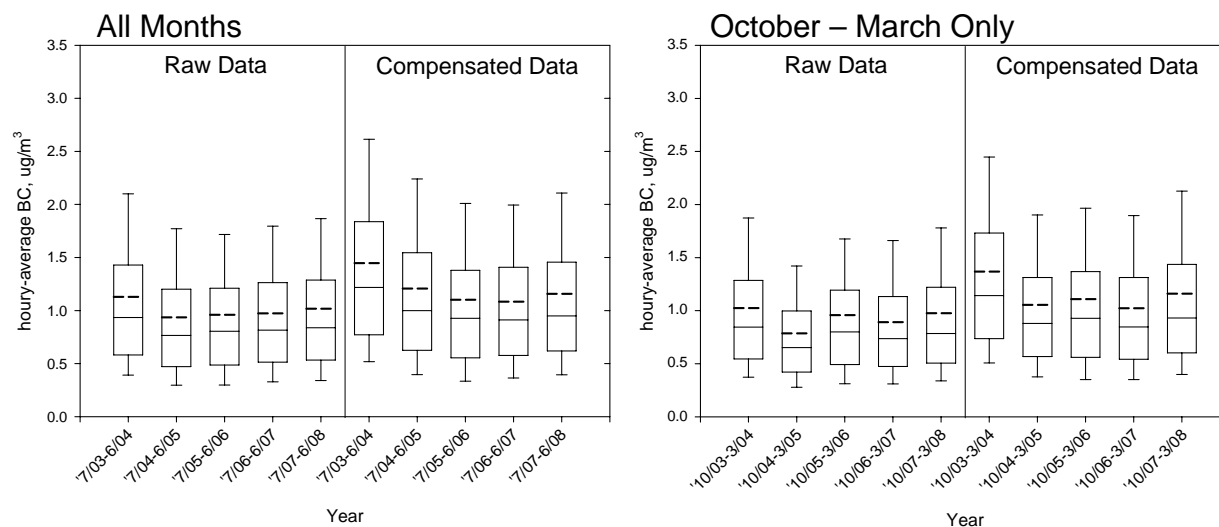
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FIGURE 3 Negative bias in the Aethalometer reported concentration as a function of attenuation for the 1st and 99th percentile values of the smoothed BC compensation parameter, k .



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264

FIGURE 4 Adjustments to the hourly-average BC concentration data for the time periods before (left) and after (right) the change in the instrument configuration from monitoring BC only to monitoring BC and UVC.

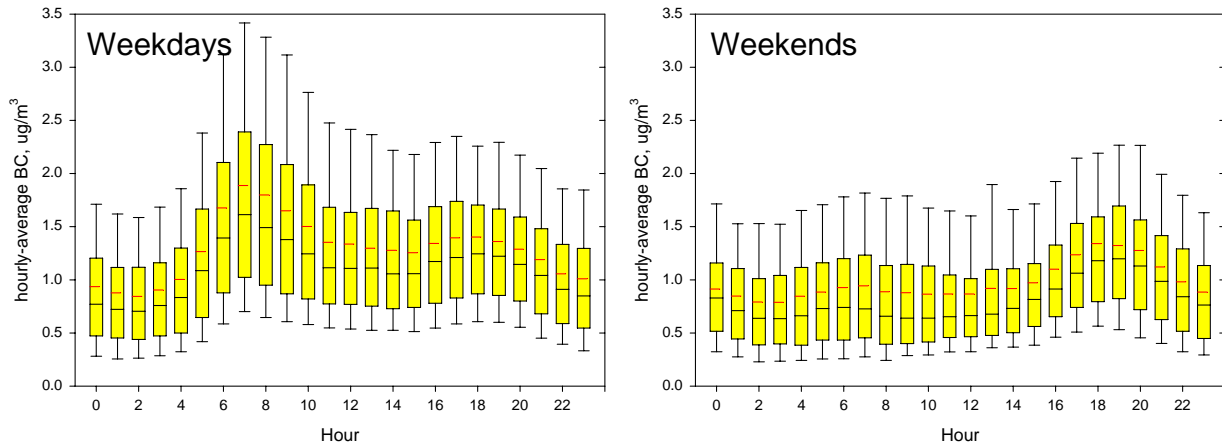


265
 266 **FIGURE 5** Yearly distributions (from July through June) of raw and compensated hourly
 267 **BC** data for all months (left) and from October through March (right). Whiskers are 10th
 268 **and 90th** percentiles.
 269

270
 271 the BC concentration. The decrease in BC concentrations over the 2003-2004 time period was
 272 also observed at two sites in Boston with Aethalometers operating since at least 2000 (G. Allen,
 273 unpublished).
 274

275 Diurnal Trends

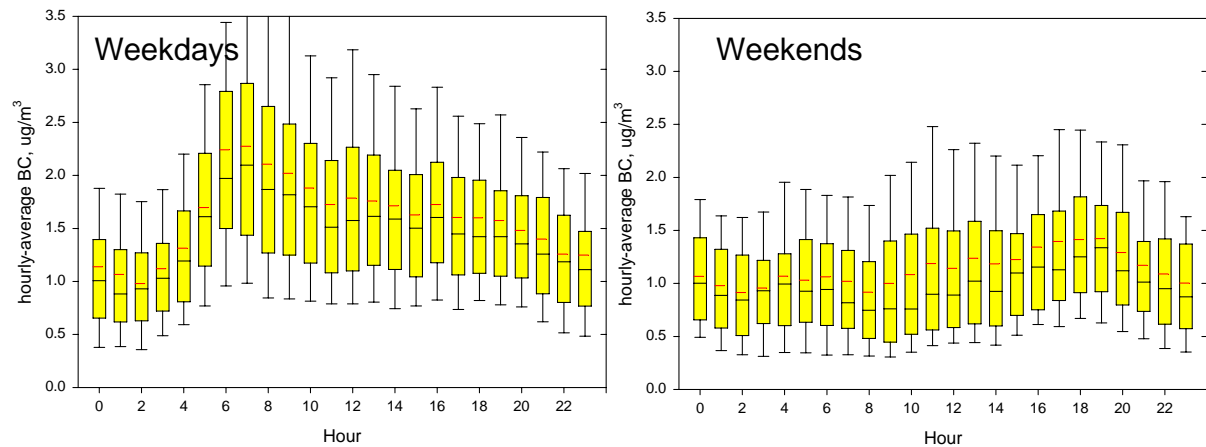
276
 277 **Figure 6** shows BC diurnal profiles for the entire five year data set stratified by weekdays and
 278 weekends. For weekdays, maximum BC concentrations are observed from 0600 to 0900 EST,
 279 consistent with the morning rush hour, followed by a local minimum at midday from 1100 to
 280 1500 EST. A modest secondary maximum occurs from 1600 to 2000 EST, consistent with the
 281 evening rush hour. Lowest BC concentrations are observed at night from 2200 to 0400 EST.
 282 The midday minimum could arise from reduced emissions during off-peak commute hours
 283 coupled with a midday maximum in the mixing layer depth which dilutes the ground-level
 284 emissions. The nighttime minimum occurs despite a shallow mixing layer depth which
 285 suppresses vertical dilution of any ground-level emissions. The weekend pattern is markedly
 286 different with the absence of a BC maximum during morning rush hour and instead a BC
 287 maximum in the evening from 1700 to 2100 EST. **Figure 7** shows diurnal profiles for both the
 288 summer (June – August) and winter (December – February) periods, again stratified by
 289 weekdays and weekends. BC concentrations are higher in the summer compared to the winter,
 290 and also exhibit a wider range of concentrations in the summer. Summertime weekdays do not
 291 exhibit the evening local maximum in BC concentration that is observed for summer weekends,
 292 winter weekdays, and winter weekends. Winter weekends exhibit a modest local maximum in
 293 the BC concentration during morning rush hour period that is not observed for summer weekends.
 294
 295



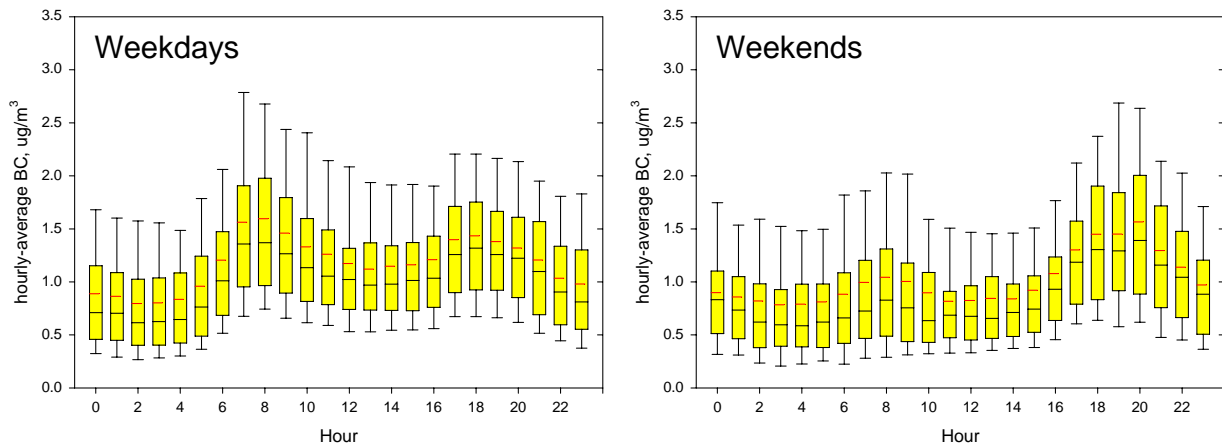
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299
300

FIGURE 6 Diurnal profiles of hourly-average compensated BC for weekdays (left) and weekends (right). Whiskers are 10th and 90th percentiles. The interior black line is the median and the red line is the arithmetic mean.

SUMMER



WINTER



301
302
303
304

FIGURE 7 Summer (top) and winter (bottom) diurnal profiles of hourly-average compensated BC for weekdays (left) and weekends (right). Whiskers are 10th and 90th percentiles. The interior black line is the median and the red line is the arithmetic mean.

305 Median diurnal profiles are shown in **Figure 8** for the summer and winter periods stratified by
 306 weekdays and weekends, as well the difference between weekday and weekend median diurnal
 307 profiles. The enhancement in the weekday BC compared to the weekends exhibits similar
 308 diurnal profiles for both seasons with a maximum at 0700-0800 EST followed by a nearly
 309 monotonic decrease throughout the day. Within each season, the median nighttime
 310 concentrations for weekdays and weekends are nearly identical.

311
 312 In aggregate, the diurnal profiles of Figures 7 and 8 support mobile sources being the dominant
 313 contributor to at least the within-season diurnal variability in the BC concentration. However,
 314 this does not mean that mobile sources are the only contributors to BC concentrations. The
 315 wavelength dependence of absorption is commonly expressed as a power law relationship –

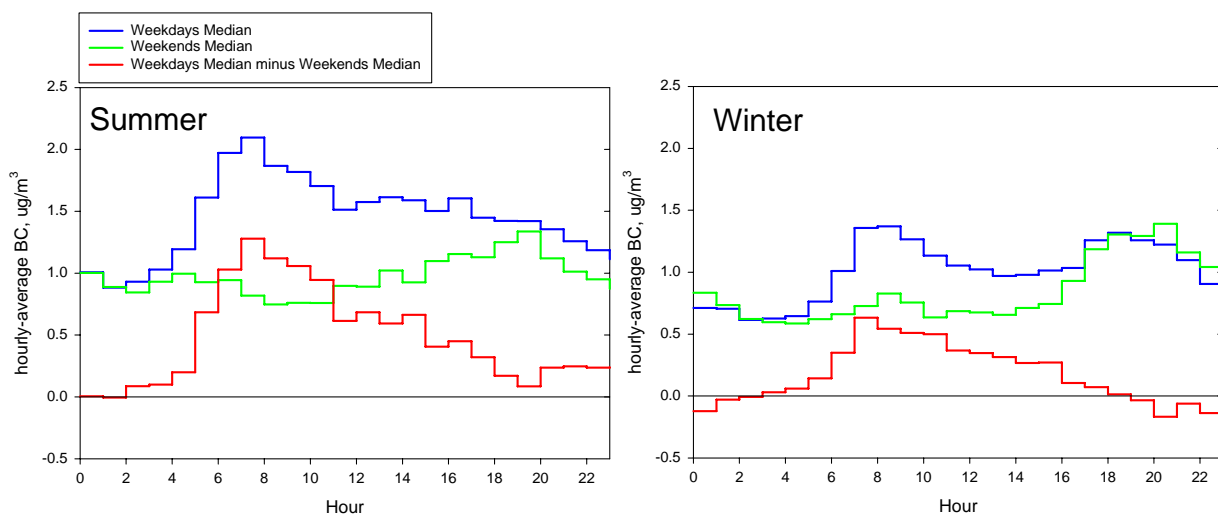
$$316 \quad b_{abs}(\lambda) = K\lambda^{-\alpha}$$

317 where K and α are absorption Angstrom coefficients and α is called the Angstrom exponent.
 318 Kirchstetter *et al.* (2) have shown that $\alpha \sim 1$ for aerosols from fossil fuel combustion and $\alpha \sim 2$
 319 for aerosols from biomass/biofuel burning and for mineral dust. Given the PM_{2.5} cutpoint will
 320 eliminate most of the airborne mineral dust and given the relatively low mass absorption
 321 efficiency for mineral dust, high Angstrom exponents in this physical setting are assumed to
 322 reflect biomass/biofuel combustion. **Figure 9** shows diurnal profiles for the UVC/BC ratio for
 323 the summer and winter periods stratified by weekdays and weekends. This ratio is related to the
 324 Angstrom exponent by –

$$325 \quad \alpha = 1 + 1.15 \times \ln(UVC / BC)$$

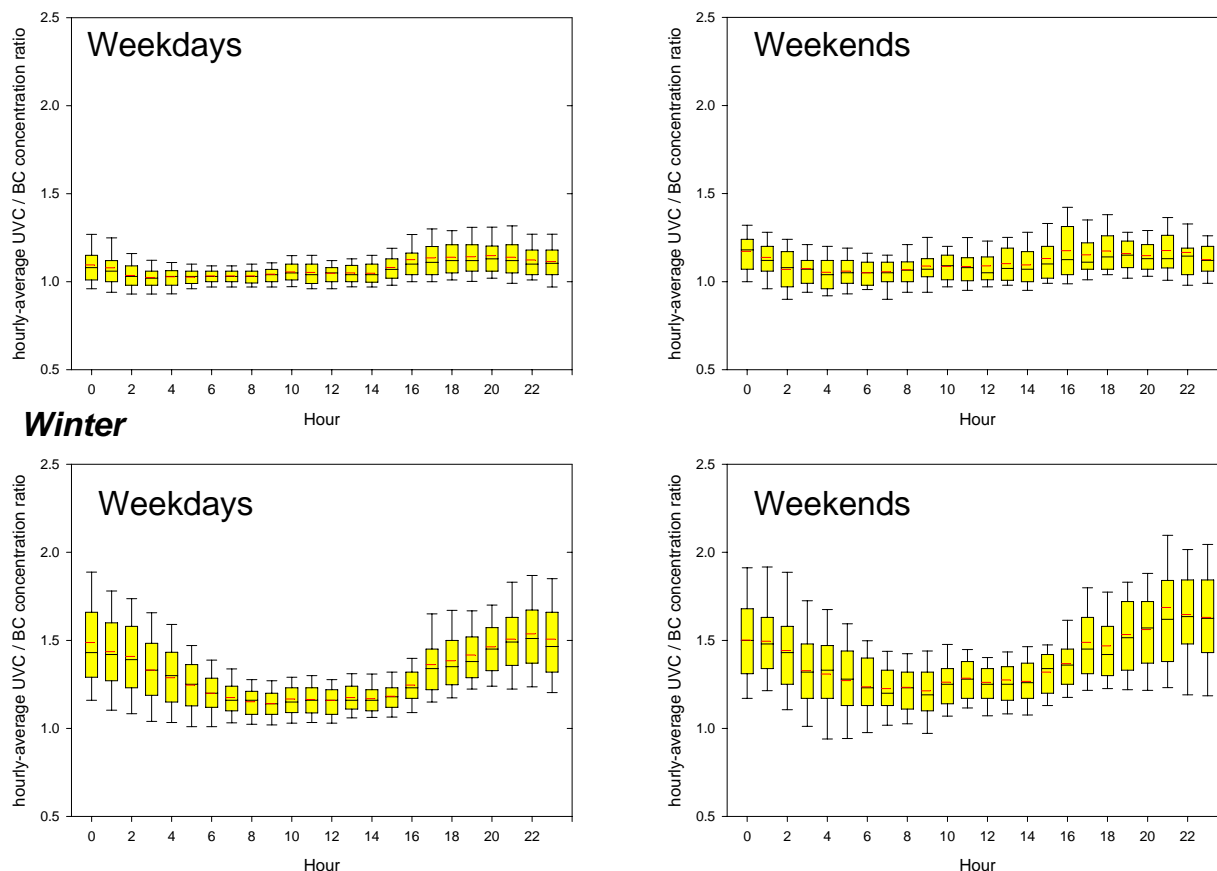
326 for the programmed Aethalometer absorption cross-sections; and UVC/BC and α differ by no
 327 more than 5% for UVC/BC ratios between 0.9 and 1.5. Thus, UVC/BC is a reasonable estimate
 328 for the Angstrom exponent. Summer UVC/BC ratios are near unity throughout the day with only
 329 modest evening enhancements. In contrast, winter UVC/BC ratios exhibit a distinct diurnal
 330 profile with a nighttime maximum and midday minimum. Median UVC/BC ratios are slightly

331
 332



333
 334 **FIGURE 8 Summer (left) and winter (right) diurnal profiles of the median hourly**
 335 **compensated BC for weekdays (blue line), weekends (green line) and the difference**
 336 **between weekday and weekend median values (red line).**

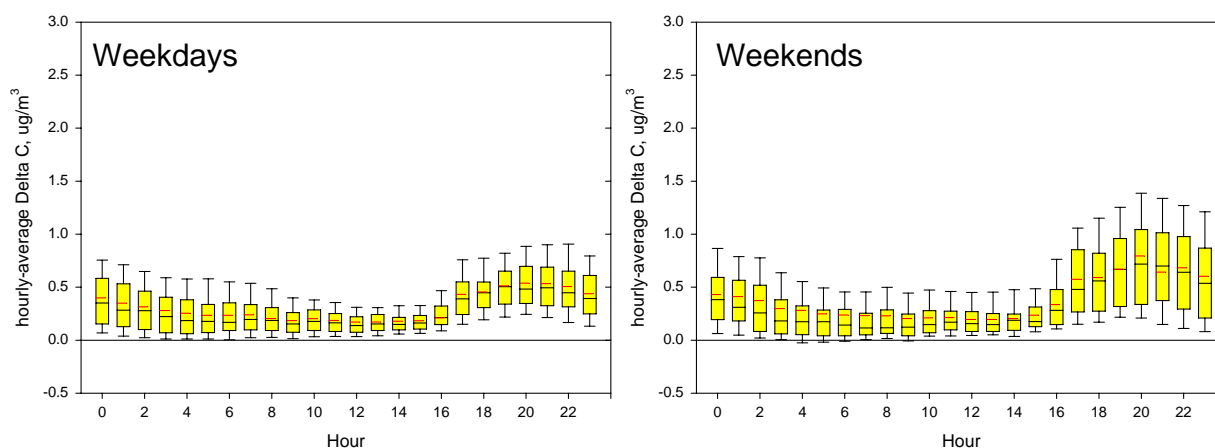
Summer



337
 338 **FIGURE 9 Summer (top) and winter (bottom) diurnal profiles of hourly-average UVC/BC**
 339 **ratios for weekdays (left) and weekends (right). Whiskers are 10th and 90th percentiles. The**
 340 **interior black line is the median and the red line is the arithmetic mean. Data from**
 341 **September 2005 through June 2008.**

342
 343
 344 higher on weekends compared to weekdays. This pattern suggests significant biomass
 345 combustion contributions during the winter evenings. Sandradewi *et al.* (6) observed similar
 346 summer and winter diurnal patterns for the Angstrom exponent of the absorption coefficient
 347 measured in an Alpine valley in Switzerland which is known to have both year round traffic
 348 influences and wintertime wood smoke influences. Summer median Angstrom exponents were
 349 in the range 1.0-1.1 and winter Angstrom exponent diurnal profile exhibited a maximum median
 350 value of 1.8 (which corresponds to a UVC/BC ratio of 2.0) at 0000 CET. In this study, we
 351 observed summer median UVC/BC ratios of 1.0-1.1 and a maximum in the median winter
 352 UVC/BC diurnal profile of 1.5-1.6 at 2200 EST.

353
 354 **Figure 10** shows winter diurnal profiles for ΔC which is defined as UVC minus BC. ΔC
 355 increases rather abruptly at 1700 EST with the evening ΔC concentrations higher on weekends
 356 compared to weekdays (and modestly higher on Sundays compared to Saturdays). The presence
 357 of the evening maximum in ΔC for both weekdays and weekends suggests the use of biomass



358
 359 **FIGURE 10 Diurnal profiles of hourly-average ΔC (UVC minus BC) for weekdays (left)**
 360 **and weekends (right). Whiskers are 10th and 90th percentiles. The interior black line is the**
 361 **median and the red line is the arithmetic mean. Data from September 2005 through June**
 362 **2008.**

363

364

365 combustion for space heating, and leads to our assignment of this source category as wood
 366 smoke. Modestly enhanced evening ΔC concentrations for weekends compared to weekdays
 367 suggests contributions from recreational fireplace use, which is also supported by the top 5th
 368 percentile of daily-average ΔC including a disproportionately high number of holidays
 369 (Thanksgiving weekend, Christmas eve and day, New Year's eve and day).

370

371 Sandradewi *et al.* (4-6) presented and applied a two-source model for interpreting multi-
 372 wavelength Aethalometer data. Generalizing their derivation to emission sources *A* and *B* with
 373 Aethalometer measurements at wavelengths λ_1 and λ_2 , using absorption cross-sections which
 374 vary inversely with wavelength (as was the case in our study), and assuming the mass absorption
 375 efficiencies at a given wavelength are the same for particles from sources *A* and *B*, the
 376 concentration $C_i(\lambda)$ apportioned to sources *A* and *B* at λ_2 are –

377

$$C_B(\lambda_2) = \frac{C(\lambda_1) - [\lambda_2 / \lambda_1]^{\alpha_A - 1} C(\lambda_2)}{[\lambda_2 / \lambda_1]^{\alpha_B - 1} - [\lambda_2 / \lambda_1]^{\alpha_A - 1}}$$

$$C_A(\lambda_2) = C(\lambda_2) - C_B(\lambda_2)$$

378 where α_i is the Angstrom exponent for source *i*. Sandradewi *et al.* (11) applied the two source
 379 model to traffic (source *A*) and wood smoke (source *B*) emissions and assigned Angstrom
 380 exponents $\alpha_A = 1.01$ and $\alpha_B = 1.86$ based on literature values and their own estimates. For our
 381 measurements at $\lambda_1 = 370$ nm (UVC) and $\lambda_2 = 880$ nm (BC) the BC concentration apportioned to
 382 wood smoke (WS) and traffic (T) reduces to –

383

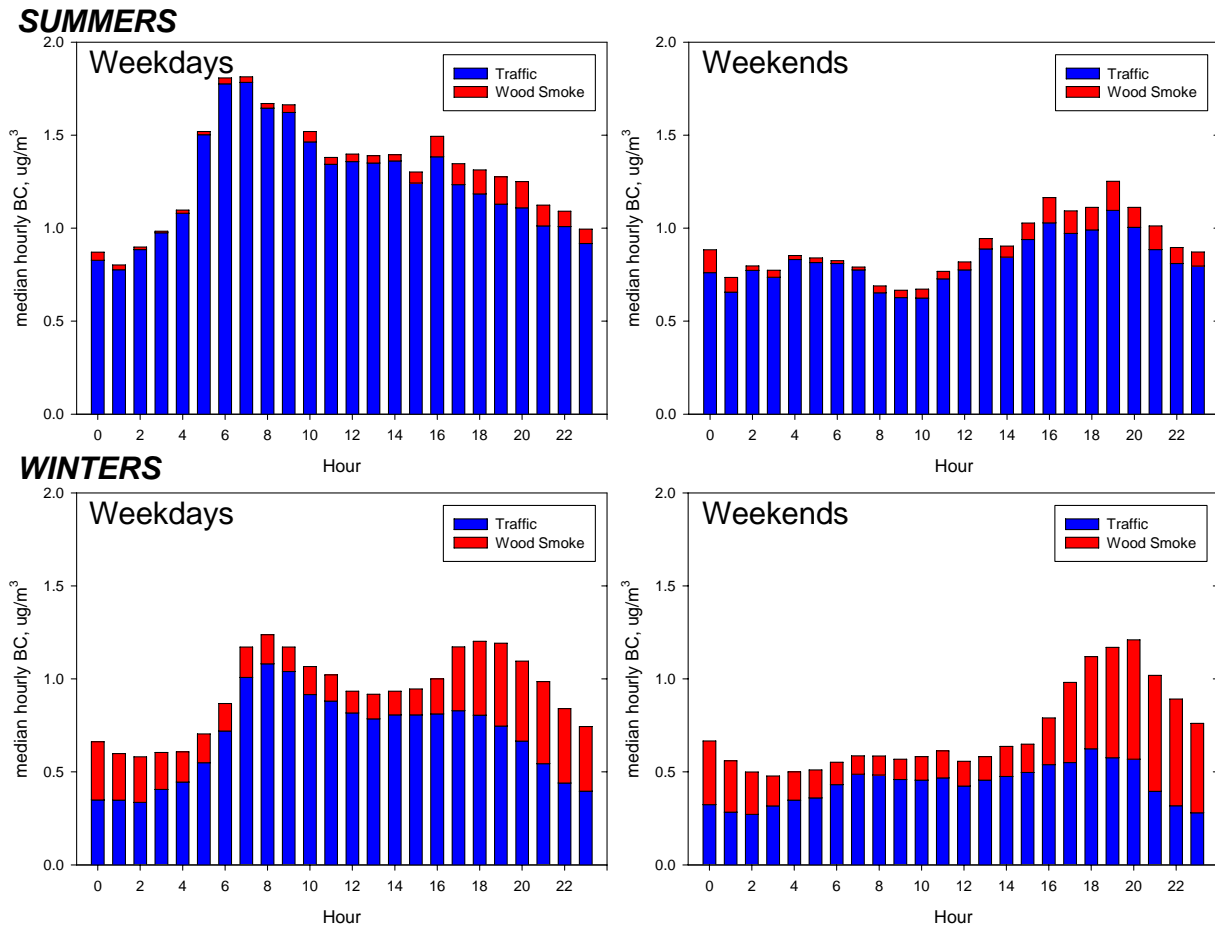
$$BC_{WS} = 0.91 \times [UVC - 1.01 \times BC]$$

$$BC_T = BC - BC_{WS}$$

384 Using this approach, wood smoke accounted for 15% of the BC over the entire study period.
 385 Wood smoke was 5% of the summer BC and 28% of the winter BC. **Figure 11** shows diurnal
 386 profiles for the median BC contribution from wood smoke and traffic (or, more generally, mobile
 387 sources). There is very little wood smoke contribution in the summer. In contrast, there are
 388 significant wood smoke contributions in the winter with the wood smoke BC on weekday nights
 389 approaching 50% of the total BC and the wood smoke BC on weekend nights representing more
 390 than 50% of the total BC.

391
 392 **Conclusions**

393
 394 An examination of five years of fine particle Aethalometer black carbon data for the North End
 395 site in Boston demonstrates dominant contributions from mobile sources but also significant
 396 contributions from biomass combustion, likely wood smoke from space heating and recreational
 397 fireplace use, in the winter evenings. While such biomass combustion contributions are
 398 generally understood to be important in urban centers in the Pacific Northwest such as Seattle
 399



400
 401 **FIGURE 11 Summer (top) and winter (bottom) diurnal profiles of median hourly BC**
 402 **contributions by traffic and wood smoke sources for weekdays (left) and weekends (right).**
 403 **Data from September 2005 through June 2008.**

404
 405

406 (13), it is now clear that biomass combustion can also be a significant contributor to black carbon
407 in urban centers in the Northeastern U.S. such as Boston. This finding has implications to the
408 interpretation of black carbon as a surrogate for diesel particulate matter in urban centers, and
409 thus to black carbon exposures ascribed to mobiles sources.

410

411 **ACKNOWLEDGEMENTS**

412

413 The authors gratefully acknowledge the Massachusetts Department of Environmental Protection,
414 Bureau of Waste Prevention, for providing the raw Aethalometer data.

415

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463

**Appendix H: Annual and Seasonal Boston/Logan
Wind Roses, 2000-2012.**

Appendix H.

Boston/Logan Wind Roses

Wind speed and direction play a significant role in the seasonal and annual variation of BC. These wind rose plots show the seasonal and year-to-year variation in wind.

Figure 1 shows all wind data for 2000-2012. Figure 2 shows wind for March, and Figure 3 for August for the same time period; these two months are typically the lowest and highest BC months. Figure 4 shows wind for 2009-2012, the duration of the second spatial analysis.

The remaining plots show each year of wind from 2000 to 2012.

Figure 1. 2000-2012

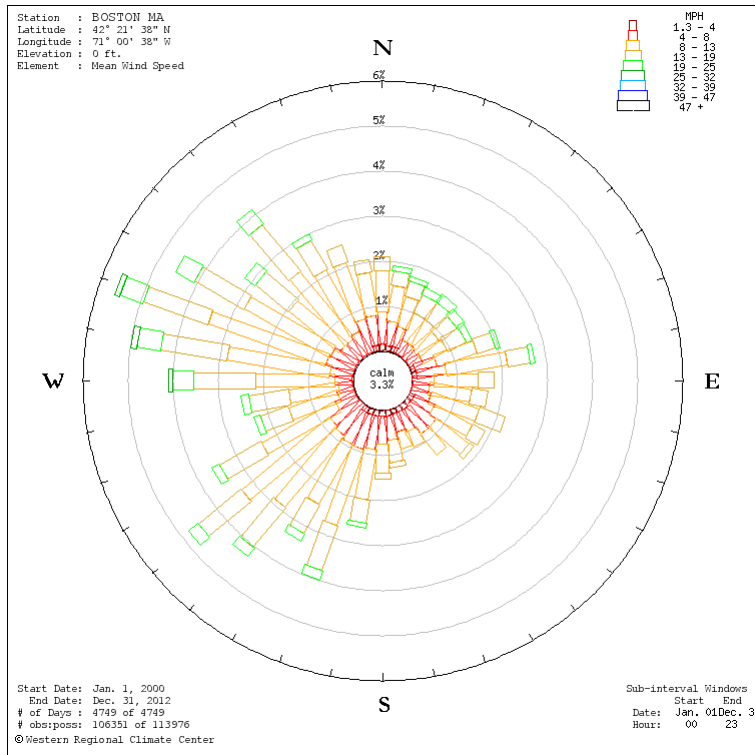


Figure 2. March 2000-2012

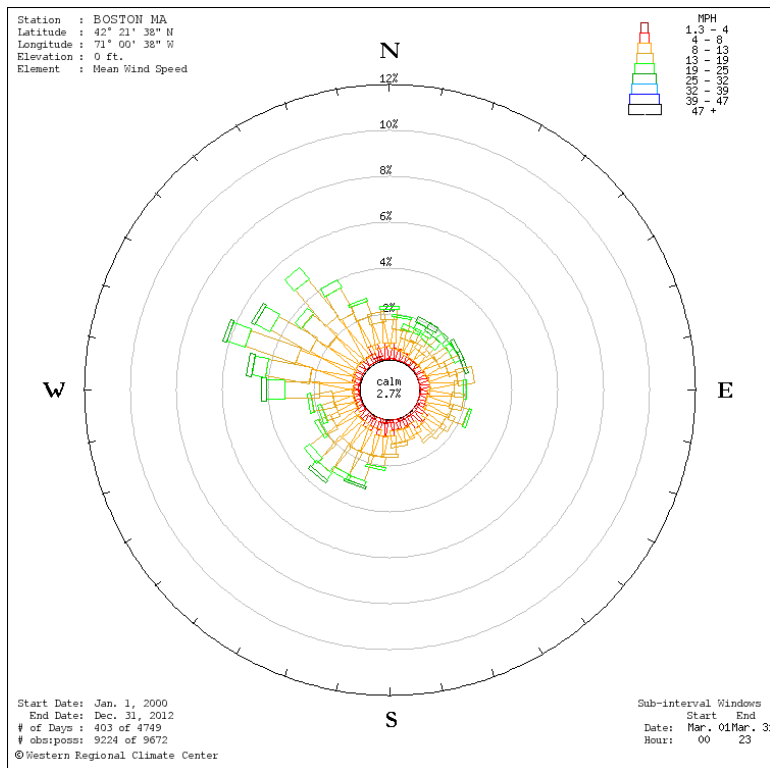


Figure 3. August 2000-2012

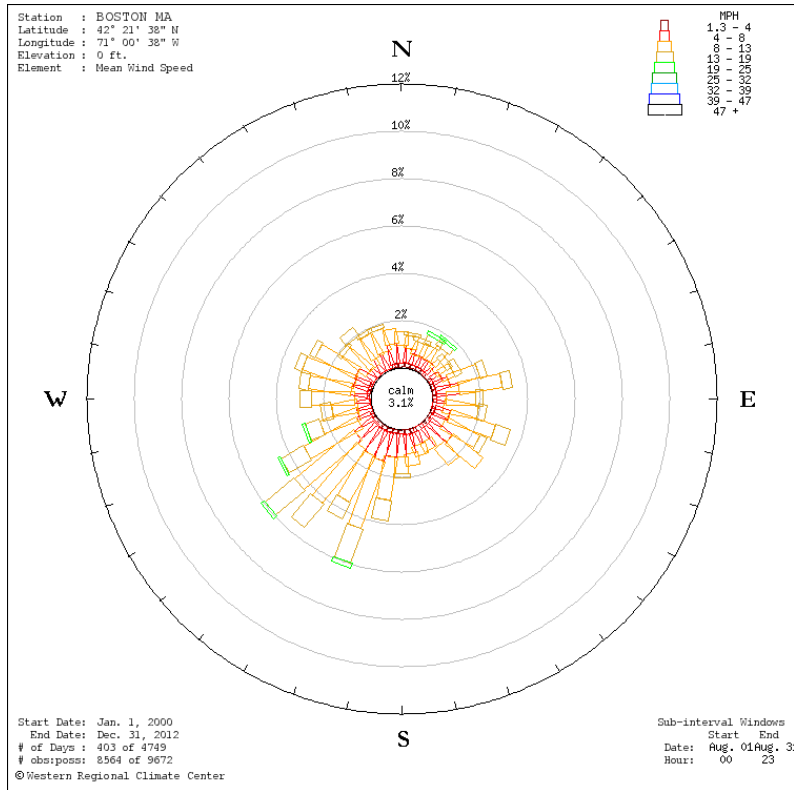
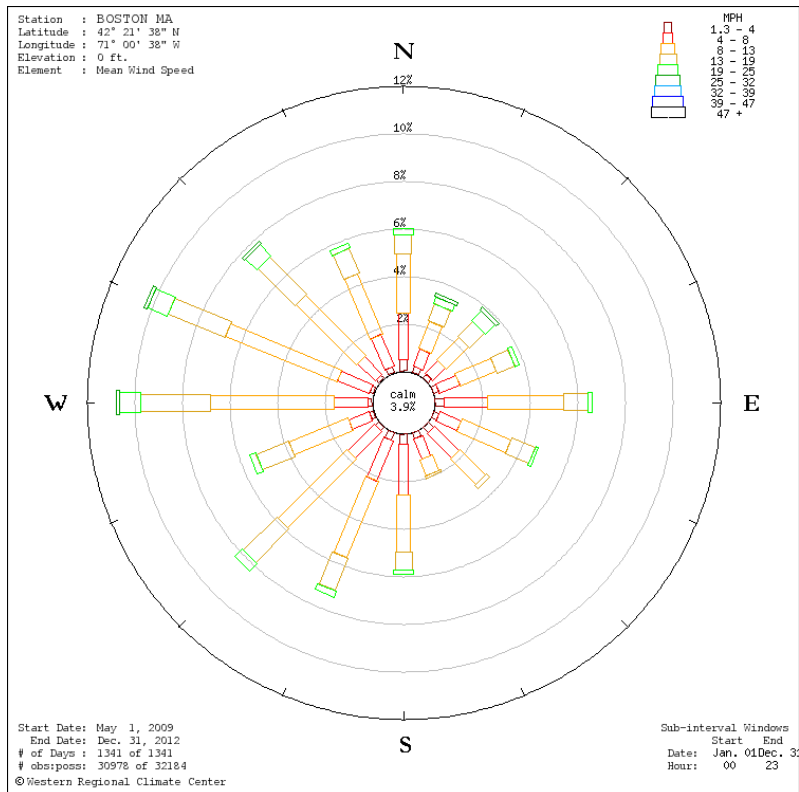
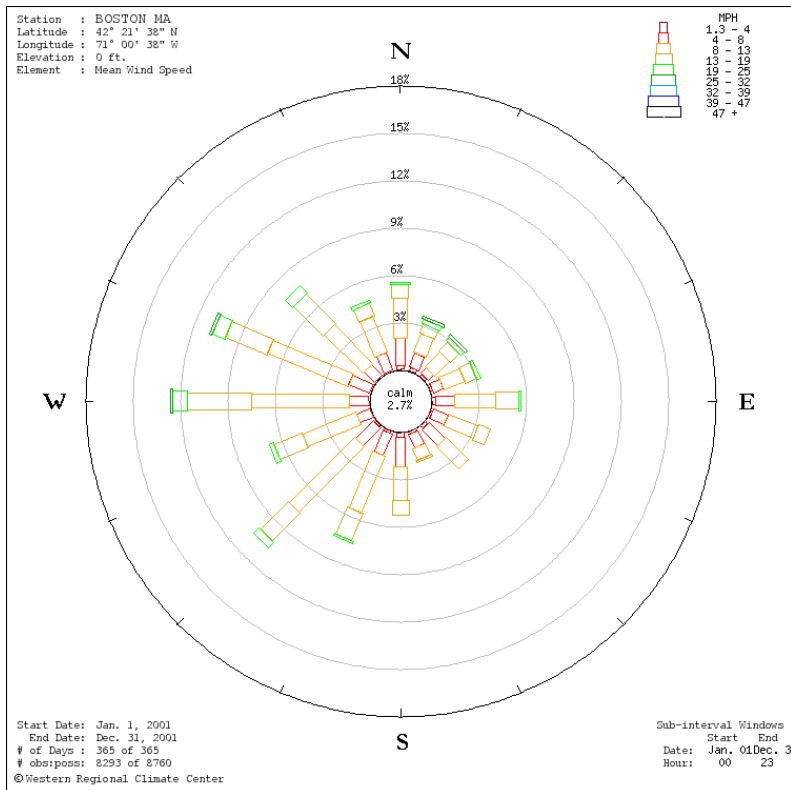
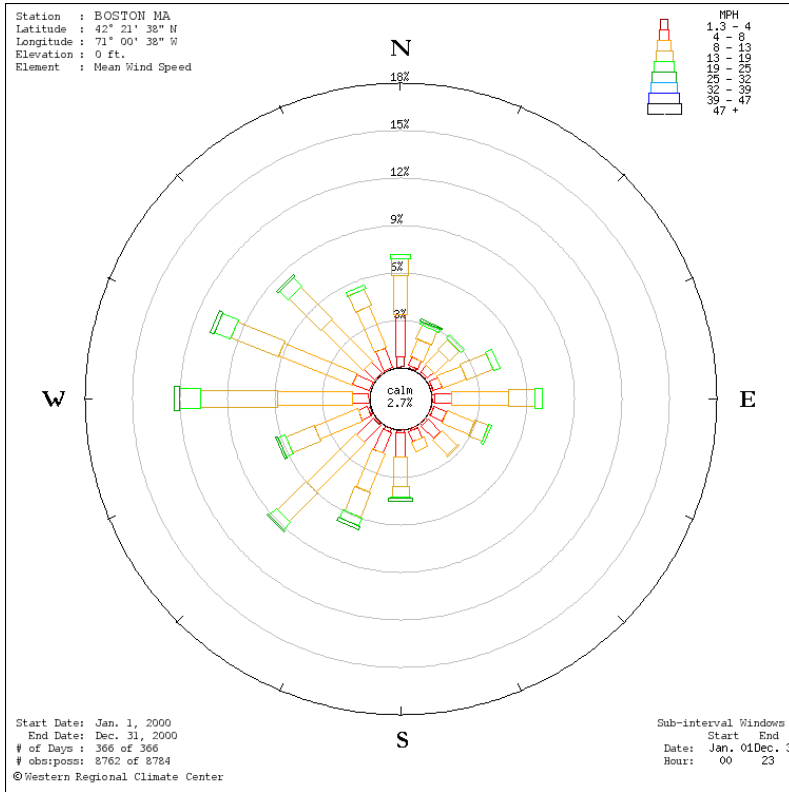


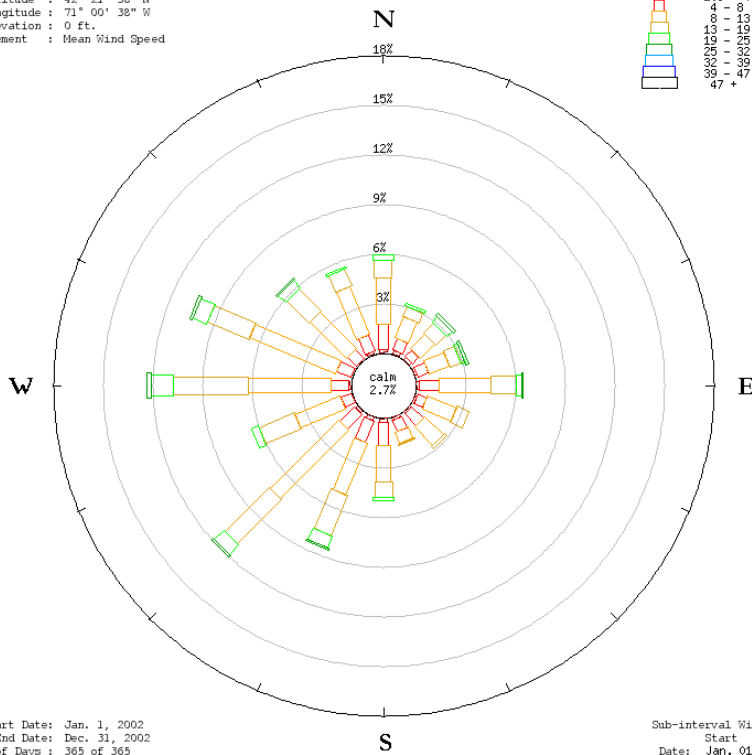
Figure 4. 2009-2012



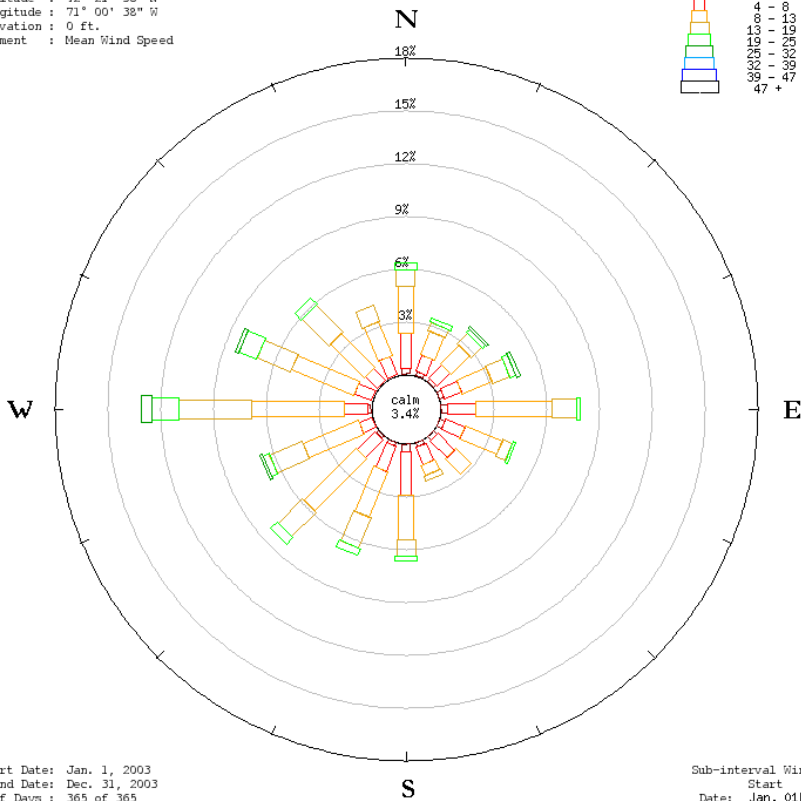
Annual Wind Roses, 2000-2012

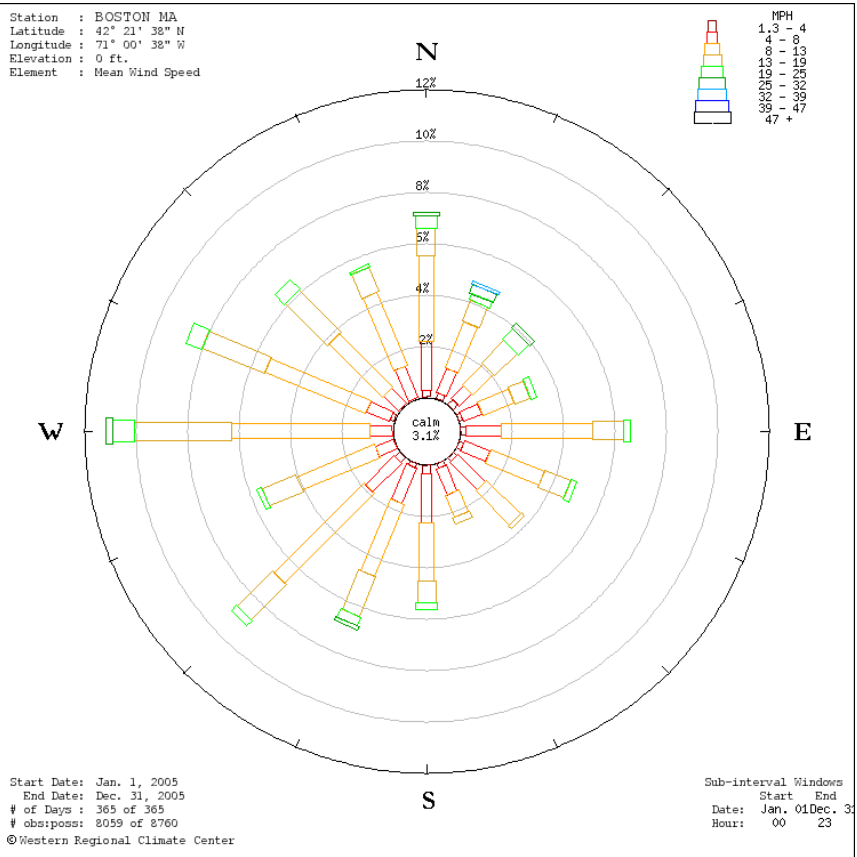
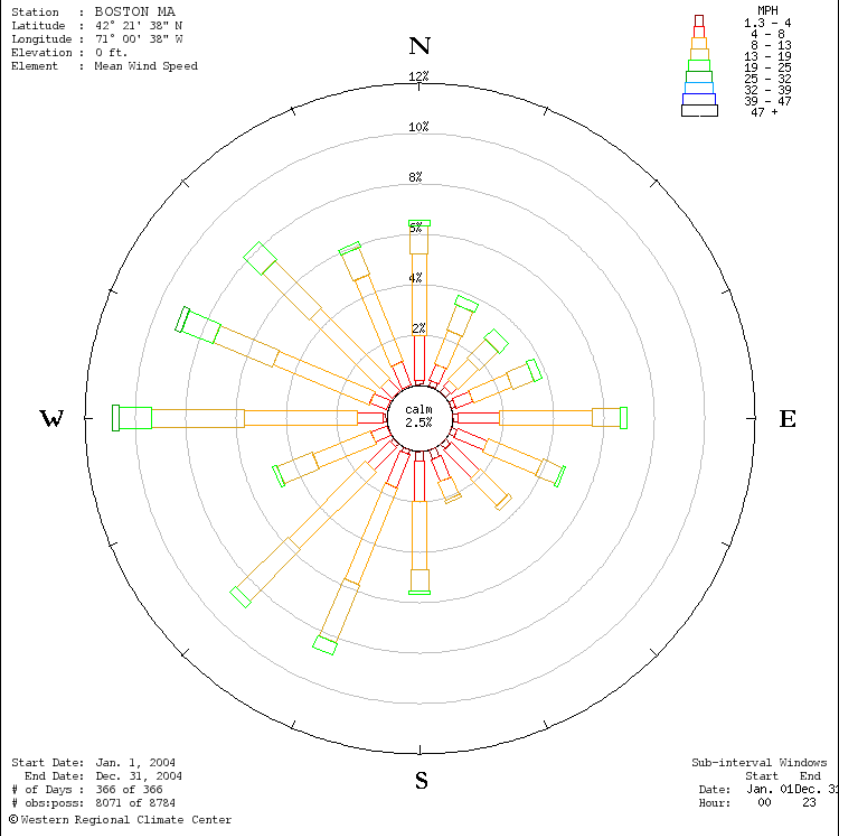


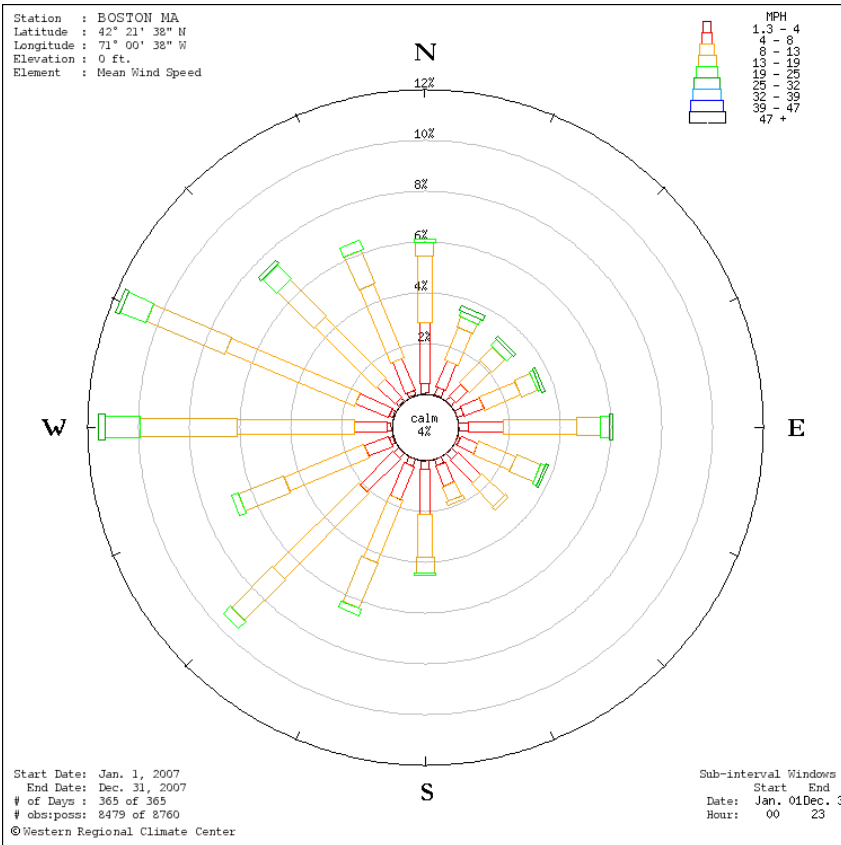
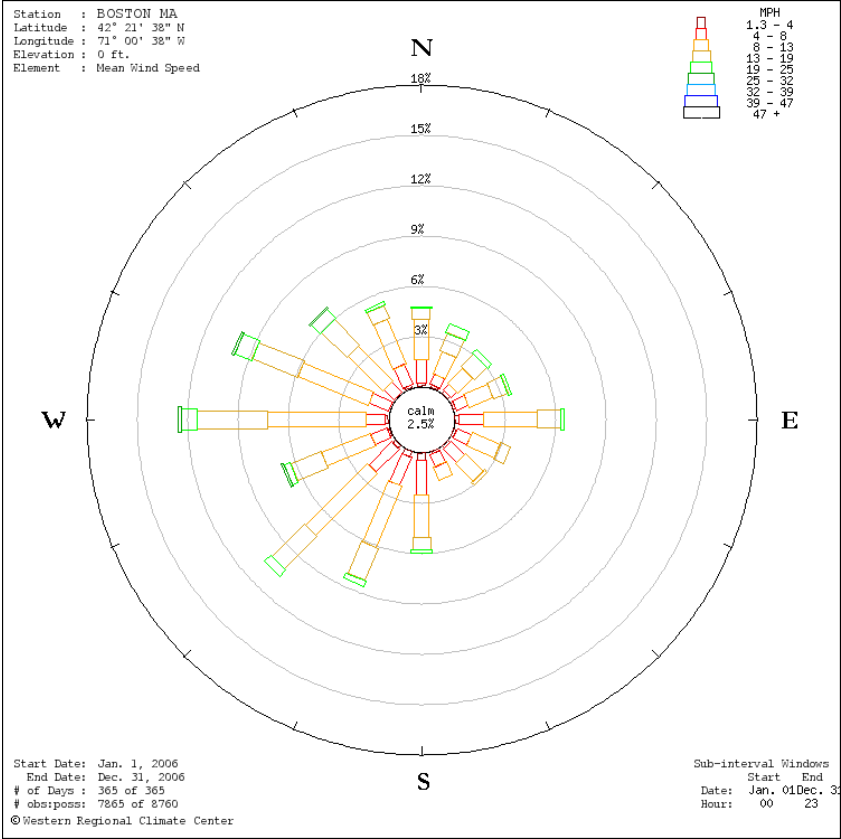
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 Latitude : 42° 21' 38" N
 Longitude : 71° 00' 38" W
 Elevation : 0 ft.
 Element : Mean Wind Speed



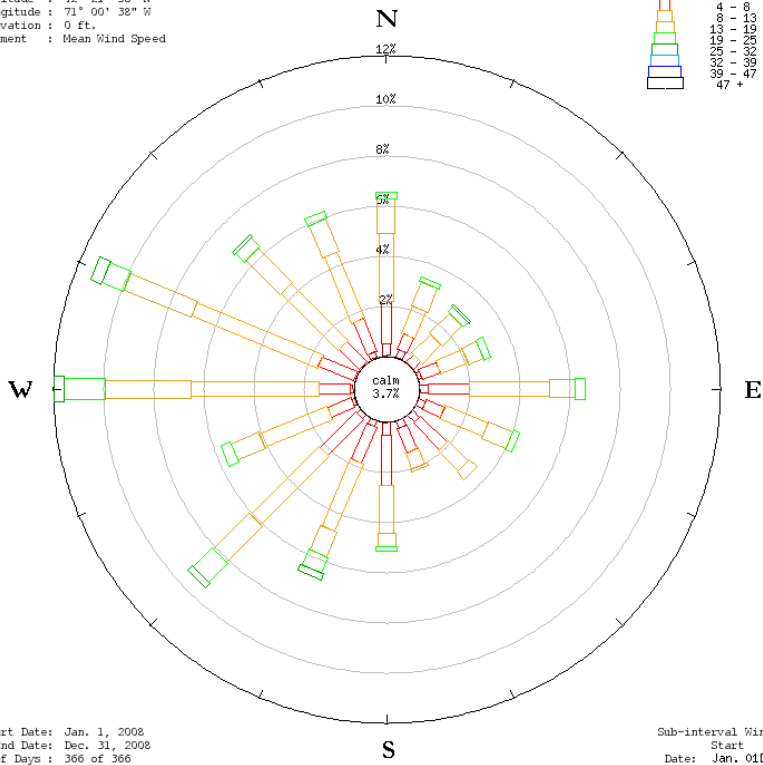
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 Latitude : 42° 21' 38" N
 Longitude : 71° 00' 38" W
 Elevation : 0 ft.
 Element : Mean Wind Speed



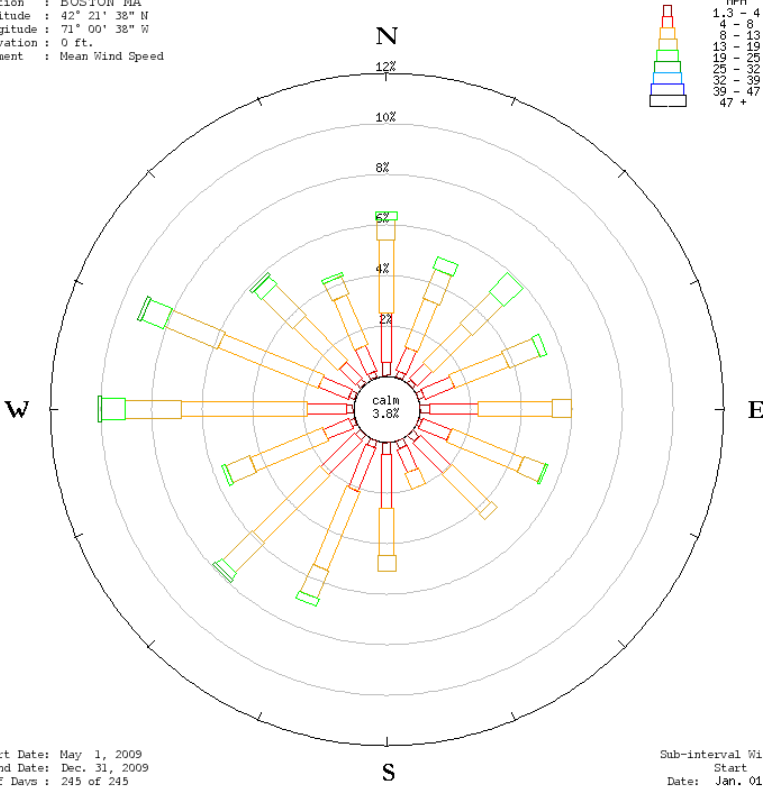




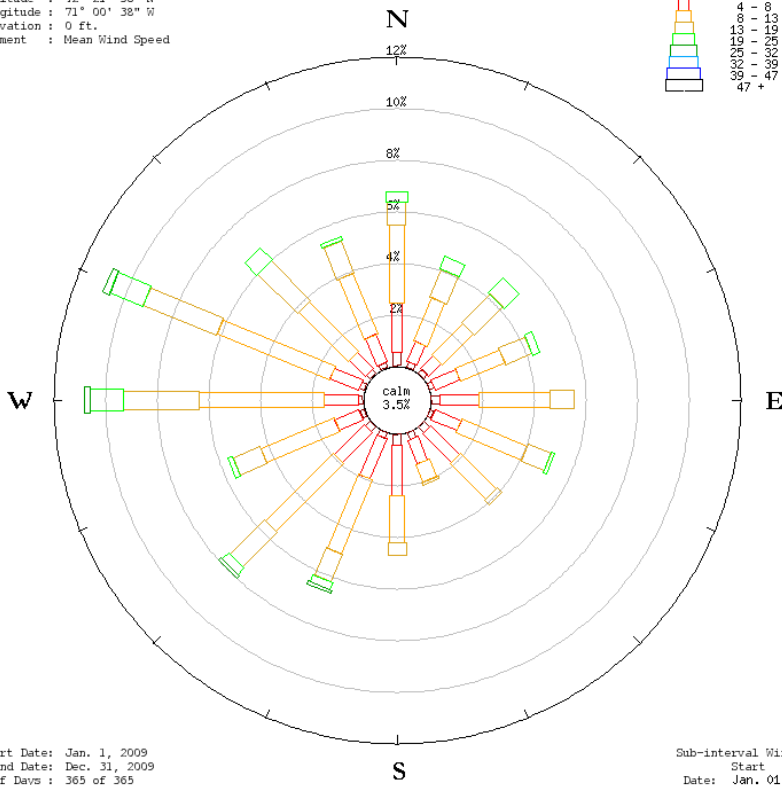
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 Latitude : 42° 21' 38" N
 Longitude : 71° 00' 38" W
 Elevation : 0 ft.
 Element : Mean Wind Speed



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 Latitude : 42° 21' 38" N
 Longitude : 71° 00' 38" W
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 Latitude : 42° 21' 38" N
 Longitude : 71° 00' 38" W
 Elevation : 0 ft.
 Element : Mean Wind Speed



Station : BOSTON MA
 Latitude : 42° 21' 38" N
 Longitude : 71° 00' 38" W
 Elevation : 0 ft.
 Element : Mean Wind Speed

