

# **Evaluation of Spatial Gradients and Temporal Trends of Black Carbon in Boston MA.**



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## **1. Introduction.**

The Massachusetts Dept. of Environmental Protection (MA-DEP), Bureau of Waste Prevention, is applying for a grant to assess the spatial and temporal trends of existing black carbon (BC) data in the metropolitan Boston area. BC has been shown to be a useful indicator of local mobile source aerosol emissions in urban areas (Janssen et al., *Atmos. Environ.* 1997, 8:1185–1193).

Local mobile sources in large urban areas contribute to elevated levels of a wide range of air toxics pollutants, including PM from both automotive (spark ignition) and diesel vehicles. PM from mobile sources has the potential to be highly toxic, and is thought to be a major factor in the observed PM health effects reported by a wide range of epidemiological studies over the last decade.

From a policy perspective, an improved understanding of both the spatial patterns (gradients) and long-term (8-10 year) temporal trends of “tailpipe” related PM in large urban areas as represented by BC is important both for implementation and assessment of control strategies. It also aids in understanding the exposure dynamics of potential environmental justice-related “hot-spots” such as the Dudley Square area of Roxbury (Boston). Finally, understanding trends and gradients is critical for understanding and improving estimates of exposures used in health effect studies; for example, the Harvard-led EPA Boston PM Center has used MA-DEP air pollution data in several studies over the last decade. However, when a single monitor is used to represent an urban area for locally generated pollutants such as BC, exposure mis-classification can occur; this can bias estimates of health effects toward lower (less hazardous) values (Kunzli et al., *EHP* 2005, 113:201–206). This project will directly support EPA’s Strategic Plan Goals 1.1 (healthier outdoor air) and 1.1.2 (Reduced risk from Toxic air pollutants).

MA-DEP will partner with NESCAUM on this project. NESCAUM staff have a wide range of expertise in data analysis, including assessment of data quality and potential bias or artifacts in BC measurements, the primary metric for this project. NESCAUM also collected much of the data for the 2003 spatial study, and is familiar with the strengths and weakness of that work.

## **2. Background and Motivation.**

In this proposal, we describe an approach for analysis of existing aerosol data (PM<sub>2.5</sub> and BC) in the greater Boston MA area that will lead to a better understanding of both urban gradients and temporal trends of BC (locally generated mobile source aerosols, including diesel PM) over the last decade. Quantification of BC spatial gradients and temporal trends are related tasks, since both effect characterization of mobile-source aerosol exposures over space and time. Data sources include monitors at on-going MA-DEP sites and the Harvard School of Public Health's (HSPH) EPA PM-center

The primary metric to be used for this analysis is BC from Aethalometers™, a measurement that is in common use nationally as part of the EPA National Air Toxics Trends Stations (NATTS) program. The Aethalometer is a simple measurement of how dark the aerosol is; the more graphitic carbon soot (primarily from diesel in urban areas but also from spark ignition vehicles), the higher the reported BC concentration in micrograms per cubic meter.

## Trend Analysis.

For trend analysis, Boston is unique in having three long-term sites with BC measurements; two are run by MA-DEP (Roxbury and the North End), and one is run by HSPH near Brigham Circle. The North End site started measuring BC in mid-2003; the other two sites started in 1999, making them some of the longest continuously running urban BC sites in the country and providing a unique opportunity for trend analysis with almost a decade of data. All three sites will continue through at least 2008.

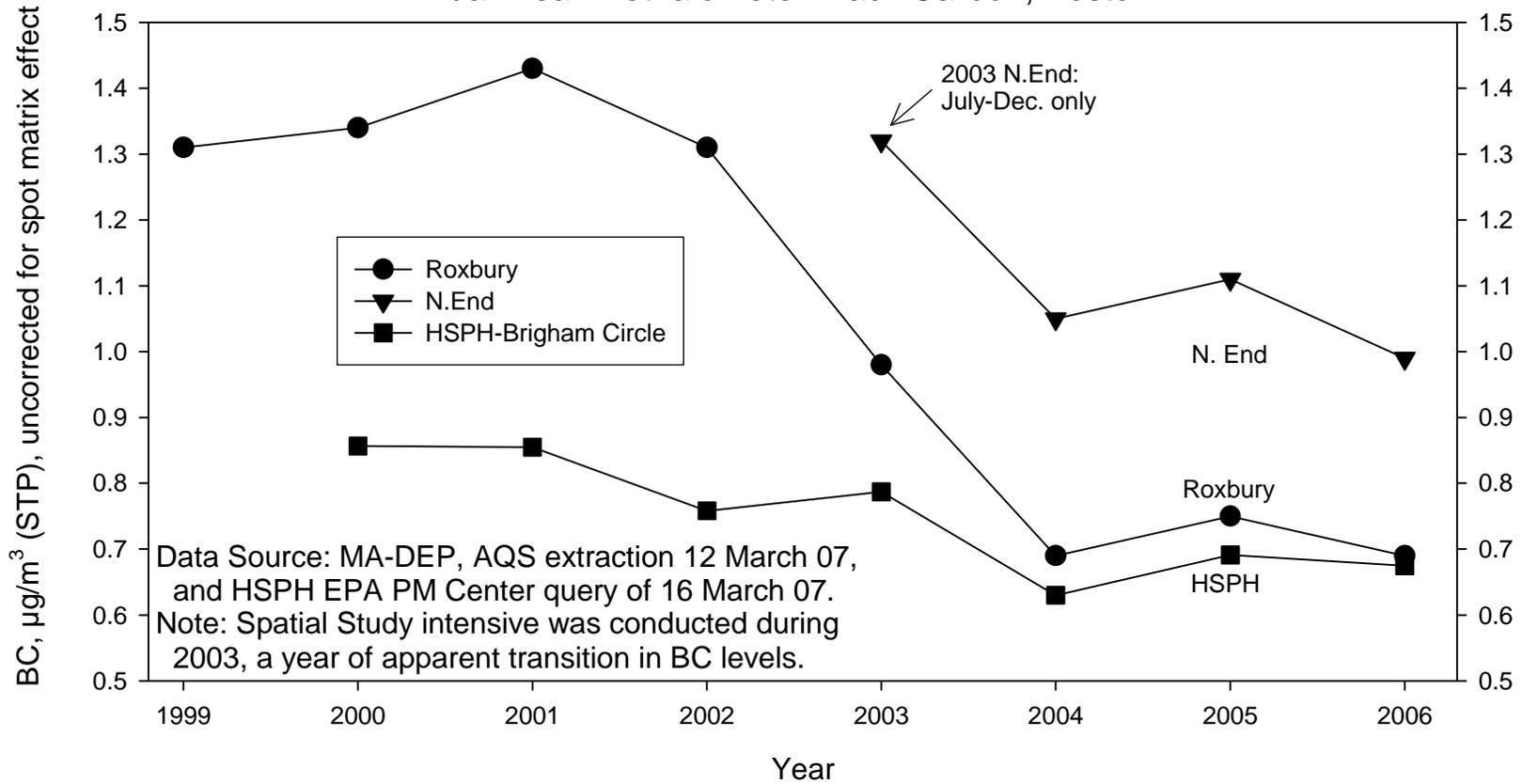
### Site Descriptions for key BC trend sites.

- The Roxbury MA-DEP site is located one block away from Dudley Square, a busy MBTA commuter bus station. A large “bus barn”, the Bartlett yard, was located approximately 300 meters from Dudley Square until it was shut down in 2004. The site is classified as neighborhood scale; probe heights are approximately 4 to 5 meters above ground.
- The North End MA-DEP site is located in a residential neighborhood on the top of a 3-story building, 30-40 meters from the entrance of two traffic tunnels that go to Logan Airport and points north. The probe height is 14 meters above ground.
- The HSPH PM-Center site is near Brigham Circle in Boston, approximately 1.5 km NW of the Roxbury site. However it is approximately 25 meters above street level, making it more of an urban scale site for fine-mode aerosols.

A preliminary evaluation of BC trends using available MA-DEP data is shown in Figure 1, using data extracted from AQS for the two Boston BC sites. It is clear that there was a substantial reduction of BC at the Roxbury site between 2002 and 2004, even with the potential of changing measurement artifacts over this time period. It is not clear from these limited data if the reduction was city-wide or primarily at Roxbury (the 6 months of data from the North End for 2003 are suggestive but not conclusive).

It will be one of the goals of this project to determine the spatial scope of the reduction in BC observed at Roxbury by (a) bringing in data from other sites such as HSPH and extending the N.End BC back to 2001 using reflectance analysis on FRM filters, and (b) assessing what control programs may have contributed to the apparently dramatic improvement in air quality both at the Roxbury site and others in downtown Boston.

### Annual Mean Aethalometer Black Carbon, Boston MA



### Spatial Gradient Analysis.

For the spatial gradient analysis component of this project, we will primarily use one year of BC data collected at 6 sites during 2003 for an initial study conducted by NESCAUM to assess the spatial gradients of BC across the greater Boston metropolitan area. That study was primarily designed to assess the impact of urban mobile-source related hot-spots, and also to define how rapidly the “urban excess” BC dropped off as a function of distance from downtown Boston. Figure 2 shows the site locations. However due to lack of any project funding, only a simplistic and preliminary data analysis was performed, and only on a portion of the entire dataset. The data analyzed also included significant instrument BC artifacts that have not been corrected or accounted for. Still, the analysis performed to date demonstrate both large gradients and the utility of BC as a good indicator for local mobile aerosol sources in urban areas. Figures 3 and 4 below show nine months of BC data for six sites, three in the core urban area, and three further out to a regional background site 36 km NW of Boston. Figure 2 is the distribution of the hourly data, and shows a factor of three in BC concentrations between downtown Boston and a regional background site (Stow MA).

Figure 3 shows the same data as diurnal plots stratified by work and non-workdays. A large morning rush-hour BC peak is present for the urban sites on workdays, but not non-workdays. The only significant difference in local sources between workdays and non-workdays for the 5 am to 9 am morning (rush-hour) period is mobile source activity; this demonstrates that BC dynamics at these core urban sites are due mostly to local primary mobile source emissions. As a result of early analysis of these data, a “saturation” study was also conducted for two months during the summer of 2003 to assess the spatial variability of BC within the urban core - generating a unique data set over a much smaller spatial scale than the year-long project. Analysis of data from these 9 urban sites will also be included in this project.

Figure 2. Six year-long “core” Spatial BC Monitoring Sites.

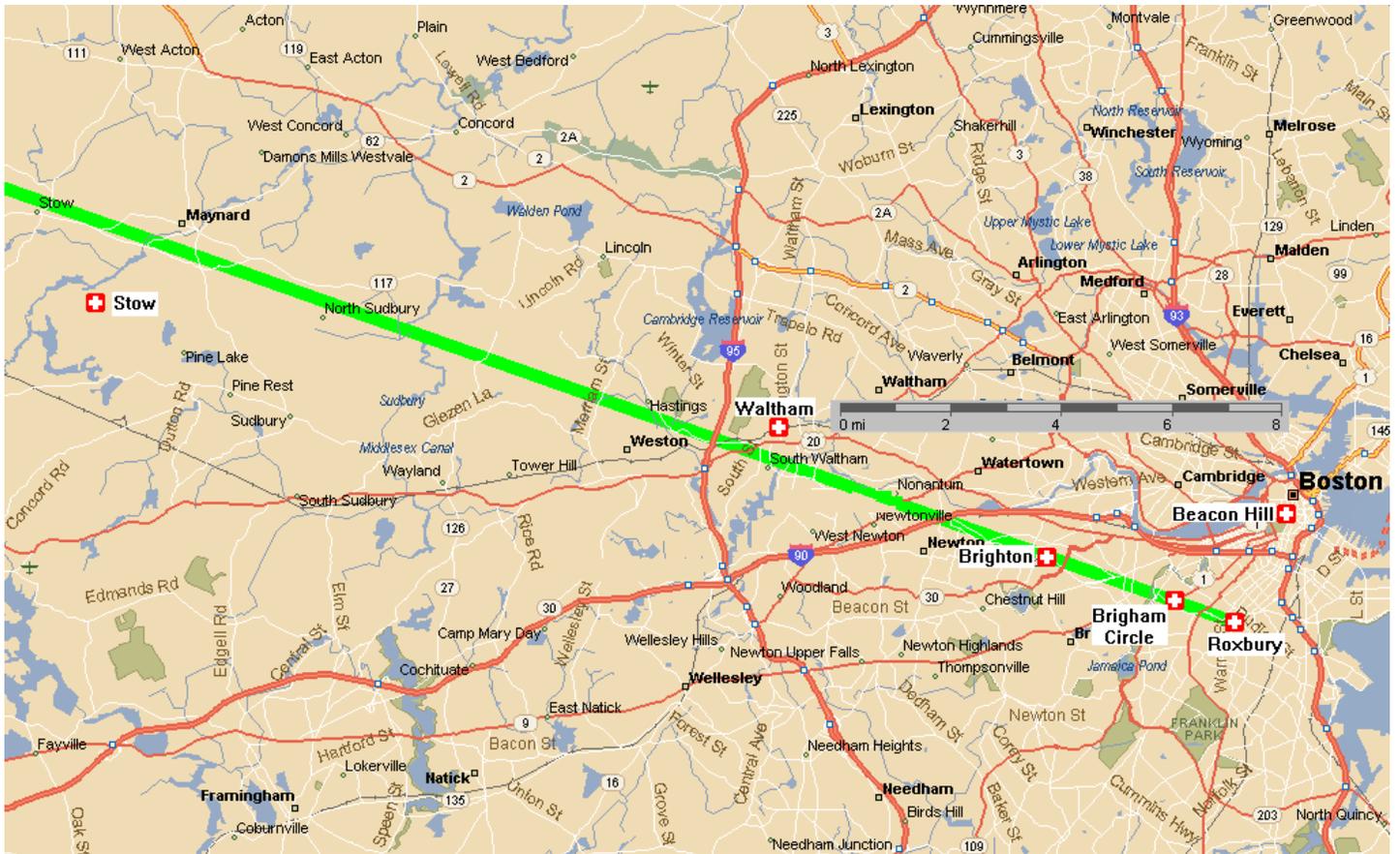


Figure 3. Preliminary Spatial Gradients using uncorrected and incomplete data.

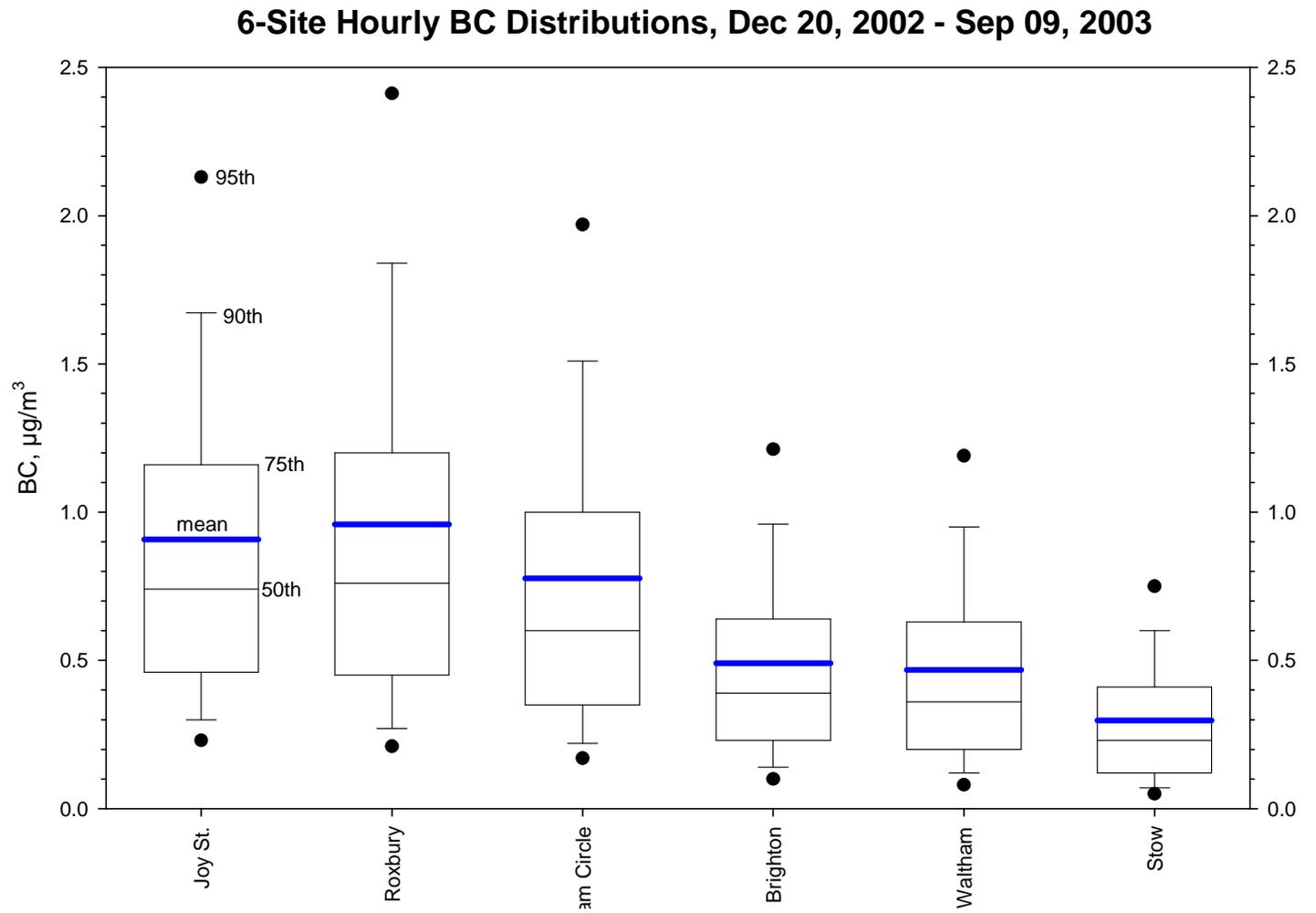
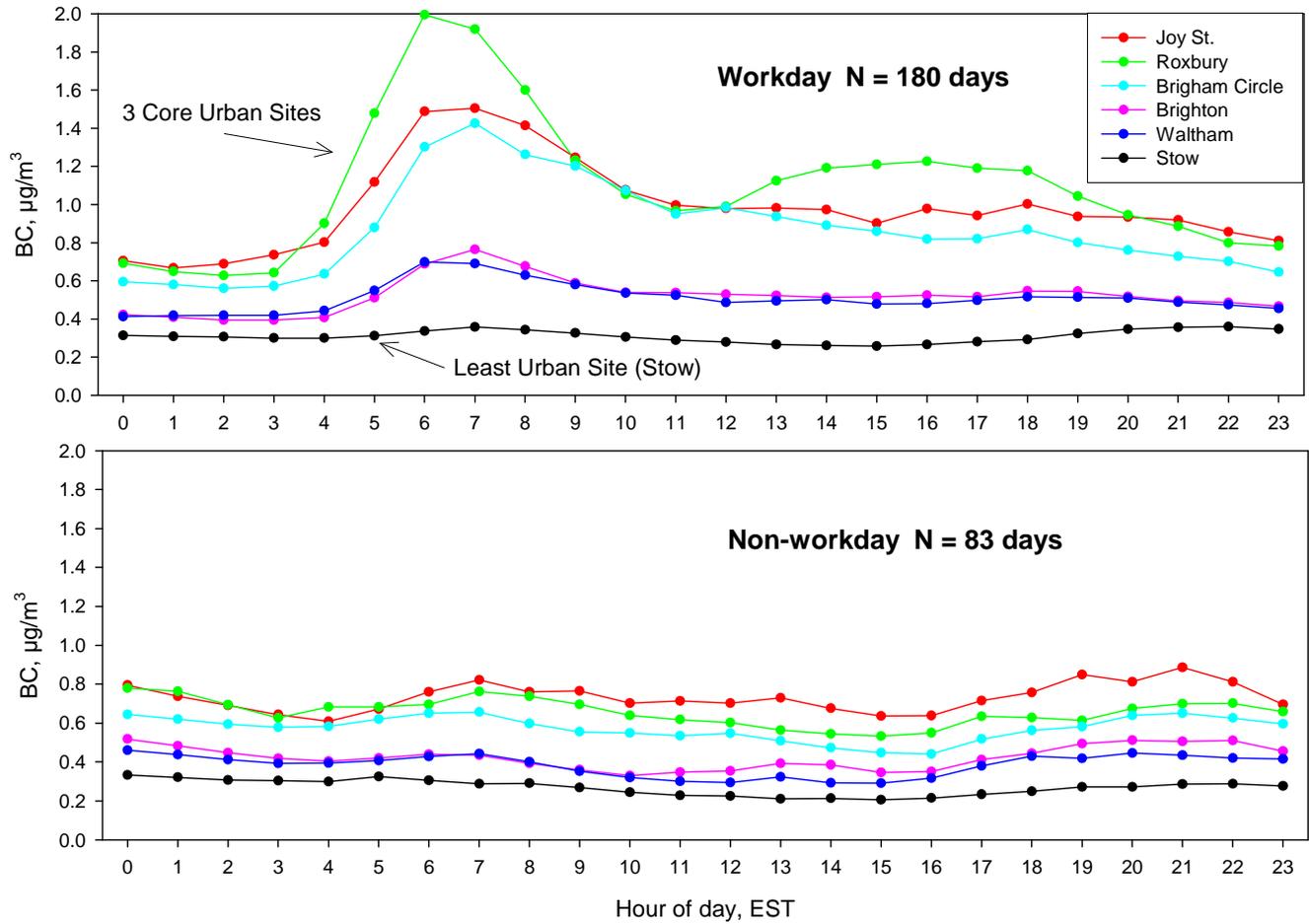


Figure 4.

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



### 3. Project Objectives.

This project has two main and related data analysis objectives:

1. To better characterize the spatial gradients of BC as a marker for local mobile source aerosol (primarily diesel) by rigorously analyzing existing BC and PM<sub>2.5</sub> data from multiple sites, incorporating known method artifacts and biases in the analysis. An additional objective for this component is to determine how representative the 2003 data is of present-day BC gradients given the substantial drop in BC observed at Roxbury between 2002 and 2004.
2. Analyze data from two MA-DEP and the HSPH site to determine the temporal trend in BC from 2000 to 2008, and to the extent possible determine if the distinct downward trend observed at Roxbury extends to other urban Boston areas. The timing of implementation of various mobile source aerosol reduction strategies in Boston and specifically in Roxbury will be evaluated to see if they are plausible factors in the observed BC downward trend.

These two objectives are related in the sense that if there have been real and substantial improvements in BC concentrations at some sites since 2003 when the spatial gradient study data were collected, how might those trends effect the interpretation of the spatial gradients observed in 2003.

For example, is Roxbury BC still elevated compared to HSPH, a more urban-scale site? We will be able to answer those kinds of questions with this analysis.

#### **4. Project Tasks and Deliverables.**

##### Temporal Trend Analysis.

First, we will investigate the long-term trends in BC using data from the two MA-DEP and the HSPH sites. Substantial trends exist for at least some sites as noted earlier. There are four major tasks, framed here as questions to be addressed:

1. Is the trend real, a method or network artifact, or some of each?
2. Assuming it is real, is it statistically significant after any major measurement artifacts are removed?
3. Is the reduction local (e.g., just Roxbury), or did it occur across the urban core of Boston?
4. To what extent can we determine which control measures contributed to the decline in BC?

## Artifacts.

As part of the trends analysis, we will investigate possible method artifacts (task #1) in multiple ways. First, using hourly data we can separate out weekdays from weekends and the weekday rush-hour period (5 to 9am local time) from the same time period during weekends. Because there is minimal effect on local mobile source BC during weekends as noted previously, we would expect to see a similar (presumably sharper) drop in weekday rush-hour BC for this stratified trend analysis and minimal or no trend in the weekend BC trend. If a substantial drop is also seen in the weekend trend data (along the lines of the annual trend), that would indicate a method artifact is driving the trend. In addition to being a robust diagnostic tool, this analysis approach will also quantify the trend in sub-daily exposures; we hypothesize that the rush-hour weekday BC trend is substantially stronger than the annual average trend. Quantifying this would also be valuable in the context of health effects assessment. Trends can also be broken down by season, although comparisons between seasons are subject to substantial bias from the Aethalometer's spot matrix effect (discussed in the section on spatial gradient analysis below, since this bias is not a significant factor in analysis of annual BC means).

Other possible artifacts in these trend data include biases between different Aethalometers and Aethalometer configurations (up to 20-30%), and analog data logger offsets (typically no more than 10% of mean BC). These can be investigated and controlled for if necessary, using historical site records and instrument meta-data. Smoothed daily or weekly time-series plots can be used to look for step-function changes in BC, as well as investigate dynamics at the sub-annual level (e.g., if 2003 is a "transition" year for BC, what does the year look like on a monthly or weekly basis?). We will also perform trend analysis on the HSPH BC data set, which was generated independently from MA-DEP (but uses the same measurement method). If these data show a similar trend, then we have increased confidence that the change for the Roxbury site is not driven by artifacts.

Finally, all three sites have daily or third-day PM<sub>2.5</sub> gravimetric measurements; these data will be analyzed for trends, but we may not see a significant change in PM<sub>2.5</sub> (BC is almost an order of magnitude more specific to local mobile source emissions than neighborhood scale PM<sub>2.5</sub>). We will evaluate the relationship between Roxbury PM<sub>2.5</sub> and other sites during this interval to determine the value of this approach for sites without BC data. There is additional data of interest that is available for all or much of the 9-10 year period of interest. Roxbury also has speciation data from the Speciation and Trends Network (including elemental carbon), as well as CO, NO<sub>x</sub>, SO<sub>2</sub>, and NATTS gaseous air toxics data. The HSPH site has filter-based elemental and organic carbon measurements from 2000 through 2005, the core period of interest. To the extent that these supplemental data are useful in supporting the BC trend analysis, these other pollutants will also be analyzed for trends during this time period.

## BC Trend Significance.

We will perform various tests of significance on the BC trend data for these three sites (question #2) using various analysis techniques, including those described in EPA web training courses (<http://www.epa.gov/quality/trcourse.html>). For Roxbury and HSPH, we will have data through much or all of 2008, giving us at least 4 years before and after the apparent 2003 transition year. ANOVA will be performed on the four pre-post 2003 annual means and also on the individual annual means themselves using the underlying hourly data. Since the Roxbury BC drop pre-post 2003 is large, corrections for climatology will not be done unless preliminary analysis indicate that it is necessary to improve the trend estimate of significance. It must be noted here that any statistical assessment of trend significance does not take into account the potential influence of method bias over time; statistical significance is likely to be high simply because “N is large” (the hourly data set has over 8000 observations per year and 9-10 years of data for the Roxbury and HSPH sites). The interpretation of statistical results must incorporate the impact of possible changes in method bias over this time period; the latter is likely more important in assessing how “real” any observed trends may be.

## Spatial Scale of BC Trend.

We will determine the spatial scale of the observed Roxbury trend (question #3) using multiple approaches. First, since the HSPH site is less influenced by mid to micro-scale local traffic sources than Roxbury, if a trend of similar magnitude is seen, then the trend is likely to be similar across the Boston core urban area (e.g., within a few km of downtown). Because the HSPH BC data play an important role in understanding the spatial scale of these trends, we need to better understand the effective spatial scale of that site. To accomplish this, NESCAUM will install and operate a MA-DEP Aethalometer near the existing HSPH site (150 meters to the east), but near the major local roadway and about 15 feet above the ground. This siting is consistent with the siting for the other BC sites used in this data analysis. This site will run for approximately 6 months, to capture both cold and warm weather patterns. Data from this site will be compared to both the existing HSPH site and the Roxbury site. Instrument collocations with both existing sites will be performed by NESCAUM to control for inter-instrument bias.

Second, the N.End BC trend is critical to understanding the spatial scale of the BC trend observed at Roxbury. We will estimate missing N.End BC data for 2001 to mid-2003 by analyzing PM2.5 FRM filters from that site by optical reflectance, a surrogate of BC. For this period, every sixth-day samples will be analyzed to generate annual means. To calibrate reflectance measurements to BC, twelfth day samples will be analyzed from mid-2003 through 2004 and compared to the measured BC for the same days. A total of 200 filters will be analyzed at HSPH, where these measurements are done routinely.

### Likely role of control programs in observed BC trends.

In attempting to answer question #4 (to what extent can we determine which control measures contributed to the decline in BC?), we will investigate and document various changes that have occurred near both MA-DEP monitoring sites during this time period. The Roxbury site is near a major bus station (Dudley square) and a bus yard for MBTA commuter busses (the Bartlett yard was closed in 2004 and was cited for idling violations in 2002). All MBTA and Boston school busses have been either retrofitted with DPFs or converted to CNG during or around 2004; a detailed time line of all fleet retrofits will be obtained. In the N. End, potential impacts in addition to the change in the Boston bus fleets include the change in local traffic patterns when the last lanes of the expressway were put underground (the Big Dig) in December 2003. This site is near both the Big Dig and a major MBTA bus station (Haymarket). Other possible factors might include enforcement of anti-idling laws, and a decrease in sulfur content of on-road diesel fuel; we will investigate the timing of these factors with respect to observed changes in BC at Roxbury and the other two Boston BC sites.

## Analysis of Spatial Gradients of BC

A preliminary analysis of some of the data from the 2003 BC spatial gradient study and additional background information is at: <http://www.nescaum.org/documents/allen-spatial-bc.pdf/>

Before a rigorous analysis of these data can be performed, the dataset must be corrected for several known artifacts such as between instrument bias and the Aethalometer BC spot matrix effect.

The “spot matrix effect” is a relatively recently recognized problem in the Aethalometer method that introduces both a short term (hourly to daily) variable negative artifact, a strong (factor of two) seasonal bias, and a bias that is a function of the “Maximum Attenuation” setting of the Aethalometer. This artifact can vary from large positive factors to smaller negative factors as a function of the aerosol composition on the instrument’s filter spot. For additional information on this artifact, see:

<http://www.epa.gov/ttn/amtic/files/ambient/2006conference/allenaethalometer.pdf>

An important issue to consider is if the spatial gradient study data collected in 2003 is still representative now. Given the apparent large change observed in BC between 2002 and 2004 at the Roxbury site, it is possible that the 2003 spatial study data no longer properly represent present-day BC gradients. The multi-site trends data analysis described above includes components that should allow assessment of the spatial scale of the decrease in BC observed at Roxbury. If we can show that a roughly proportional decrease occurred at multiple sites and especially the HSPH site (more urban scale siting), then we can have reasonable confidence that although the actual concentrations measured in the 2003 spatial study may have decreased, the overall urban spatial gradients have not changed substantially. Annual BC means for the three long-term sites will be assessed to evaluate the stability of the 2003 spatial analysis.

### Specific Tasks for Spatial Study Data Analysis:

1. Re-process the entire raw 5-minute 2003 Aethalometer data set for all 12 sites with a new version of Jay Turner's "data masher" program ( <http://users.seas.wustl.edu/jrturner/aethalometer/> ). By summer 2007, this program will include corrections that compensate for the "spot matrix effect".
2. Normalize the artifact-corrected BC data to correct for inter-instrument bias. Most of the Aethalometers used in this study were collocated either before or after the study period, but these data have never been evaluated or used to correct study data. In one case, a pair of instruments used in this study was shown to be different by 20%. This is a very significant amount given the objectives of the study; within the urban core, many sites differ by only 20% to 30%.
3. Create a daily mean data set from the corrected hourly data. Analyze the corrected hourly and daily data sets for a wide range of scenarios, including simple annual [or summer] means and diurnal patterns. ANOVA techniques will be used to assess the significance of differences between sites. Correlation matrixes will be created for both hourly and daily BC metrics. Data will be stratified by season and day of week analysis. Event case studies will be analyzed to determine the dynamic behavior and spatial patterns during periods of unusually high urban BC concentrations.
4. Compare the 2003 spot matrix artifact-corrected data for N.End, Roxbury, and HSPH with the original data to assess the extent of bias in the uncorrected data on a monthly basis.
5. Construct a longer BC annual mean record for the N.End site using reflectance analysis on filters.
6. Compare annual means for these three long term sites to assess stability of the 2003 spatial analysis.
7. Collect BC data at a site near the existing HSPH site but close to street level for 6 months to better characterize the spatial scale of the HSPH BC data set. Although this is a data analysis project, this minor monitoring component will substantially enhance the interpretation of the data analysis; since the scope of the monitoring is limited, the added cost is not a substantial portion of the proposed budget.