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 PM2.5 across urban areas:
- Compliance issues (PM2.5 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 $\mu g/m^3$
- 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)

Study design details and winter 2003 pilot study results are available at:
http//64.2.134.196/General/030403pres-blkcarb.pdf

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 PM2.5 of 15.0 µg/m^3. Data from PM2.5 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 PM2.5 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 PM2.5 (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 PM2.5 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; Regional Background site for Metro Boston

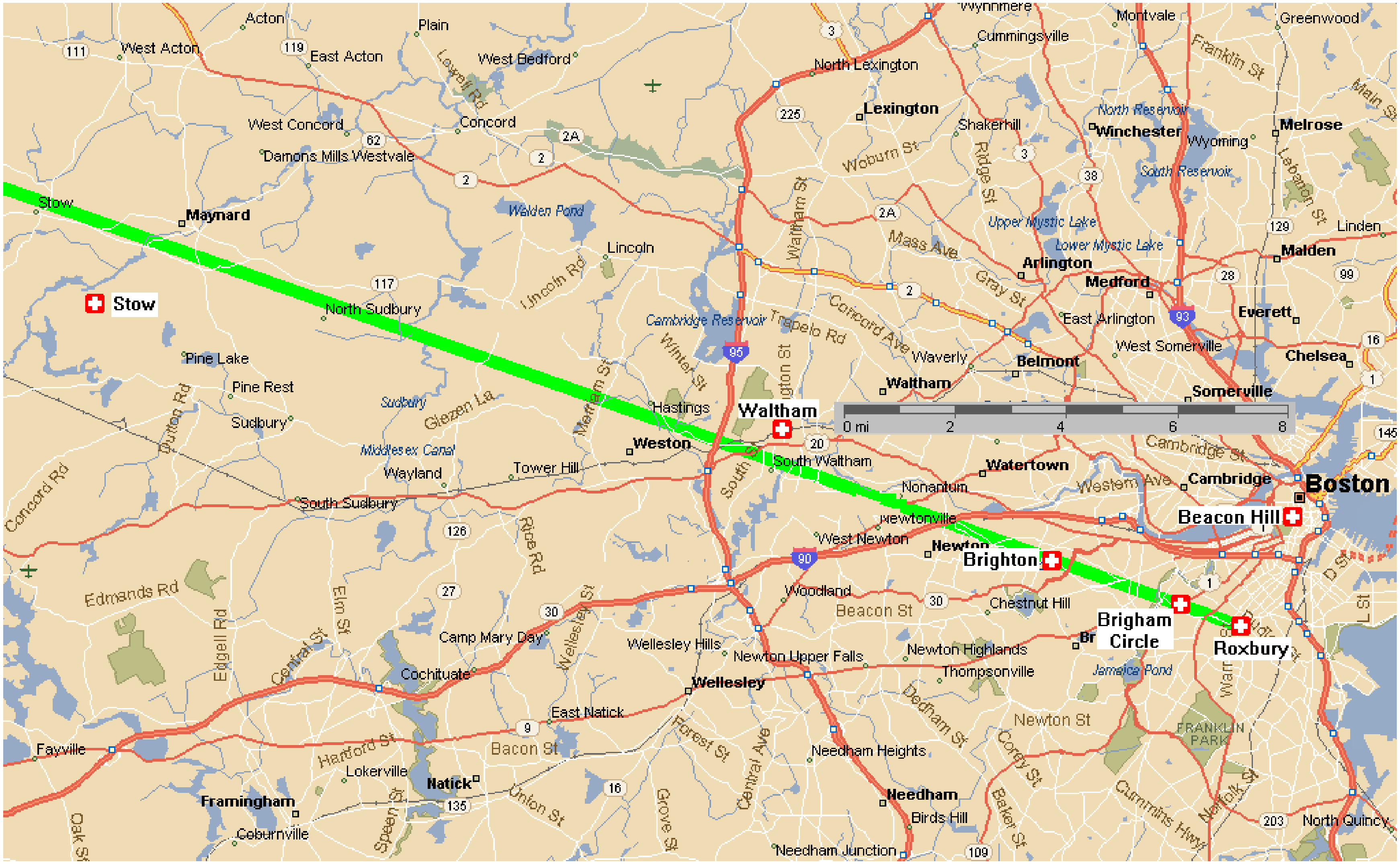


Figure: 6-Site Hourly BC Distributions, Dec. 20, 2002 to Sep. 09, 2003.

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 (95^{th} 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.

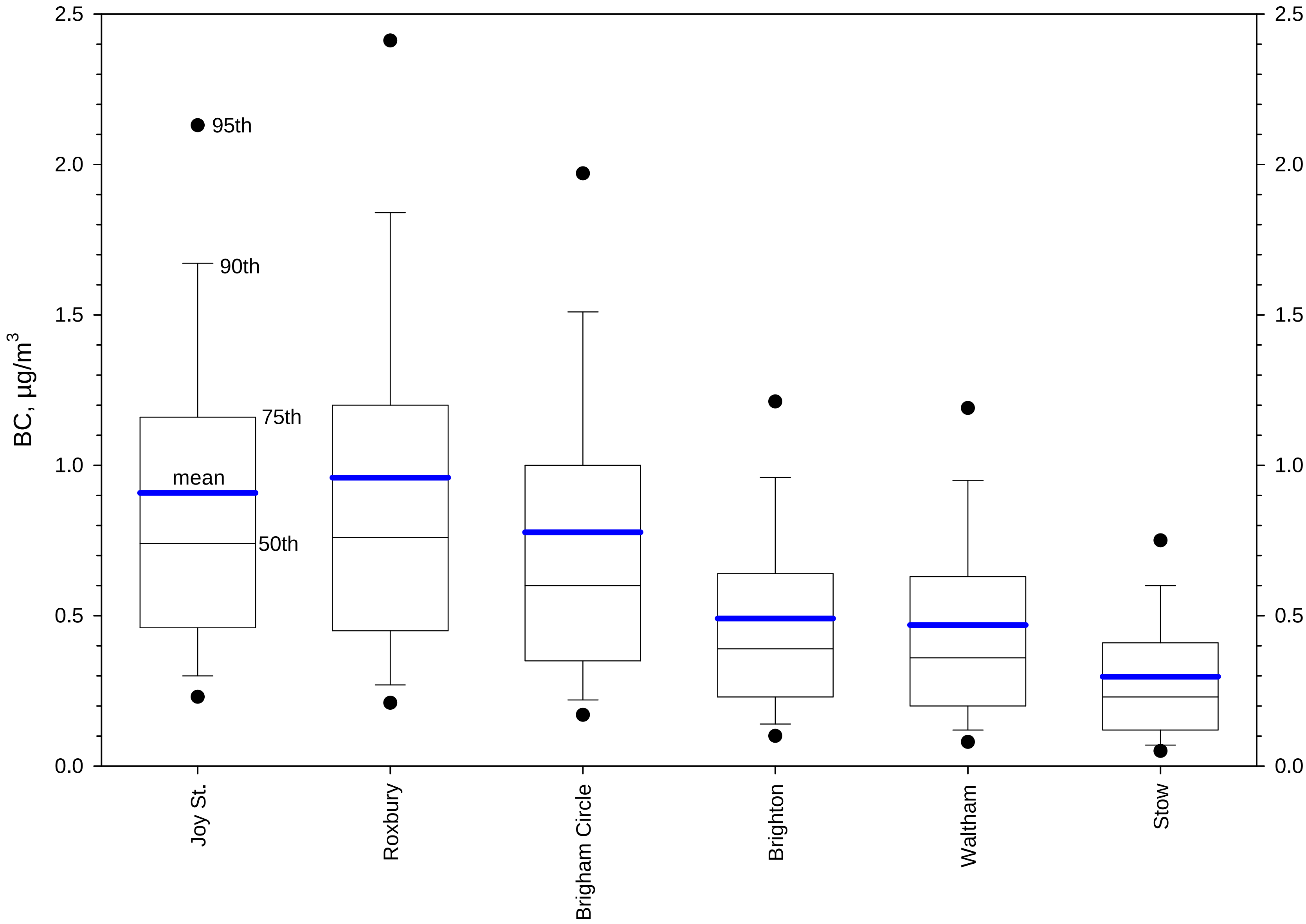
Figure: Workday/Non-Workday BC means across sites.

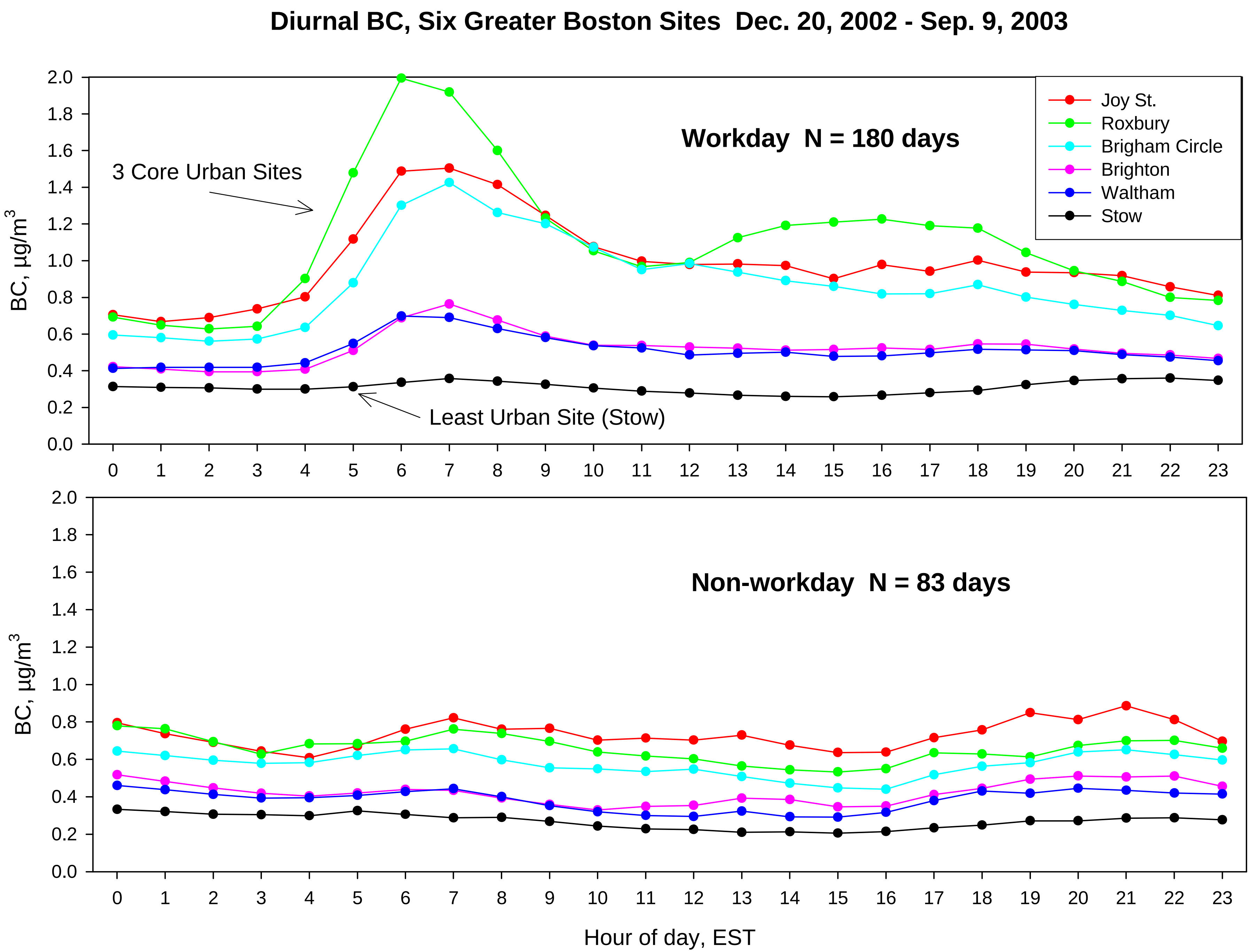
This plot shows the mean BC across sites by work/non-work day. As would be expected, the differences are highest (about 70%) at the sites with the most local traffic influence and highest BC levels, and decrease to about 10% at the Stow background site.

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

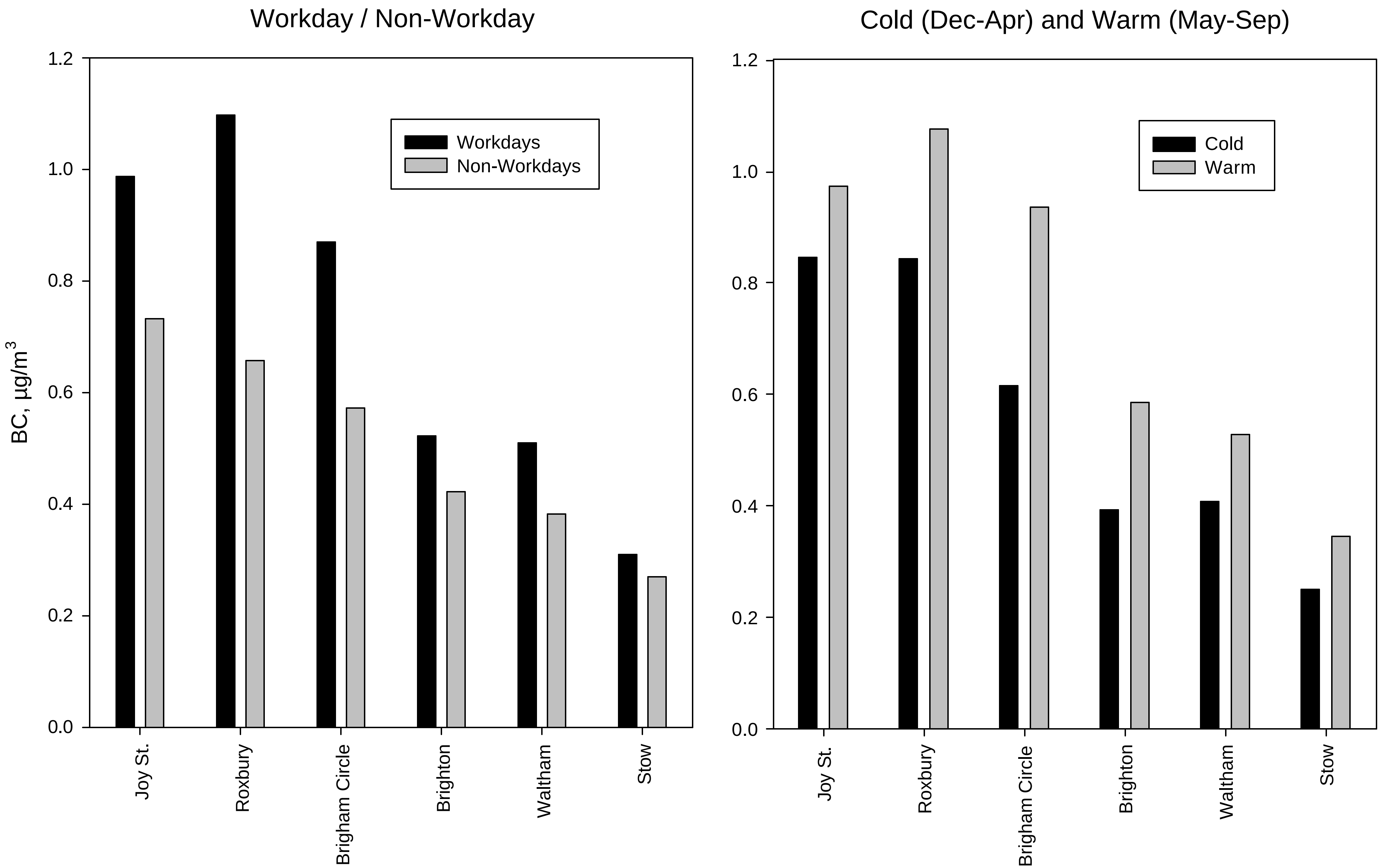
Although winter might be expected to have more of a local mobile source influence than summer (more and stronger inversions), these data show that all sites had substantially lower mean BC in the winter. This is most likely an artifact due to the unusually stormy weather in winter 2003; compared to Jan-Feb 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
Workday N = 180 days
3 Core Urban Sites
Least Urban Site (Stow)
Joy St.
Roxbury
Brigham Circle
Brighton
Waltham
Stow
BC, µg/m3
Non-workday N = 83 days
Hour of day, EST

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



"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).

Site Locations	Km	Site Description
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

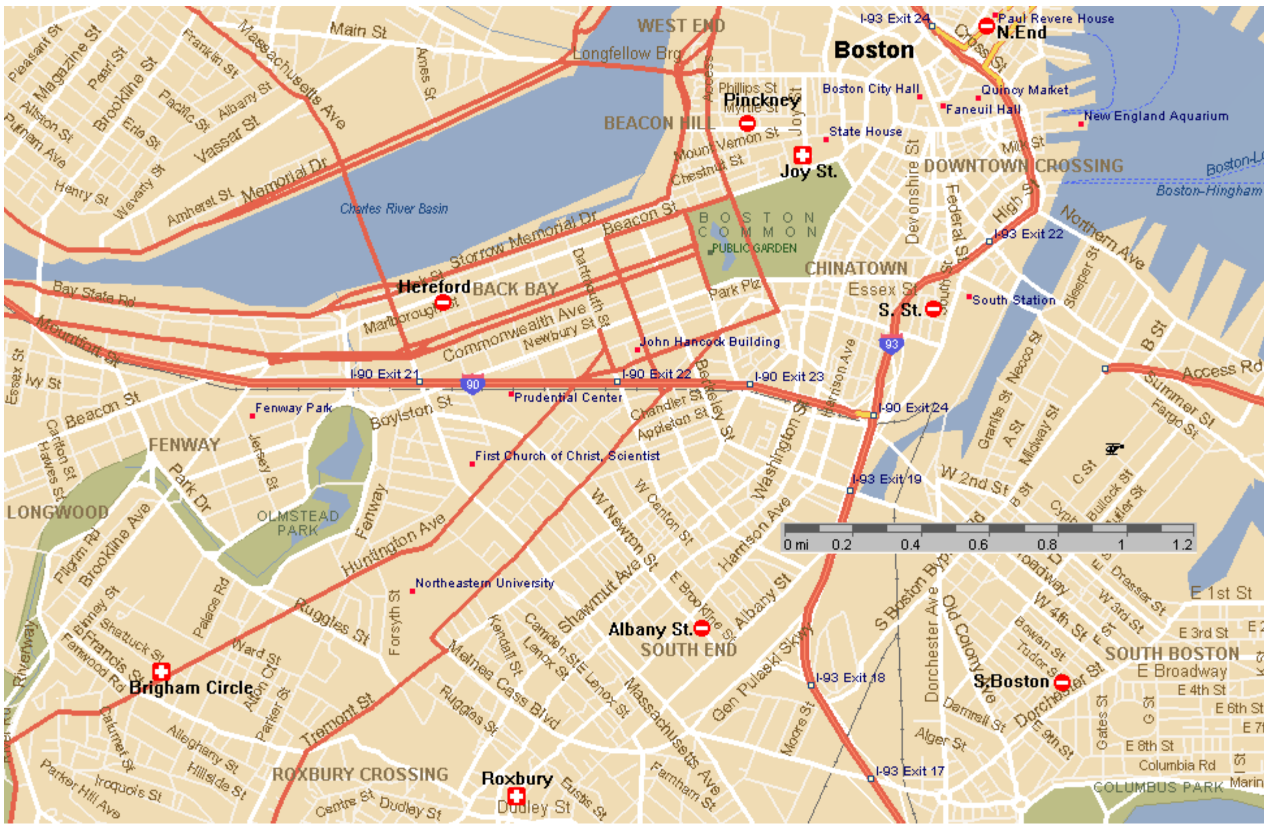


Figure: Summer 2003 1-hour Boston BC frequency distributions

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 $\mu g/m^3$, 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 $\mu g/m^3$, 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.

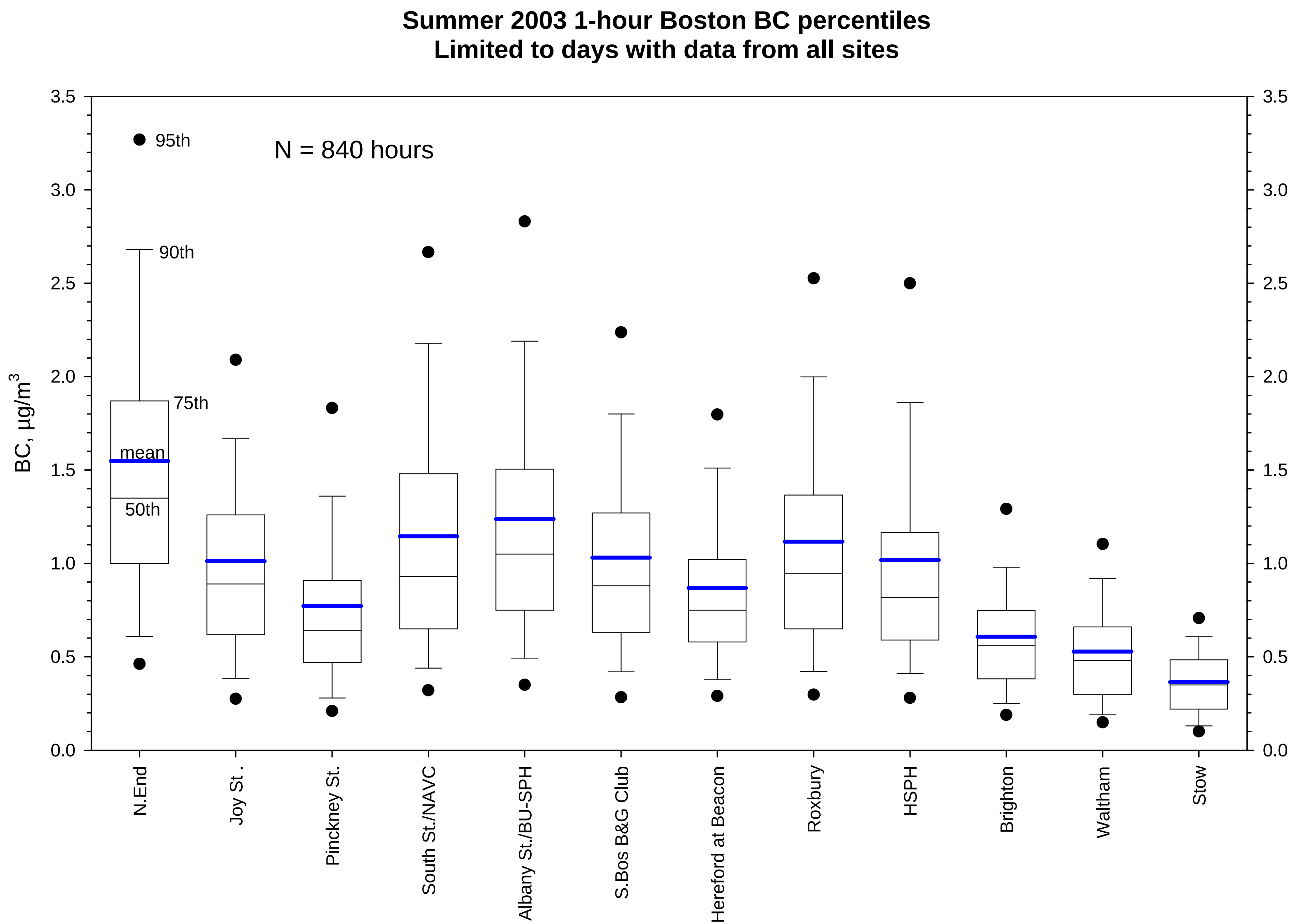
Figure: Joy St., Pinckney St. and Roxbury, April - August 2003

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.

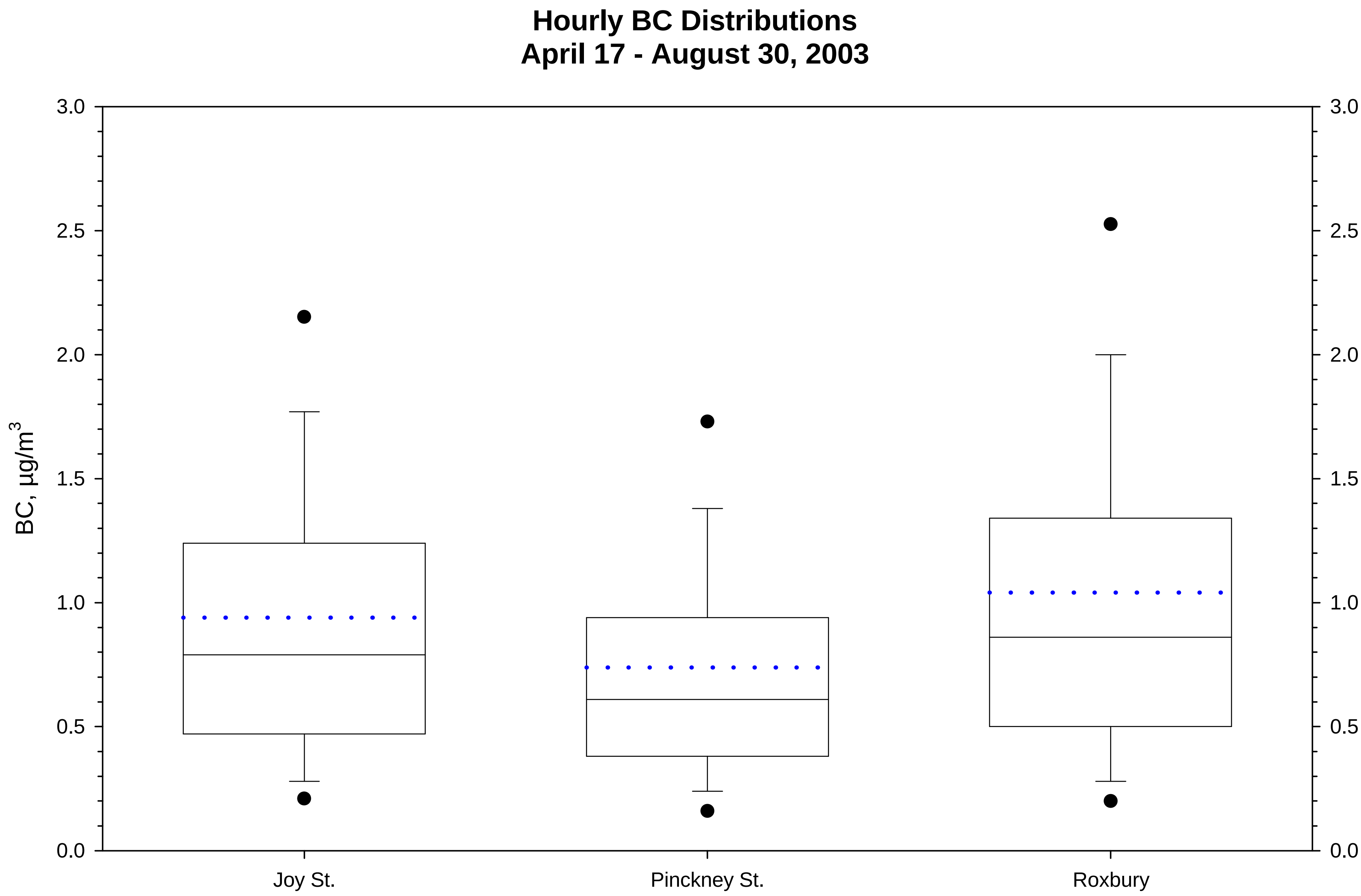
For this longer period, mean BC for Joy, Pinckney, and Roxbury are 0.94, 0.74, and 1.04 $\mu g/m^3$ respectively. The ratio of Pinckney to Joy St. is 0.79 for both the mean and the 95th percentile values. This suggests that part of Joy St.'s BC is traffic that is very local (micro-scale), but gradients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

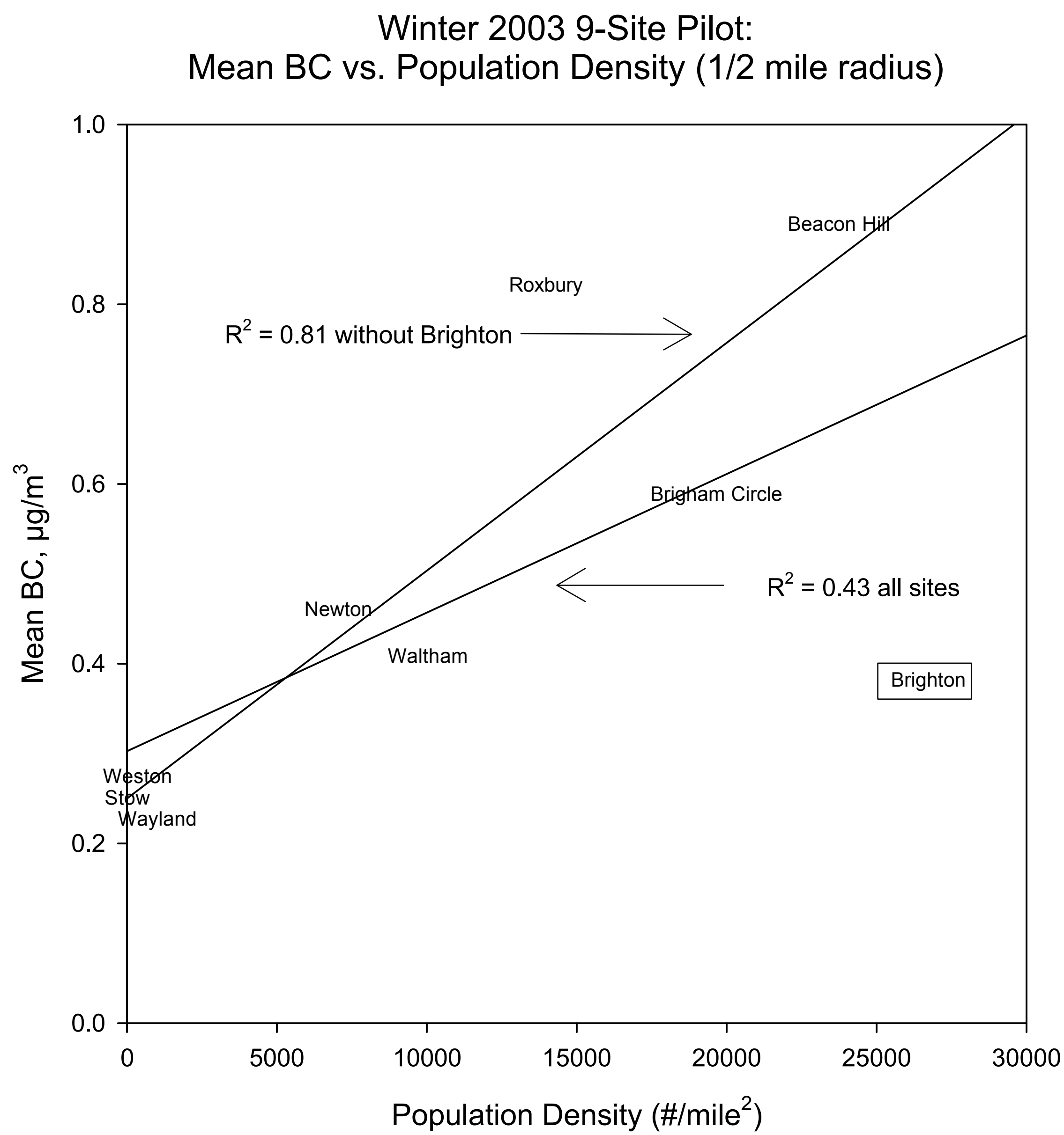
Figures: BC vs. Population Density, Winter 9-site and Summer Boston 10-site Regressions

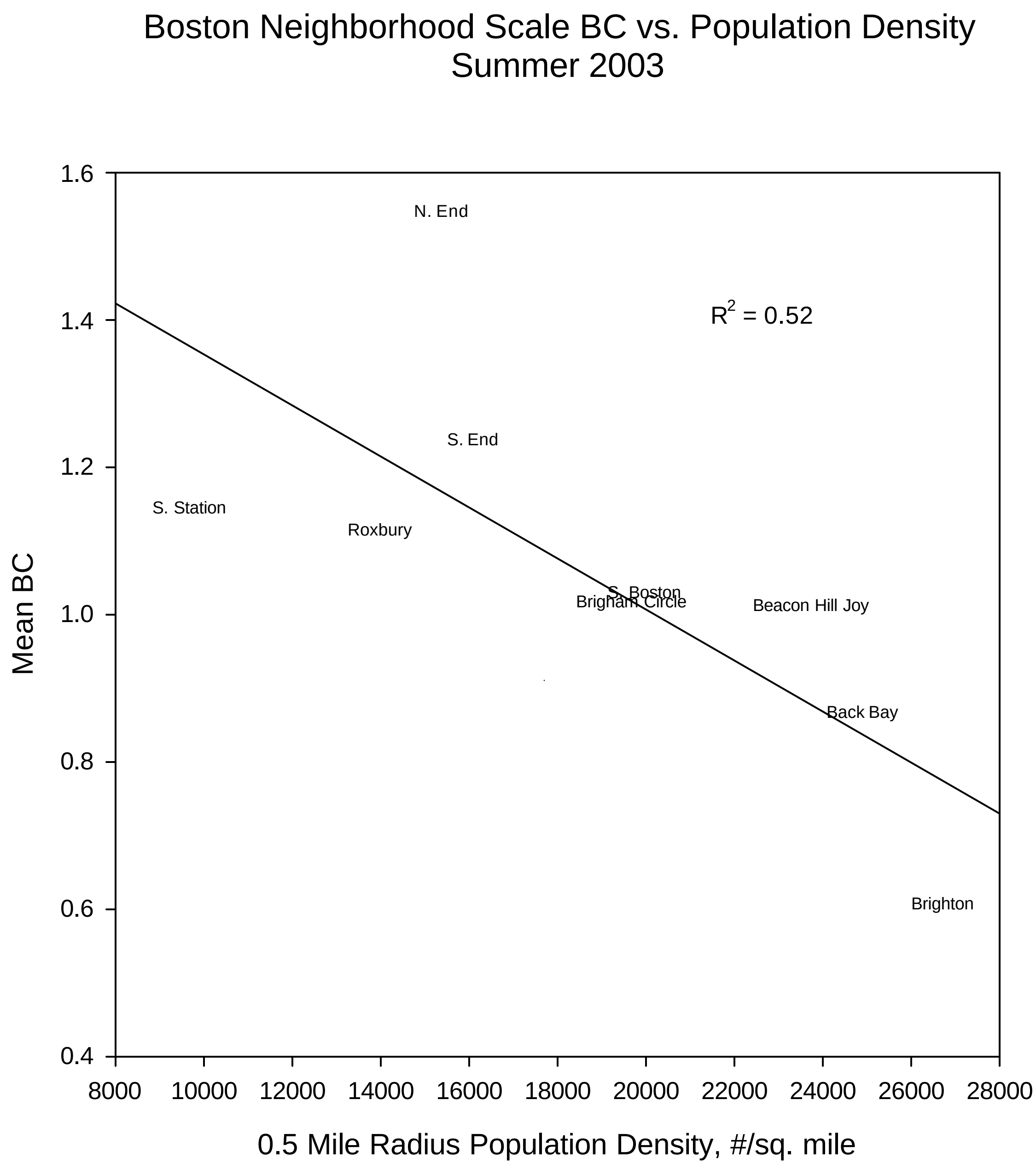
The first plot shows data from the winter 2003 pilot study that suggested that population density might be a useful surrogate for average BC concentrations over a large spatial scale and with a large range of BC concentrations (R^2 of 0.81 if Brighton removed). The same analysis was performed on the 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
N = 840 hours
95th
90th
75th
mean
50th
BC, µg/m3
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
N.End
Joy St .
Pinckney St.
South St./NAVC
Albany St./BU-SPH
S.Bos B&G Club
Hereford at Beacon
Roxbury
HSPH
Brighton
Waltham
Stow


Hourly BC Distributions
April 17 - August 30, 2003
BC, µg/m3
3.0
2.5
2.0
1.5
1.0
0.5
0.0
Joy St.
Pinckney St.
Roxbury


Winter 2003 9-Site Pilot:
Mean BC vs. Population Density (1/2 mile radius)
Mean BC, µg/m3
Population Density (#/mile2)
1.0
0.8
0.6
0.4
0.2
0.0
0
5000
10000
15000
20000
25000
30000
Beacon Hill
Roxbury
R2 = 0.81 without Brighton
Brigham Circle
R2 = 0.43 all sites
Newton
Waltham
Brighton
Weston
Stow
Wayland


Boston Neighborhood Scale BC vs. Population Density
Summer 2003
Mean BC
0.5 Mile Radius Population Density, #/sq. mile
1.6
1.4
1.2
1.0
0.8
0.6
0.4
8000
10000
12000
14000
16000
18000
20000
22000
24000
26000
28000
N. End
R2 = 0.52
S. End
S. Station
Roxbury
S. Boston
Brigham Circle
Beacon Hill Joy
Back Bay
Brighton

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 $\mu g/m^3$ 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 $\mu g/m^3$); without those values the mean for this hour is 6 $\mu g/m^3$, more similar to the other sites.

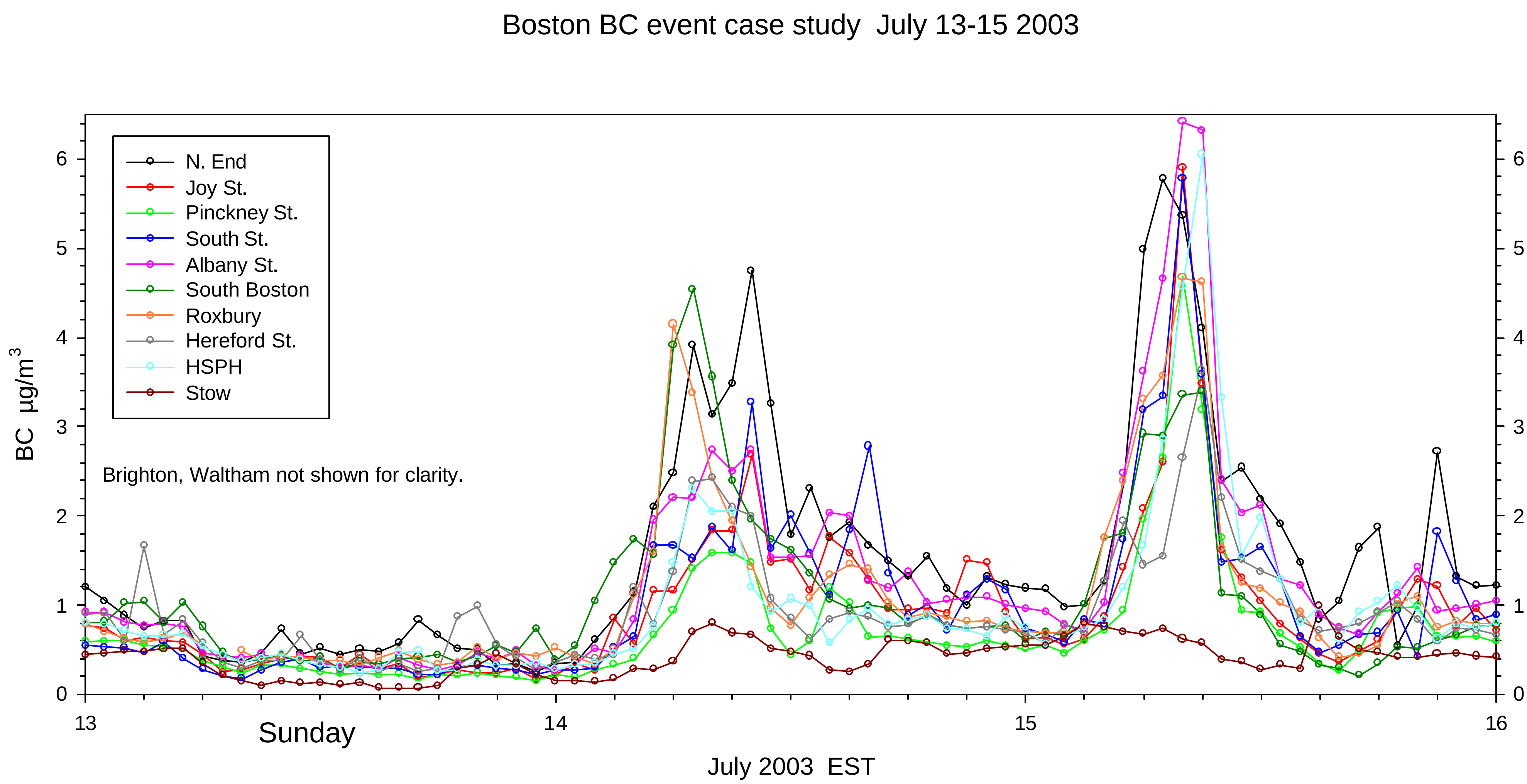
Both of these time-series plots show a very distinct "clean Sunday and dirty work-day" effect.

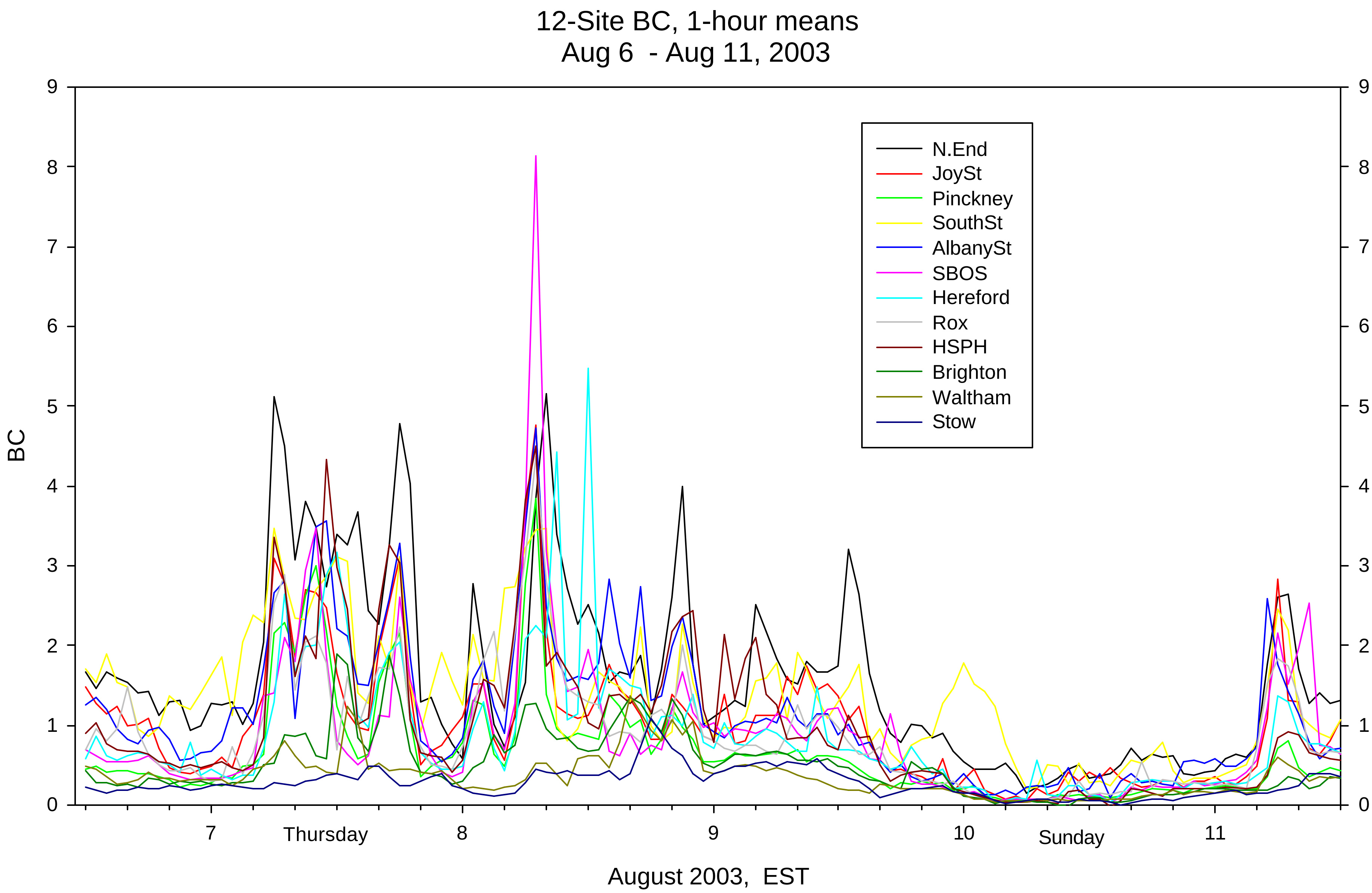
Figure and Table: August 6-11 case study: hourly scatter plots and correlation matrix

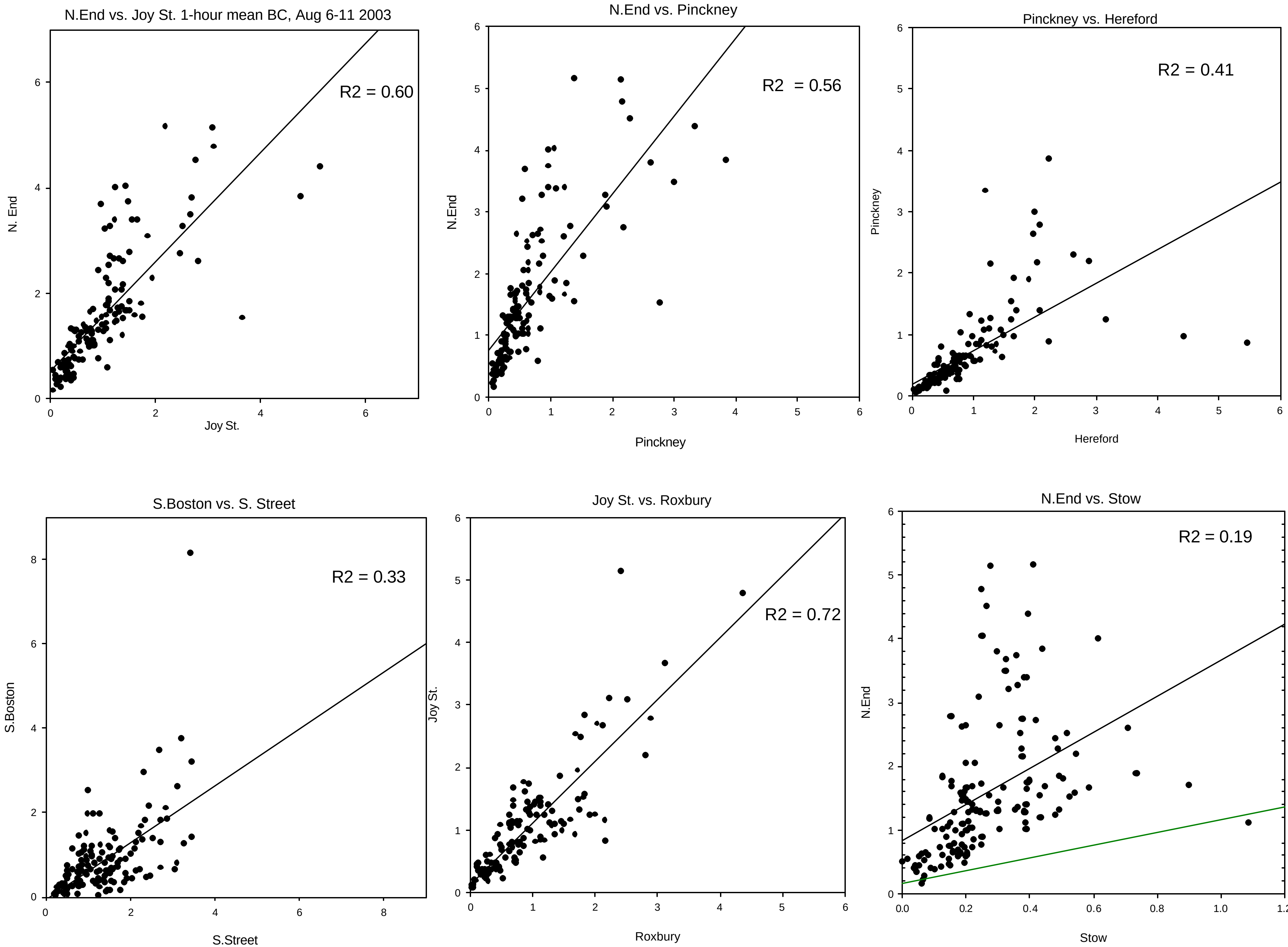
Scatter plots for hourly BC for six site-pairs and a table for all site pairs during this August 5-day period are shown as examples of the short-term relationships across different spatial scales. There is a wide range of correlation, from reasonably high (R^2 = 0.85 for the two Beacon Hill sites) to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

Distance between sites is not always a predictor of good 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 $\mu g/m^3$ 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
N. End
Joy St.
Pinckney St.
South St.
Albany St.
South Boston
Roxbury
Hereford St.
HSPH
Stow
Brighton, Waltham not shown for clarity.
BC μg/m3
0
1
2
3
4
5
6
13
14
15
16
Sunday
July 2003 EST


12-Site BC, 1-hour means
Aug 6 - Aug 11, 2003
BC
0
1
2
3
4
5
6
7
8
9
N.End
JoySt
Pinckney
SouthSt
AlbanySt
SBOS
Hereford
Rox
HSPH
Brighton
Waltham
Stow
7
Thursday
8
9
10
Sunday
11
August 2003, EST


N.End vs. Joy St. 1-hour mean BC, Aug 6-11 2003
R2 = 0.60
N. End
Joy St.
N.End vs. Pinckney
R2 = 0.56
N.End
Pinckney
Pinckney vs. Hereford
R2 = 0.41
Pinckney
Hereford
S.Boston vs. S. Street
R2 = 0.33
S.Boston
S.Street
Joy St. vs. Roxbury
R2 = 0.72
Joy St.
Roxbury
N.End vs. Stow
R2 = 0.19
N.End
Stow

1-hour R^2 Matrix, August 6-11 2003

R2 .70 or higher   R2 .50 to .69

	N.End	Joy St.	Pinck-ney 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	.85	.54	.79	.62	.36	.72	.59	.38	.18	.14
Pinckney	.56	.85	x	.51	.79	.64	.41	.69	.67	.44	.19	.13
S. St.	.54	.54	.51	x	.57	.33	.28	.53	.48	.35	.21	.08
Albany	.63	.79	.79	.57	x	.55	.46	.73	.66	.51	.31	.19
S.Bos	.40	.62	.64	.33	.55	x	.34	.70	.48	.24	.09	.10
Hereford	.46	.36	.41	.28	.46	.34	x	.41	.48	.45	.27	.17
Rox	.59	.72	.69	.53	.73	.70	.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” PM2.5 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 tp 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.

Future Work

Future Data Analysis Plans with HSPH (Schwartz et al.): EPA PM-Center funds at Harvard will allow a detailed spatial analysis of these and other related data that will incorporate several variables:

- Geocode all the addresses with monitoring data;
- Fit a regression predicting values – based on:
  - season, population density, distance from roads, length of road segments, estimated traffic counts, land use data, distance from possible sources such as bus depots, etc.
- Incorporate spatial smoothing (kriging, other?)
- Capture geographic patterns in unexplained measurement variance

Goal: to make a surface of estimated pollution exposure

Incorporate meteorology for "case studies" such as shown here with the South Boston August 8 peak hour.

Perform more detailed seasonal analysis with a full year of data; apply ANOVA analysis to between site gradients.

Compare 2003 seasonal means with 5-year historical data (2 sites) as was done with winter:

- Winter 2003 was distinctly cleaner for BC than previous winters

Contingent on finding funding: Investigate microscale cases (Brigham Circle, Brighton, urban and non-urban "hotspots"); gradients from major roadways, other urban areas, other directions from downtown. Investigate micro-scale (< 100 meters) and mid-scale spatial gradients in non-urban populated settings that are impacted by mobile sources.

Collaborate with Turner et al. (Washington University - St. Louis) on validation and application of the "zone of representation" approach: Use 5-minute BC data to independently estimate middle-scale (0.1 to about 1 km – short duration) vs. larger scale contributions. For details, see St. Louis Supersite 2003 Q1 report at:
http://www.epa.gov/ttn/amtic/files/ambient/super/stl13.pdf
See also: Estimating Middle-, Neighborhood-, and Urban-Scale Contributions to Elemental Carbon in Mexico City with a Rapid Response Aethalometer". Watson and Chow, Nov. 2001, J. Air & Waste Manage. Assoc. 51:1522-1528

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