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Methods to Select Metropolitan Areas of Epidemiologic Interest for Enhanced Air Quality Monitoring

Air Quality Monitoring by the EPA Speciation Trends Network, 2001-2005

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Executive Summary

The U.S. Environmental Protection Agency's current Speciation Trends Network (STN) covers most major U.S. metropolitan areas and a wide range of particulate matter (PM) constituents and gaseous co-pollutants. However, using filter-based methods, most PM constituents are not measured daily and the lack of daily air quality data complicates epidemiologic analyses of the potential adverse health effects of these PM constituents.

Possible criteria for the identification of metropolitan areas with the greatest epidemiologic value for enhanced monitoring are population, mean levels and variation of criteria air pollutants and PM constituents, correlations among these pollutants, and the relationship of these correlations to the coefficient of variation.

Using a review of air quality measurements from 49 STN monitors for 2001-2005 as an illustration of this criteria, we selected metropolitan areas that had the appropriate population size, sufficient $PM_{2.5}$ concentration levels, variability for most pollutants, and appropriate correlations between pollutants.

Once these criteria had been met, the geographical distribution of the selected cities was further examined. Due to an over-representation on Northeastern cities and an under-respresentation of Western cities, the final list was adjusted to include western sites
Thus, as an example the list of candidatemetropolitan areas of greatest epidemiologic interest for enhanced air quality monitoring area,:

Sacramento, CA
San Diego, CA
Atlanta, GA
Baltimore, MD
Boston, MA
Newark, NJ
Cleveland, OH
Pittsburgh, PA
Providence, RI
Salt Lake City, UT
Milwaukee, WI

Using the presented methodology daily monitoring of the widest range of particulate matter (PM) constituents and gaseous co-pollutants at these locations would be of great advantage for future epidemiologic time series studies.

Contents

Contents (continued)

Contents (continued)

Tables

Figures

Figures (continued)

Figures (continued)

Figures (continued)

List of Appendix Tables

Acronyms

Al	Aluminum
Br	Bromine
Ca	Calcium
CBSA	Core Based Statistical Areas
Cl	Chlorine
CO	Carbon Monoxide
Cu	Copper
EC	Elemental carbon
EPA	U.S. Environmental Protection Agency
Fe	Iron
K	Potassium
Mn	Manganese
Na	Sodium
NAAQS	National Ambient Air Quality Standards
Ni	Nickel
NO_2	Nitrogen dioxide
NO_3	Nitrate
O_3	Ozone
OC	Organic carbon
PM	Particulate matter
$PM_{2.5}$	Particulate matter less than 2.5 microns
PM_{10}	Particulate matter less than 10 microns
Se	Selenium
Si	Silicon
SO_2	Sulfur dioxide
SO_4	Sulfate
STN	Speciation Trends Network
Ti	Titanium
Zn	Zinc

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Chapter 1
Introduction

Background

Epidemiologic studies of daily mortality and hospitalization (time series studies) have previously demonstrated adverse human health effects of particulate matter (PM), both in the fine fraction ($PM_{2.5}$) and in the coarse fraction ($PM_{10-2.5}$), and gaseous criteria air pollutants, including carbon monoxide (CO), nitrogen dioxide (NO_2), sulfur dioxide (SO_2), and ozone (O_3) (United States Environmental Protection Agency 2009c). Although the epidemiology has been supported by recent toxicological studies in both humans and animals, the identification of the specific etiologic agents responsible for the observed health effects remains a major uncertainty in the science underlying the current National Ambient Air Quality Standards (NAAQS). Furthermore, the specific constituents of PM vary among the different major air pollution sources with consequences for control strategies (United States Environmental Protection Agency 2009c). The Environmental Protection Agency's (EPA's) Multi-Year Plan for Clean Air included a commitment to the investigation of multipollutant health effects (United States Environmental Protection Agency 2008b). The EPA's current Strategic Research Action Plan for Air, Climate and Energy continues this commitment to the investigation of multipollutant health effects.

The evaluation of the health effects of specific PM constituents has been the focus of many recent epidemiologic and clinical studies. Using single pollutant models, several studies have observed associations between various PM constituents and mortality (Mar et al. 2000; Burnett et al. 2000; Ostro et al. 2007). An alternative to the constituent-by-constituent approach is the consideration of major sources as characterized by multiple constituents. For example, previous time-series analyses indicate that, of the sources of PM, motor vehicle exhaust usually has the strongest associations with all non-accidental or cardiovascular mortality (Mar et al. 2000; Laden et al. 2000; Janssen et al. 2002).

Time series studies have also shown that certain chemical species significantly modify the association between $PM_{2.5}$ and mortality (Franklin and Schwartz 2008), while others have examined the role of gaseous pollutants such as O_3 as either potential confounders or surrogates to $PM_{2.5}$ exposures (Sarnat et al. 2001). The degree of spatial and temporal variability can differ by pollutant, having implications for epidemiologic research. Heterogeneity in city-specific and seasonal-specific estimates have been demonstrated in PM_{10} (Peng et al. 2005; Dominici et al. 2003), $PM_{2.5}$ (Franklin et al. 2007), and ozone (Bell et al. 2005; Bell et al. 2004; Levy et al. 2005; Ito et al. 2005) as well as with the effects of $PM_{2.5}$ on hospital admissions (Bell et al. 2006).

The various constituents of PM probably exert their adverse effects through different modes of action and over different time scales (National Research Council 2004). Long-term exposures to spatial gradients, primarily driven by regional-scale air pollutants, have been associated with decreased survival in well-defined cohorts (Puett et al. 2009; Laden et al. 2006) and decreases in city-specific life expectancy (Pope et al. 2009). Short-term exposures to temporal gradients, driven by more local-scale air pollutants, have been associated with increased hospitalization and mortality both at very prompt effects (Zanobetti and Schwartz 2009; Bell et al. 2006; Franklin et al. 2008) and over more extended periods (Zanobetti et al. 2003). Using biological indicators of early adverse effects, investigators have recently reported very immediate adverse effects on cardiac and endothelial function (Schneider et al. 2008) and delayed inflammatory effects (Mann et al. 2010).

The current air quality monitoring network covers most major U.S. metropolitan areas and a wide range of PM constituents and gaseous co-pollutants. However, using filter-based methods, most PM constituents are measured not daily, but only 1 in 3 days or 1 in 6 days. The lack of daily monitoring of PM constituents complicates time-series analyses (Franklin et al. 2008). At a 2008 EPA workshop, epidemiologists raised the lack of daily monitoring data as a major impediment to future studies of PM constituents and requested enhanced monitoring at selected metropolitan areas (United States Environmental Protection Agency 2008a). Because monitoring resources are constrained, identifying the most important locations for daily monitoring is critical.

This report focuses on ambient air quality monitoring in support of epidemiologic time series studies. Other longitudinal study designs, such as repeated clinical measures in a small closed cohort, have made substantial contributions to our understanding of PM health effects. Some of the design criteria, such as population of the metropolitan area, are simply not relevant to the selection of a study location for a clinical panel, which is generally limited by the clinic location.

Design Criteria

To maximize the research potential, the enhanced monitoring frequency should be conducted in major metropolitan areas with short-term variations in criteria air pollutants in the vicinity of the current NAAQS. An ability to distinguish between various PM constituents would be enhanced by independent variation in these constituents as characterized by low pair-wise correlations. However, low correlations between pairs of constituents may be due to a variety of factors, including a lack of variation of one or both constituents.

The five criteria for the identification of metropolitan areas with the greatest epidemiologic value for enhanced monitoring frequency are:

1. Population of the metropolitan area (Chapter 4);
2. Mean levels of criteria air pollutants (Chapter 5);
3. Variation in levels of criteria air pollutants and particulate matter constituents (Chapter 6);
4. Correlation among criteria air pollutants and particulate matter constituents (Chapters 7 and 8); and
5. Relationship of correlations to the coefficient of variation (Chapter 9).

The population of the metropolitan area largely determines the daily number of clinical events, such as mortality and hospitalizations, and thus the statistical power to detect potential adverse health effects of air pollutants, as reflected in the confidence intervals around their effect estimates. Small cities with relatively few daily events will have more uncertainty for their city-specific effect estimates and less statistical power to detect potential adverse health effects of air pollutants. Metropolitan areas of more than 100,000 people with an average of more than three clinical events per day will have smaller confidence intervals and more power to detect potential adverse effects. Since statistical power increases only as the square root of sample size, further increases in population provide much less improvement in the confidence intervals. Geographically extensive metropolitan areas of more than 1,000,000 people may also enter the summary analysis of effects across multiple areas with extreme influence and may be less well characterized by a single, central-site monitor. Thus, the most populated metropolitan areas are not necessarily the most critical areas for daily speciation monitoring.

Through the standard setting process, EPA has already determined that air pollutant levels in excess of the current NAAQS are hazardous to human health. Further studies at levels far above the NAAQS cannot address issues of uncertainty in the science underlying the current standards. One such uncertainty in the city-to-city variation in $PM_{2.5}$ health effect estimates. Therefore, ambient concentrations in the vicinity of the current standard provide more relevant information. Ideally, the metropolitan areas identified for daily speciation monitoring would experience a range of air pollution levels at or below the current NAAQS.

In the vicinity of the current NAAQS, greater temporal variation in the measured levels of criteria air pollutants enhance the statistical power to detect potential adverse health effects of criteria air pollutants and PM constituents. Metropolitan areas with greater variation will be more informative than areas with little variation in ambient concentrations.

Since recent and future epidemiologic and clinical studies are focused on resolving the relative independent and joint effects of PM constituents and co-pollutants in a multipollutant context, the ability to detect potential adverse health effects will be enhanced by low correlations among the various pollutants. Observational studies cannot resolve the independent effects of highly correlated air pollutants. So, low correlations among criteria air pollutants and PM constituents are an important factor in the identification of metropolitan areas for daily speciation monitoring.

However, a low correlation between a pair of air pollutants (criteria 4) may also indicate a lack of temporal variability in one or both pollutants (criteria 3). So, a final criterion concerns the relationship pair-wise correlations of air pollutants to the temporal variation in each pollutant. The ideal city for daily speciation monitoring would have both low pair-wise correlations and high temporal variation in air pollutants.

This report analyzes existing air quality monitoring data from a selection of major U.S. metropolitan areas for these five major criteria with the goal of identifying a few areas for daily speciation monitoring from these metropolitan areas. This report also examined the spatial and temporal patterns of selected air pollutants as a guide to the interpretation of results from recent and future epidemiologic and clinical studies.

Chapter 2
Methods

Creation of Database

Ambient concentrations of the criteria pollutants are measured at more than 4,000 air quality monitoring stations operated by state, local, and tribal environmental agencies and compiled by the EPA's Aerometric Information Retrieval Service (AIRS) Database (United States Environmental Protection Agency 2009b). Since 2001, the EPA's Speciation Trends Network (STN) has been measuring fine particle components at 174 sites: 54 permanent sites intended to capture long-term trends and 120 additional locations identified as "supplemental speciation."

These STN monitors are a subset of the Chemical Speciation Network (CSN) that also includes State and Local Monitoring Stations (SLAMS). As of January 1, 2011, the National Core (NCore) Multipollutant Monitoring Network became operational consisting of 80 sites; 63 urban sites and 17 rural sites. NCore integrates several advanced measurement systems for particles, pollutant gases and meteorology. Most of the STN monitors (26) described in this report have been included as part of the NCore network design. For an additional 16 STN sites, the additional platform requirements of NCore necessitated a shift of the monitor location within the same county. Only 7 STN monitors were not included in the NCore network.

For this analysis, the focus is on metropolitan areas with STN monitors during 2001–2005. Four monitors that were not operational for the entire 2001–2005 time period were excluded: New Haven County, CT (New Haven); Monroe County, NY (Rochester); Henrico County, VA (Richmond); and Kanawha County, WV (Charleston). Monitors located in Cass County, ND (background site) and Guaynabo County, Puerto Rico (outside the continental United States) were also excluded. Following common practice, the urban speciation site in Washington, DC that is part of the EPA/National Park Service Visibility (IMPROVE) program was included as an STN-equivalent monitor. Thus, 49 STN monitors (or STN-equivalent) were included in this analysis.

The 49 STN monitors selected for this analysis were those that had data from 2001-2005. The purpose of this report is to illustrate the selection criteria. The metropolitan areas chosen for such as future epidemiologic study may differ slightly depending on the STN monitoring area analyzed.

All of the STN monitors were collocated with measurements of total $PM_{2.5}$ mass. Where available, measurements of PM_{10} and the gaseous co-pollutants were obtained from collocated monitors, otherwise the next nearest non-STN monitor was used (Table 2-1). For comparability with the 24-hour filter-based measurements of PM constituents, hourly measurements of gaseous pollutants (CO, NO_2, O_3, and SO_2) were combined into 24-hour averages.

Meteorologic data were generally obtained from National Weather Service stations, except in two cases where a U.S. Air Force weather station was much closer than the nearest National Weather Service Station. Data from monitors located more than 20 miles (32 kilometers) from the STN monitors were excluded from this analysis. Site identifiers for the monitors used in this report are provided in Appendix A, Table A-1.

The precise locations of these selected STN monitors were verified using aerial photography and are available as a keyhole markup language (kml) file (United States Environmental Protection Agency 2010).

The definitions for the 49 multi-county metropolitan areas in this report are based on the Core-Based Statistical Areas (CBSA) of the White House Office of Management and Budget (Appendix A). While the division of metropolitan areas across multiple county-level jurisdictions is largely unrelated to air quality considerations, the segregation of the population into urban core and suburban ring for some metropolitan areas may introduce an air quality relationship. Sacrificing precision to readability, CBSAs in this report will be referenced by the name of the largest city in the CBSA (shortened name underlined in Appendix A, Table A-2).

Mortality data for 2001 through 2005 at the county level was obtained from the National Center for Health Statistics under a restricted data use agreement.

Table 2-1. Distance in Miles from the STN Monitor to the Nearest PM_{10} and Gaseous Monitor and to the Nearest Weather Station: 49 STN Monitors, 2001–2005

City	CO	NO_2	O_3	SO_2	PM_{10}	Weather
Birmingham, AL	1	>100[b]	3	7	-[a]	3
Phoenix, AZ	-	-	-	-	-	7
Bakersfield, CA	-	-	-	82[b]	-	5
Fresno, CA	-	-	-	>100[b]	-	3
Oxnard, CA	-	-	-	22[b]	-	8
Riverside, CA	-	-	-	-	-	4
Sacramento, CA	-	-	-	-	-	7
San Diego, CA	13	-	-	13	-	3
San Jose, CA	-	-	-	28[b]	-	2
Denver, CO	1	1	1	1	-	6
Washington, DC	2	-	-	4	4	5
Miami, FL	<1	<1	5	13	-	5
Tampa, FL	6	15	15	9	14	6
Atlanta, GA	-	-	-	8	7	8
Boise City, ID	1	>100[b]	17	>100[b]	1	9
Chicago, IL	7	-	-	-	5	4
Indianapolis, IN	3	3	3	6	3	5
Baton Rouge, LA	-	-	-	-	3	5
Baltimore, MD	-	-	-	-	-	8
Boston, MA	-	-	-	-	-	4
Springfield, MA	6	-	-	6	13	1
Detroit, MI	-	11	-	7	-	7
Minneapolis, MN	2	16	13	2	2	5
Gulfport, MS	68[b]	33[b]	-	31[b]	31[b]	1
Kansas City, MO	-	-	-	-	-	2
St Louis, MO	-	3	-	2	-	10
Missoula, MT	2	>100[b]	>100[b]	>100[b]	-	6
Omaha, NE	4	>100[b]	3	4	-	5
Reno, NV	-	-	-	>100[b]	-	3
Rockingham, NH	36[b]	-	-	-	-	3
Edison, NJ	-	-	-	-	-	0
Newark, NJ	2	-	5	-	-	3
New York, NY	-	-	-	-	11	3
Charlotte, NC	-	-	-	-	2	9
Cleveland, OH	5	-	5	-	-	10
Tulsa, OK	4	-	-	4	4	5
Portland, OR	6	6	6	>100[b]	-	6
Philadelphia, PA						6
Pittsburgh, PA	5	-	-	3	2	7
Providence, RI	1	4	4	1	-	6
Charleston, SC	<1	6	14	6	4	9
Memphis, TN	4	6	3	8	4	8
Dallas, TX	-	-	-	-	3	3
El Paso, TX	-	-	-	<1	-	5
Houston, TX	-	-	-	-	-	9
Salt Lake City, UT	-	-	-	6	-	6
Burlington, VT	-	-	-	-	-	3
Seattle, WA	-	-	-	-	-	8
Milwaukee, WI	-	-	-	-	3	8

[a] Measurements that were collocated with STN monitor are indicated by a hyphen.

[b] Monitors more than 20 miles (32 kilometers) from the STN monitor were excluded.

Data Analysis

A database of concentrations for 19 $PM_{2.5}$ components, $PM_{2.5}$ mass, PM_{10}, CO, NO_2, O_3, and SO_2 was developed based on data obtained from the EPA's Technology Transfer Network's Air Quality System Data Mart (United States Environmental Protection Agency 2009d). These $PM_{2.5}$ species were chosen based on their potential to be markers for a particular source. SO_4, SO_2, and Se can be markers for coal combustion and Mn, Ni, and V can be markers of residual oil fly ash (Kim and Hopke 2008). Biomass combustion can consist of K, Br, Ca (Watson et al. 2001; Maykut et al. 2003). Mobile sources can be identified by concentrations of Cu, Zn, NO_3, NH_4, EC, and OC as well as CO and NO_2 (Maykut et al. 2003; Lough et al. 2005). Finally, Al, Fe, Si, and Ti are considered crustal elements, and Na and Cl can be markers for sea spray and road salt (United States Environmental Protection Agency 2009b).

The mean and variability of the pollutants were calculated for each site during the 2001–2005 time period and seasonally. The variability was represented by the coefficient of variation, a dimensionless number that is a normalized measure of dispersion, defined as the ratio of the standard deviation to the mean pollutant concentration. The overall and seasonal spearman correlations between selected pollutants were also calculated.

Seasons were based on 3-month periods, with December through February as winter season, March through May as spring, June through August as summer, and September through November as fall.

For the site selection, epidemiologists are interested in identifying cities with low bi-pollutant correlations for each of the two pollutants. However, the low correlations may be due to a low coefficient of variation for one of the pollutants. Therefore interest was in cities that had low correlations between pollutants that still had high coefficients of variation for those pollutants. To examine these three dimensions, bubble plots were created to plot two pollutant coefficients of variations against one another with the areas of those circles proportional to the correlations. Although informative, the interpretation of these numerous bubble plots was tedious and these plots are not presented in this report.

Interpretation of Figures

For all box plots, the shaded box provides the interquartile range with a solid line indicating the median, whiskers indicating the 10^{th} and 90^{th} percentiles, and dots indicating outliers beyond the tenth and ninetieth percentiles.

For the maps with mean concentrations, the monitor-specific mean concentrations are expressed by filled bars representing the proportion of a fixed reference concentration. Aside from selenium where a single city had an extreme mean concentration (Pittsburgh, PA at 6.7 ng/m^3), the reference concentration was fixed at an integer value slightly greater than the highest city-specific concentration.

For maps with correlations, the monitor-specific Pearson correlations (0-1) are expressed by filled bars with a completely filled bar representing a perfect correlation. Aside from winter ozone correlations with $PM_{2.5}$ that were always negative for all STN monitors, all correlations with $PM_{2.5}$ or inter-correlations among PM constituents were either positive or small, that is less than $R^2 = 0.1$.

For maps with coefficients of determination, the monitor-specific coefficients (0-1) are expressed by filled bars representing the proportion of the variation in one pollutant explained by the variation in the other pollutant.

Limitations

Limitations of these data and analyses include measurement error and detection limits. In general sampling methods were improved after 2007 so future work may include applying these criteria to data collected after 2007. In addition sulfate, followed by crustal materials, has the smallest uncertainty associated with its measurement among the components, while uncertainties for organic carbon, elemental carbon, and nitrate are larger (United States Environmental Protection Agency 1996). The organic carbon data provided by the EPA are not blank-corrected and thus have a positive bias due to sampling artifacts. Blank concentrations were not available until 2003 so the organic carbon concentrations presented in this analysis are not blank-corrected. The EPA did not include a method detection limit (MDL) with each $PM_{2.5}$ species concentration recorded until July 2003. Therefore, all reported concentrations were included. Additionally, in order to interpret these results, each pollutant was assigned to only one source category ignoring the fact that a pollutant could be generated from multiple sources. For example, iron could reflect crustal components of road dust re-suspended by vehicles, or particles generated during combustion in engines (Gotschi et al. 2005). Silicon (road dust) and bromine can also represent traffic emissions (Martuzevicius et al. 2004). To examine the potential overlap, the correlation within source categories was studied and the finding indicates that they differed depending on the city.

Chapter 3
Existing Speciation Trends Network (STN) Monitors, 2001–2005

Location of Current Monitors

The 49 selected Speciation Trends Network (STN) monitors are most densely located in the northeastern metropolitan corridor from Washington, DC through Rockingham, NH and in California from San Diego through San Jose (Figure 3-1). In four cities (San Jose, CA; Tampa, FL; Missoula, MT; and Portland, OR), the location of the STN monitors were shifted slightly sometime within the 2001−2005 time period.

Number of Monitoring Days

Not all air pollutants are measured daily so the amount of information on air quality varies by pollutant and location. By design, the filter-based samples for $PM_{2.5}$ constituents at most STN sites were collected on a schedule of either 1-in-3 day, with 67 percent missing by design, or 1-in-6 day, with 83 percent missing by design (Table 3-1). Completely (100%) missing data for a gaseous pollutant and PM_{10} indicates that the nearest alternative air quality monitor was more than 20 miles away. In addition, 17 STN sites had measurements of O_3 only during the warmer months, when O_3 levels are the highest, resulting in higher percentages of missing values. Finally, the monitoring frequency may have changed during the study period. The purpose of Table 3-1 is to describe the information available for this report and not to thoroughly describe monitoring frequency.

Figure 3-1. Map of selected Speciation Trends Network (STN) monitor sites: 49 STN monitors, 2001–2005.

Table 3-1. Percent of Days with Missing Values: 49 STN Monitors, 2001–2005

	$PM_{2.5}$ Species										
	$PM_{2.5}$	SO_4	NO_3	EC	OC	Other	PM_{10}	CO	NO_2	SO_2	O_3
Birmingham, AL	2	67	67	68	68	67	0	3	100	2	3
Phoenix, AZ	55	66	66	67	69	67	53	0	1	4	0
Bakersfield, CA	14	75	75	75	75	75	65	3	2	100	2
Fresno, CA	9	72	72	69	69	69	82	0	1	100	0
Oxnard, CA	69	87	87	87	87	87	83	1	2	100	1
Riverside, CA	8	71	71	71	71	71	68	1	2	3	2
Sacramento, CA	49	70	70	70	70	69	7	9	20	2	2
San Diego, CA	11	76	76	76	76	76	84	0	1	0	0
San Jose, CA	46	76	76	76	76	76	85	12	10	100	10
Denver, CO	65	74	74	74	74	74	3	2	11	1	1
Washington, DC	5	75	75	75	75	75	87	0	0	2	0
Miami, FL	11	74	74	74	74	74	84	2	5	9	3
Tampa, FL	5	68	68	68	68	68	20	1	2	1	0
Atlanta, GA	11	69	69	69	70	69	84	11	6	1	35
Boise City, ID	67	73	73	73	73	73	5	1	100	100	47
Chicago, IL	69	22	17	19	19	73	84	0	20	60	60
Indianapolis, IN	26	73	73	73	73	73	84	1	8	18	50
Baton Rouge, LA	2	79	79	78	78	79	84	1	0	1	0
Baltimore, MD	7	76	76	76	76	76	53	0	2	1	17
Boston, MA	41	73	73	73	73	73	90	8	4	3	2
Springfield, MA	40	80	80	77	77	81	84	1	4	0	16
Detroit, MI	11	69	69	70	70	69	84	3	9	7	51
Minneapolis, MN	46	73	73	73	73	73	90	1	1	2	27
Gulfport, MS	66	77	77	77	77	72	100	100	100	100	37
Kansas City, MO	68	76	76	77	77	77	90	5	3	3	0
St. Louis, MO	2	68	68	68	68	68	92	0	0	0	41
Missoula, MT	69	69	69	69	69	69	10	53	100	100	100
Omaha, NE	10	80	80	78	78	78	83	1	100	7	44
Reno, NV	66	70	70	70	70	70	84	0	20	100	0
Rockingham, NH	71	72	72	73	73	73	91	100	49	48	73
Edison, NJ	71	73	73	73	89	73	85	1	1	0	1
Newark, NJ	11	76	76	76	76	75	94	80	1	2	80
New York, NY	67	68	68	68	68	68	88	1	6	1	1
Charlotte, NC	8	73	73	73	73	73	85	2	2	2	42
Cleveland, OH	68	72	72	73	73	73	9	2	1	0	43
Tulsa, OK	36	70	70	70	70	70	84	2	16	1	1
Portland, OR	40	69	69	67	67	67	74	50	60	100	62
Philadelphia, PA	13	71	71	70	70	70	86	1	5	4	1
Pittsburgh, PA	4	71	71	72	73	72	4	5	0	1	1
Providence, RI	9	86	86	86	86	86	85	1	3	1	51
Charleston, SC	9	69	69	69	69	69	70	4	6	2	2
Memphis, TN	7	73	73	74	74	73	84	3	23	0	33
Dallas, TX	6	70	70	70	70	70	84	3	2	1	0
El Paso, TX	56	71	71	70	70	71	99	1	5	81	1
Houston, TX	4	71	71	71	84	71	85	2	5	3	2
Salt Lake City, UT	3	68	68	68	68	68	8	0	0	2	58
Burlington, VT	67	69	69	69	69	69	94	2	19	1	1
Seattle, WA	3	27	22	11	19	67	91	1	8	3	62
Milwaukee, WI	52	68	68	68	68	68	88	0	28	21	30

Chapter 4
Metropolitan Characteristics

Most previous observational studies of mortality and hospitalization have examined daily counts of health events aggregated across single or multiple counties. The statistical power of such studies is determined, in part, by the average number of daily events, and thus is proportional to the population of the study area. A study area with a larger population will have greater statistical power and a more precise effect estimate.

For time-series models, the analytic choice between an entire multi-county metropolitan area and the single county with the Speciation Trends Network (STN) monitor involves several competing factors. Restricting the analysis to the STN-monitored county may improve the assessment of population exposure by a single, central-site air quality monitor, but the restriction may dramatically reduce the average number of daily deaths, decrease the precision of the subsequent point estimate and alter the relative weight of that metropolitan area in subsequent pooled analyses.

Small numbers of daily deaths will have a major statistical impact on any time-series analysis. The daily counts of non-accidental deaths are expected to follow a Poisson distribution with the probabilities of discrete event counts (N) as a function of the average daily number of deaths (Figure 4-1). For an average daily deaths of three or more deaths per day, 80 percent or more of the days would be expected to have two or more deaths. Conversely, for a mean of one or less death per day, at least 75 percent of the days would be expected to have a count of one or zero.

Since time-series models evaluate the impact of varying air quality on the day-to-day variation in mortality, the relatively modest contribution of air pollution to risk may be difficult to determine in time-series with a high proportion of days with one or zero events. While no decision rule can be formulated regarding a minimum average number of events per day, the fitting of age-specific temporal smoothing functions may be very problematic for metropolitan areas with low populations and average deaths per day, especially for specific causes of death.

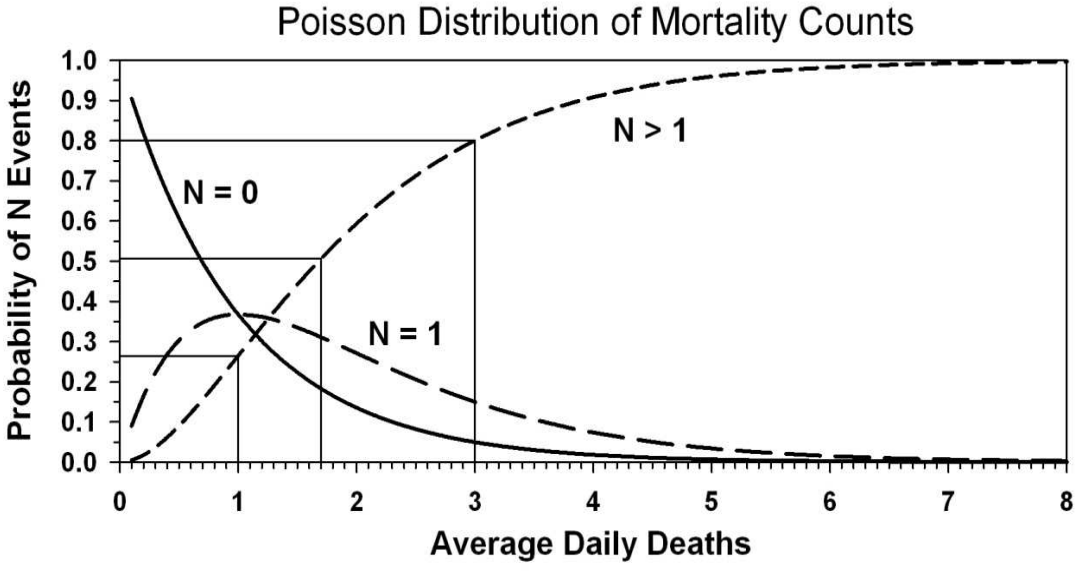

Figure 4-1. Probabilities of specified event counts (N) as a function of average daily deaths for Poisson distributed counts.

Population

The population in 2000 (U.S. Census Bureau 2010) varied greatly across these 49 metropolitan areas (Figure 4-2, Table 4-1). The two most populous Core Based Statistical Areas (CBSAs), New York City, NY (11,298,122) and Chicago, IL (7,628,496), were not included in the figure so that the lower portion of the distribution would be more legible. While most of the CBSAs with STN monitors had populations of more than 750,000, populations less than 250,000 were present for three metropolitan areas (Gulfport, MS; Missoula, MT; and Burlington, VT) and for the monitored county of one additional metropolitan area (Kansas City, MO-KS).

As expected, the monitored county within the CBSA was often much less populous than the CBSA as a whole. Only seven CBSAs with an STN monitor were comprised of a single county. For most CBSAs, the monitored county comprised less than two-thirds of the CBSA population. The population of the monitored county comprised less than 20 percent of the entire CBSA for five metropolitan areas: Washington, DC (District of Columbia with 15%); Atlanta, GA (Fulton County with 19%); Kansas City, MO-KS (Wyandotte County with 9%); St. Louis, MO-IL (St. Louis City with 13%); and New York, NY (Bronx Borough with 12%).

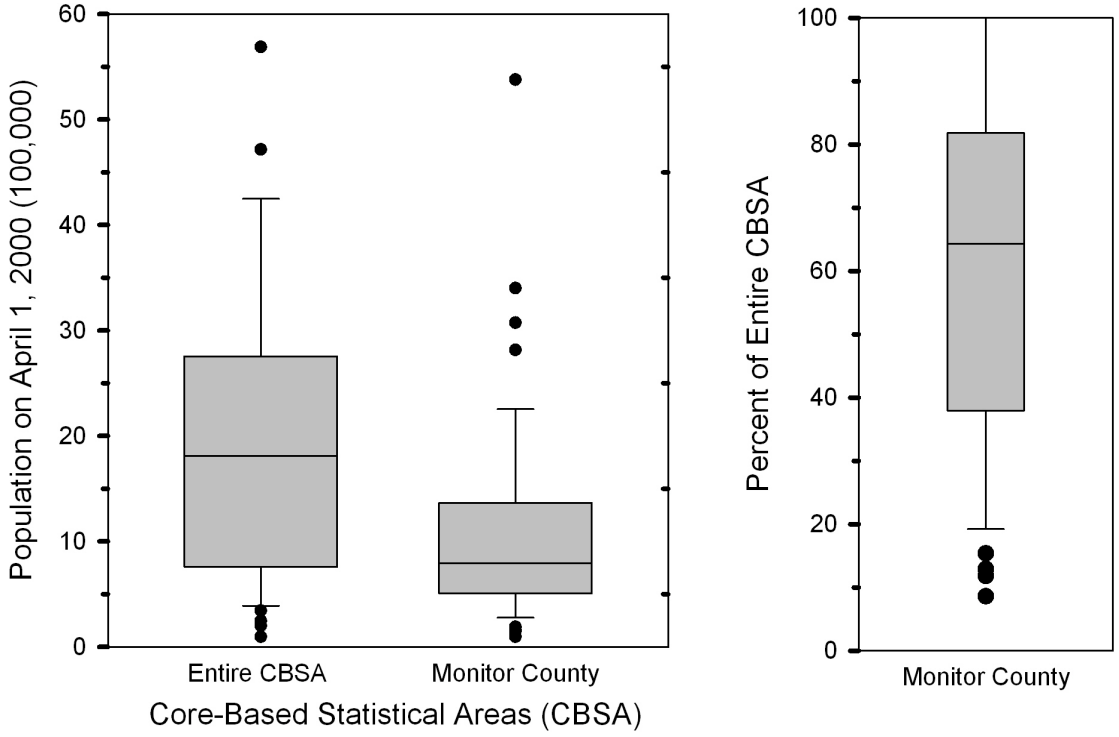

Figure 4-2. Population on April 1, 2000 of Core Based Statistical Areas (CBSA) with an STN monitor and the percentage of the CBSA population in the monitor county: 49 STN monitors, 2001–2005 (presented data excludes New York City and Chicago).

Table 4-1. Population on April 1, 2000 of Core Based Statistical Areas (CBSA) with an STN Monitor: 49 STN Monitors, 2001–2005

CBSA	2000 Population	Monitor County	2000 Population
Birmingham-Hoover, AL	1,051,300	Jefferson, AL	662,062
Phoenix-Mesa-Scottsdale, AZ	3,251,888	Maricopa, AZ	3,072,168
Bakersfield, CA	661,649	Kern, CA	661,649
Fresno-Madera, CA	921,930	Fresno, CA	798,821
Oxnard-Thousand Oaks-Ventura, CA	753,186	Ventura, CA	753,186
Riverside-San Bernardino-Ontario, CA	3,254,817	Riverside, CA	1,545,374
Sacramento-Arden-Arcade-Roseville, CA	1,796,852	Sacramento, CA	1,223,497
San Diego-Carlsbad-San Marcos, CA	2,813,834	San Diego, CA	2,813,834
San Jose-Sunnyvale-Santa Clara, CA	1,735,818	Santa Clara, CA	1,682,584
Denver-Aurora, CO	2,179,343	Denver, CO	553,691
Washington-Arlington-Alexandria, DC-VA-MD-WV	3,727,452	District of Columbia	572,055
Miami-Miami Beach-Kendall, FL	2,253,786	Miami-Dade, DL	2,253,786
Tampa-St. Petersburg-Clearwater, FL	2,396,014	Hillsborough, FL	998,943
Atlanta-Sandy Springs-Marietta, GA	4,248,021	Fulton, GA	815,827
Boise City-Boise City, ID	464,842	Ada, ID	300,906
Chicago-Naperville-Joliet, IL	7,628,496	Cook, IL	5,376,837
Indianapolis-Carmel, IN	1,525,103	Marion, IN	860,457
Baton Rouge, LA	705,962	East Baton Rouge, LA	412,854
Baltimore-Towson, MD	2,553,022	Baltimore City, MD	651,154
Boston-Quincy, MA - Metropolitan Division	1,812,937	Suffolk, MA	689,809
Springfield, MA	680,016	Hampden, MA	456,226
Detroit-Livonia-Dearborn, MI	2,061,161	Wayne, MI	2,061,161
Minneapolis-St. Paul-Bloomington, MN-WI	2,968,812	Hennepin, MN	1,116,037
Gulfport-Biloxi-Pascagoula, MS	246,197	Harrison, MS	189,606
Kansas City, MO-KS	1,836,425	Wyandotte, KS	157,882
St. Louis, MO-IL	2,698,664	St. Louis City	348,189
Missoula, MT	95,799	Missoula, MT	95,799
Omaha-Council Bluffs, NE-IA	767,144	Douglas, NE	463,585
Reno-Sparks, NV	342,885	Washoe, NV	339,486
Rockingham-Strafford, NH Division	389,595	Rockingham, NH	277,357
Edison-New Brunswick, NJ Division	2,173,876	Middlesex, MD	749,167
Newark-Union, NJ-PA Division	2,097,523	Baltimore, MD	792,313
New York-White Plains-Wayne, NY-NJ Division	11,298,122	Bronx, NY	1,332,652
Charlotte-Gastonia-Concord, NC-SC	1,330,552	Mecklenburg, NC	913,639
Cleveland-Elyria-Mentor, OH	2,148,017	Cuyahoga, OH	1,393,848
Tulsa, OK	859,533	Tulsa, OK	563,302
Portland-Vancouver-Beaverton, OR-WA	1,927,883	Multnomah, OR	660,486
Philadelphia-Camden-Wilmington, PA-NJ-DE-MD	5,687,158	Philadelphia, PA	1,517,542
Pittsburgh, PA	2,431,086	Alleghany, PA	1,281,665
Providence-New Bedford-Fall River, RI-MA	1,582,997	Providence, RI	621,595
Charleston-North Charleston-Summerville, SC	548,974	Charleston, SC	310,099
Memphis, TN-MS-AR	1,205,196	Shelby, TN	897,472
Dallas-Plano-Irving, TX	3,451,204	Dallas, TX	2,218,792
El Paso, TX	679,622	El Paso, TX	679,622
Houston-Sugar Land-Baytown, TX	4,715,417	Harris, TX	3,400,590
Salt Lake City, UT	968,883	Salt Lake, UT	898,412
Burlington-South Burlington, VT	198,892	Chittenden, VT	146,572
Seattle-Tacoma-Bellevue, WA	3,043,897	King, WA	1,737,047
Milwaukee-Wauksha-West Allis, WI	1,490,743	Milwaukee, WI	940,165

Mortality

Across all age groups, the average daily counts for total non-accidental deaths were sufficient (great than 3) for time-series analyses across all of the CBSA with STN monitors (Figure 4-3). Only the CBSAs of Missoula, MT and Burlington, VT have average daily numbers of total non-accidental deaths for all ages of 1.7 and 3.3, respectively. Both of these areas were selected for monitoring to provide measurements of fine particles characteristics far from local sources.

Many CBSAs have low average counts for age and cause subgroups, especially for respiratory deaths and deaths below age 75 years (Figure 4-3, Table 4-2). For cardiovascular mortality, 36 of the CBSAs have averages of more than one death per day in all three age groups. Conversely, for respiratory mortality, only the Chicago, IL and New York, NY CBSAs have averages of more than one death per day in all three age groups. For time-series models of respiratory mortality, the estimation of age-specific temporal functions for the baseline hazard may be problematic for all but the two most populous CBSAs due to the very large proportion of days with only 0 or 1 event.

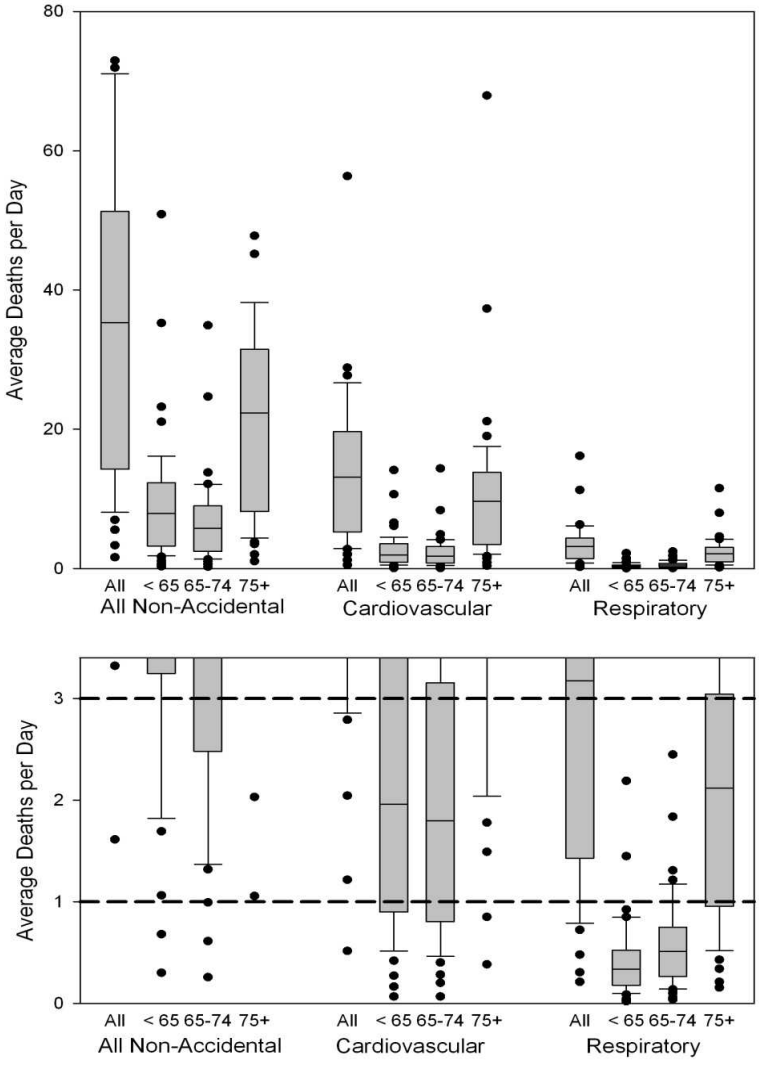

Figure 4-3. Average daily deaths by cause and age groups in CBSA with an STN monitor: 49 STN monitors, 2001–2005 (bottom graph is a blow-up of the lower portion of the top graph).

Table 4-2. Average Daily Deaths by Cause and Age Groups in CBSA with an STN Monitor: 49 STN Monitors, 2001–2005

	All Non-Accidental			Cardiovascular			Respiratory		
CBSA	<65	65-74	75+	<65	65-74	75+	<65	65-74	75+
Birmingham, AL	7.0	5.1	14.8	2.1	1.7	6.3	0.3	0.5	1.5
Phoenix, AZ	14.1	10.8	36.3	3.6	3.5	15.5	0.8	1.2	4.2
Bakersfield, CA	3.1	2.3	6.5	0.9	0.8	3.3	0.2	0.3	0.9
Fresno, CA	4.0	2.8	9.2	1.0	1.0	4.6	0.2	0.3	1.1
Oxnard, CA	2.2	1.7	6.9	0.5	0.6	3.2	0.1	0.2	0.8
Riverside, CA	14.7	11.2	33.9	4.1	4.1	16.8	0.8	1.3	4.2
Sacramento, CA	8.1	6.1	20.7	2.1	2.0	9.6	0.4	0.7	2.7
San Diego, CA	10.4	8.0	31.0	2.5	2.7	14.3	0.4	0.8	3.5
San Jose, CA	4.8	3.6	13.2	1.1	1.1	6.1	0.2	0.3	1.6
Denver, CO	8.4	5.6	19.1	2.0	1.6	7.8	0.5	0.7	2.2
Washington, DC	16.1	9.4	27.5	4.3	3.2	11.8	0.5	0.6	2.5
Miami, FL	10.4	8.0	27.2	3.0	3.1	13.7	0.3	0.5	2.4
Tampa, FL	13.8	12.1	45.2	3.7	4.0	19.0	0.7	1.2	4.1
Atlanta, GA	21.1	12.0	33.2	6.1	4.1	13.8	0.9	1.2	3.3
Boise City, ID	1.7	1.3	5.1	0.4	0.4	2.0	0.1	0.1	0.6
Chicago, IL	35.3	24.7	83.3	10.7	8.4	37.3	1.4	1.8	8.0
Indianapolis, IN	7.6	5.8	17.7	2.1	1.8	7.5	0.5	0.6	2.1
Baton Rouge, LA	4.1	2.5	7.2	1.3	0.9	3.0	0.2	0.2	0.7
Baltimore, MD	14.8	10.0	31.9	4.2	3.3	13.8	0.7	0.8	3.3
Boston, MA	7.1	5.9	22.4	1.7	1.8	8.6	0.3	0.5	2.7
Springfield, MA	3.0	2.4	10.2	0.8	0.8	4.0	0.2	0.2	1.1
Detroit, MI	12.6	7.4	22.3	4.5	3.1	11.3	0.6	0.5	1.9
Minneapolis, MN	9.8	7.3	29.4	2.2	1.8	10.2	0.4	0.7	2.8
Gulfport, MS	2.3	1.9	4.4	0.8	0.7	2.1	0.1	0.2	0.4
Kansas City, MO	8.6	6.6	22.9	2.3	2.1	10.1	0.4	0.7	2.4
St. Louis, MO	13.7	11.2	38.2	4.2	3.9	17.5	0.7	1.0	4.0
Missoula, MT	0.3	0.3	1.1	0.1	0.1	0.4	0.0	0.0	0.2
Omaha, NE	3.3	2.8	8.6	0.9	0.8	3.6	0.2	0.3	1.1
Reno, NV	1.8	1.4	3.8	0.5	0.5	1.8	0.1	0.2	0.5
Rockingham, NH	1.1	1.0	3.5	0.3	0.3	1.5	0.0	0.1	0.3
Edison, NJ	7.7	7.6	30.3	2.0	2.4	13.9	0.3	0.6	2.6
Newark, NJ	9.3	6.4	22.9	2.3	2.1	10.3	0.3	0.4	2.0
New York, NY	49.9	34.9	120.5	14.1	14.4	67.9	2.2	2.4	11.5
Charlotte, NC	6.7	4.6	13.5	1.9	1.5	5.4	0.3	0.5	1.4
Cleveland, OH	10.8	9.2	33.8	3.4	3.3	15.7	0.4	0.6	2.8
Tulsa, OK	4.7	3.6	11.1	1.5	1.3	5.3	0.3	0.4	1.1
Portland, OR-WA	8.0	5.8	22.9	1.8	1.7	9.6	0.4	0.6	2.1
Philadelphia, PA	7.9	7.9	33.7	1.9	2.3	14.7	0.3	0.7	3.5
Pittsburgh, PA	12.0	12.1	47.8	3.6	4.1	21.1	0.5	1.0	4.6
Providence, RI	6.5	5.7	25.0	1.8	1.8	11.3	0.3	0.5	2.7
Charleston, SC	3.1	2.1	5.7	0.9	0.7	2.4	0.1	0.2	0.5
Memphis, TN	8.3	4.9	13.1	2.7	1.9	6.2	0.4	0.4	1.3
Dallas, TX	14.9	8.8	25.4	4.4	3.1	11.2	0.6	0.8	2.6
El Paso, TX	2.6	2.2	5.8	0.6	0.7	2.4	0.1	0.1	0.6
Houston, TX	23.2	13.8	35.9	6.6	4.9	16.2	0.9	1.1	3.2
Salt Lake City, UT	3.2	2.1	7.8	0.7	0.6	2.9	0.2	0.3	0.8
Burlington, VT	0.7	0.6	2.0	0.2	0.2	0.8	0.0	0.1	0.2
Seattle, WA	8.1	5.9	24.0	2.0	1.8	10.1	0.4	0.6	2.5
Milwaukee, WI	6.6	5.3	20.3	1.8	1.7	8.8	0.3	0.4	2.0

Chapter 5
Mean Concentrations of Total PM Mass and Selected $PM_{2.5}$ Species

In order to address issues of scientific uncertainty underlying current standards, observational studies of ambient concentrations far above current standards will be less informative than studies of ambient concentrations at or below current standards. Conversely, observational studies of very low ambient concentrations may lack sufficient statistical power to correctly assess adverse health effects.

Relative to National Ambient Air Quality Standards

The 24-hour average $PM_{2.5}$ concentrations measured by STN monitors varied greatly across the 49 Core Based Statistical Areas (CBSAs) (Figure 5-1). While many CBSAs were well below 17.5 $\mu g/m^3$ (half the 24-hour National Ambient Air Quality Standard (NAAQS) of 35 $\mu g/m^3$) on most days, three CBSAs in Southern California had more than 10 percent of days above 35 $\mu g/m^3$: Bakersfield, Fresno, and Riverside. For the association between air quality and health, these three CBSAs may not be as informative regarding the remaining health effects of $PM_{2.5}$ concentrations at or below the current 24-hour NAAQS since the linear concentration-response relationship may be largely determined by days well above the current standard. Similarly, the nine CBSAs with 10 percent or fewer days above 17.5 $\mu g/m^3$ may also contribute less information to the analysis.

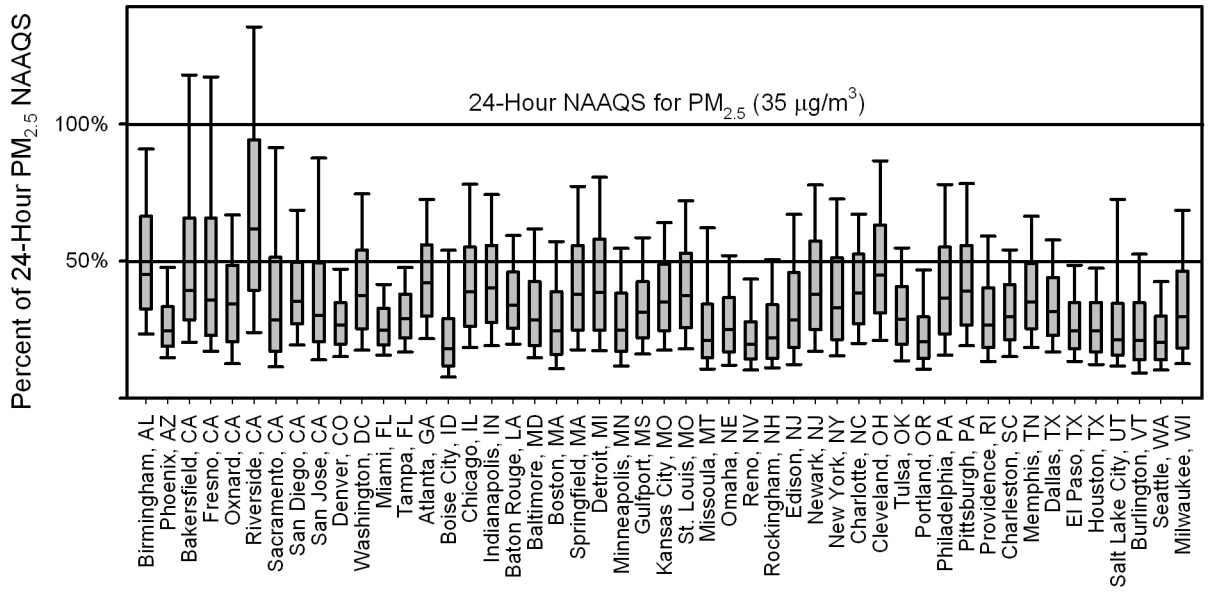

Figure 5-1. Distribution of monitor-specific 24-hour $PM_{2.5}$ concentrations compared to the daily NAAQS (35 $\mu g/m^3$): 49 STN monitors, 2001–2005.

The annual average $PM_{2.5}$ concentrations measured by STN monitors for the 5 years from 2001 through 2005 also varied greatly across the 49 CBSAs (Figure 5-2). Compared with the daily concentrations, a greater proportion of the CBSAs appeared to have at least one annual average above the NAAQS for $PM_{2.5}$ of 15 μg/m^3 during 2001–2005. The three California CBSAs with high 24-hour $PM_{2.5}$ concentrations also had high annual average concentrations. At the lower end of the distribution, 13 CBSAs consistently had average $PM_{2.5}$ concentrations below 10.5 μg/m^3 (75% of the annual NAAQS). Five of these CBSAs also had 10 percent or fewer individual days above 17.5 μg/m^3: Denver, CO; Miami, FL; Rockingham, NH; El Paso, TX; and Seattle, WA (Figure 5-1).

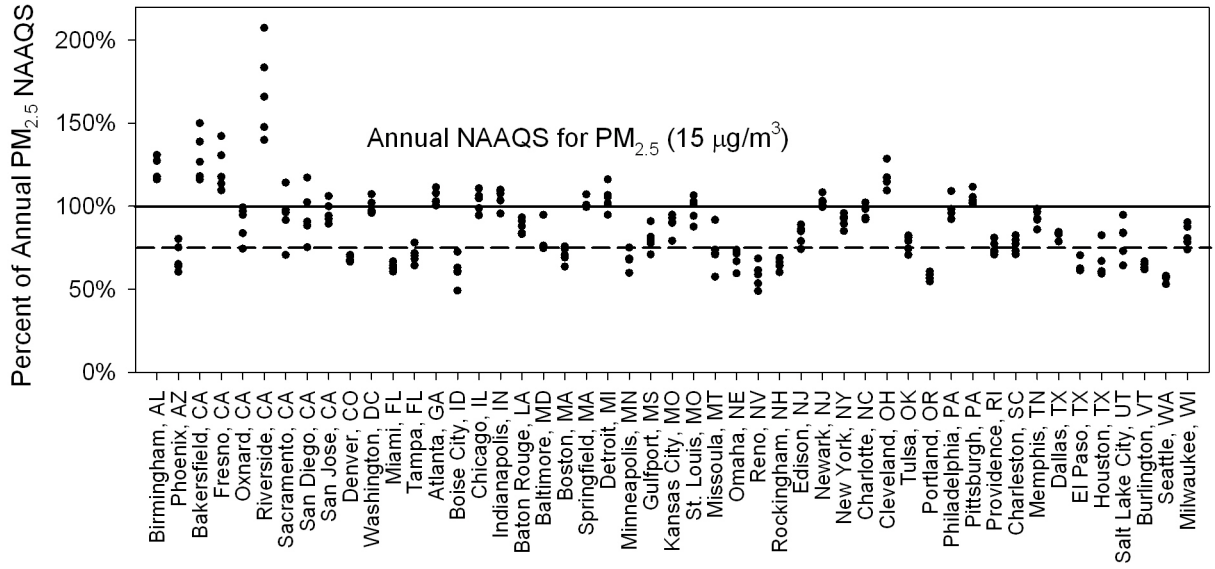

Figure 5-2. Percent of the annual NAAQS (15 μg/m^3) for each monitor-specific annual mean $PM_{2.5}$ concentration: 49 STN monitors, 2001–2005. (Circles = single year average).

Overall Mean Concentrations

The inter-comparison across all pollutants of the distributions of monitor-specific mean pollutant concentrations is complicated by varying units of measurement and by great differences in the mean concentrations (Table 5-1). Gaseous pollutants are measured in parts per hundred million (CO) or parts per billion (O_3, NO_2, SO_2). Particle mass ($PM_{2.5}$, PM_{10}), the major ions (NH_4, NO_3, SO_4), and carbonaceous material (elemental carbon [EC], organic carbon [OC]) are measured in $\mu g/m^3$. The elemental constituents of PM are measured in ng/m^3. Even for pollutants measured in the same units, the grand mean concentrations can vary by orders of magnitude, for example, sulfur at 1011.4 ng/m^3 to selenium at 1.5 ng/m^3.

The Speciation Trends Network (STN) monitors in southern California (Riverside, Fresno, and Bakersfield) had the highest concentrations of $PM_{2.5}$ and traffic-related pollutants (OC, and NO_3). The Pittsburgh, PA STN monitor had the highest concentrations of pollutants related to coal combustion (Se, SO_4, S, and SO_2). The Birmingham, AL STN monitor had the highest concentrations of Zn and Mn. The STN monitors in the northeastern urban corridor (New York, NY; Providence, RI) had the highest concentrations for residual oil fly ash (Ni and V). The highest concentrations of wood smoke pollutants and motor vehicles were not consistent in a particular STN monitor or set of monitors. Many of the crustal elements (Si, Al, Ti, and Ca) were highest in Phoenix, AZ and El Paso, TX.

Table 5-1. STN Monitors with the Highest Pollutant Concentrations Overall and by Season: 49 STN Monitors, 2001–2005

Pollutant		Mean Concentration	STN Monitor with Highest Concentration		
			Overall	Winter	Summer
Fine particulate matter	$PM_{2.5}$	13.1 $\mu g/m^3$	Riverside	Bakersfield	Riverside
Coarse particulate matter	PM_{10}	26.6 $\mu g/m^3$	El Paso	El Paso	Riverside
Ozone	O_3	26.0 ppb	Burlington	Burlington	Fresno
Sulfate	SO_4	3.1 $\mu g/m^3$	Pittsburgh	Pittsburgh	Pittsburgh
Sulfur dioxide	SO_2	4.0 ppb	New York	New York	Pittsburgh
Selenium	Se	1.5 ng/m^3	Pittsburgh	Pittsburgh	Pittsburgh
Nitrogen dioxide	NO_2	17.6 ppb	Newark	Salt Lake City	Newark
Nitrate	NO_3	2.0 $\mu g/m^3$	Riverside	Bakersfield	Riverside
Elemental carbon	EC	0.7 $\mu g/m^3$	Newark	Riverside	Newark
Organic carbon	OC	4.5 $\mu g/m^3$	Fresno	Fresno	Missoula
Carbon monoxide	CO	6.3 pphm	Missoula	Phoenix	Boise City
Manganese	Mn	3.3 ng/m^3	Birmingham	Birmingham	Birmingham
Nickel	Ni	2.7 ng/m^3	New York	New York	New York
Vanadium	V	2.8 ng/m^3	Providence	New York	Newark
Potassium	K	82.4 ng/m^3	Missoula	Missoula	Indianapolis
Bromine	Br	3.3 ng/m^3	Riverside	Salt Lake City	Riverside
Silicon	Si	133.8 ng/m^3	Phoenix	El Paso	Houston
Aluminum	Al	36.7 ng/m^3	Phoenix	Phoenix	Miami
Titanium	Ti	6.6 ng/m^3	Phoenix	Reno	Miami
Calcium	Ca	70.9 ng/m^3	El Paso	El Paso	Omaha
Iron	Fe	101.3 ng/m^3	Cleveland	Phoenix	Cleveland
Copper	Cu	5.1 ng/m^3	St Louis	St Louis	Bakersfield
Zinc	Zn	16.2 ng/m^3	Birmingham	Birmingham	Birmingham
Sodium	Na	100.4 ng/m^3	San Jose	Miami	San Jose
Chlorine	Cl	42.3 ng/m^3	Miami	Miami	Miami

The inter-comparison across all pollutants of the distributions of monitor-specific mean pollutant concentrations is facilitated by scaling each pollutant to the pollutant-specific overall STN network grand mean concentration for 2001-2005 (Figure 5-3). Criteria air pollutants ($PM_{2.5}$, PM_{10}, CO, NO_2, O_3, SO_2), for which EPA has set a National Ambient Air Quality Standard (NAAQS), show less monitor-specific variation around the overall STN grand mean than most PM constituents, which are not specifically regulated. Certain PM constituents (Se, OC, Br, and K) also show relatively tighter distributions, while Nickel (Ni) and chlorine (Cl) had the greatest variation in monitor-specific means.

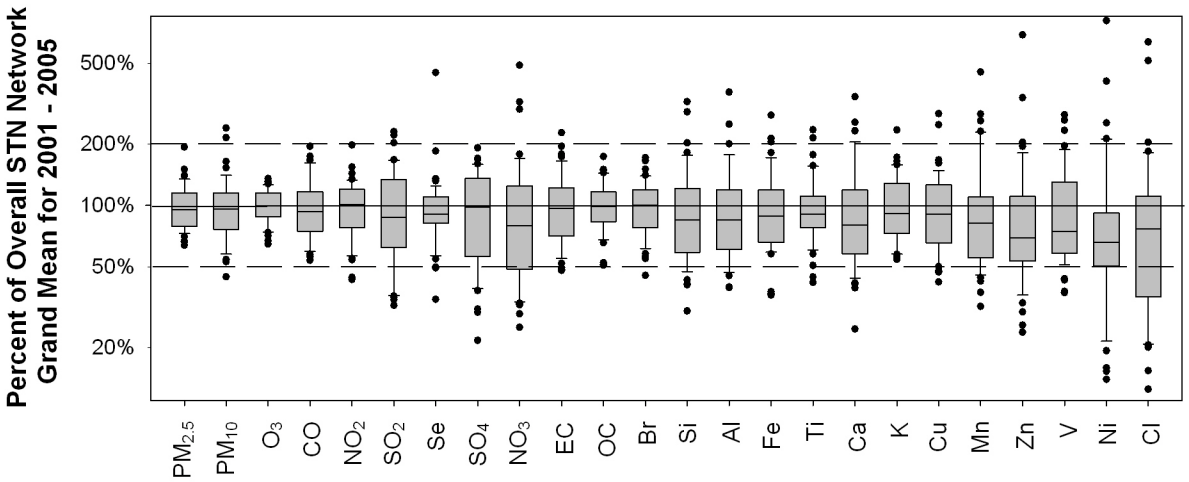

Figure 5-3. Distribution of monitor-specific mean pollutant concentrations as percent of the pollutant-specific overall STN network grand mean concentration: 49 STN monitors, 2001–2005.

Regional and Seasonal Patterns

Over this 5-year period, mean fine particulate matter ($PM_{2.5}$) concentrations were higher in the Eastern United States and in California, and lower in the Northwest, the inter-mountain West, and the Central United States. (Figure 5-4).

Seasonally, $PM_{2.5}$ concentrations were higher during the winter months (December – February) in California's central valley and in Salt Lake City, UT, but higher during the summer months (June – August) in southern California and in the Eastern United States. (Figure 5-5).

Figure 5-4. Mean fine particulate matter ($PM_{2.5}$) concentrations as proportions of 25 µg/m^3: 49 STN monitors, 2001–2005.

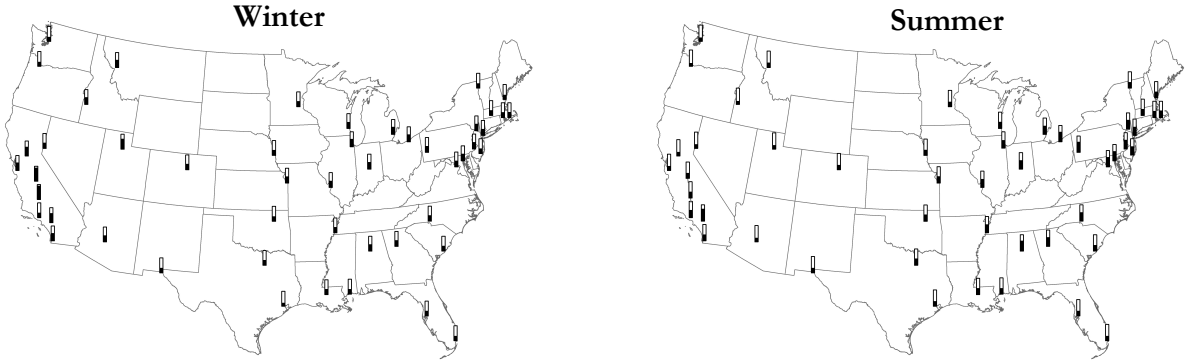

Figure 5-5. Mean winter and summer fine particulate matter ($PM_{2.5}$) concentrations as a proportion of 35 µg/m^3: 49 STN monitors, 2001–2005.

PM constituents and gaseous co-pollutants showed various patterns of seasonal means concentrations (Figure 5-6). Ozone and oxidation products (sulfate and organic carbon) were all higher in the summer, while sulfur dioxide, the precursor to sulfate particles, was higher in the winter. Elements associated with wind-blown dust (Si, Al, Ti, and Ca) (United States Environmental Protection Agency 2009c) were also generally higher in the summer. Conversely, higher winter concentrations were observed for PM constituents and gaseous co-pollutants associated with mobile source emissions (NO_3, CO, NO_2, and elemental carbon) and for chlorine and bromine (possibly from road salt) (Gao et al. 2006). Zinc (possibly an indicator of tire wear) (Davis et al. 2001) also showed higher winter concentrations. The remaining PM constituents did not show strong seasonal ratios, including elements associated with residual-fuel oil (Mn, V, and Ni) and with coal combustion (Se).

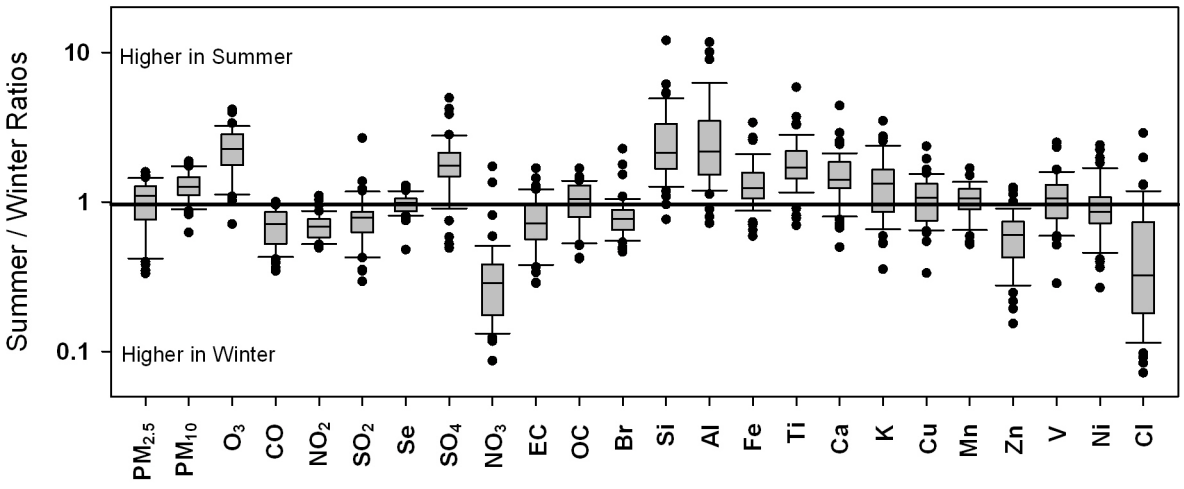

Figure 5-6. Distributions of monitor-specific ratios of mean concentrations in the summer (June – August) and the winter (December – February): 49 STN monitors, 2001–2005.

Sulfate (SO_4) concentrations were higher in the Eastern United States, reflecting the major contribution of sulfur oxides from fossil fuel combustion (Figure 5-7). Similarly, the highest SO_2 concentrations were also found in the Northeast.

Figure 5-7. Mean sulfate (SO_4) concentrations as a proportion of 10 μg/m^3: 49 STN monitors, 2001-2005.

Selenium (Se) concentrations, probably related to coal combustion and coking process (Gildemeister et al. 2007), were highest in CBSAs with steel manufacturing (Pittsburgh, PA at 6.7 ng/m^3 (not mapped) and Cleveland, OH at 2.8 ng/m^3) and higher than average in the Midwest and in the Northeastern United States (Figure 5-8).

Figure 5-8. Mean selenium (Se) concentrations as a proportion of 3 ng/m^3: 49 STN monitors, 2001-2005.

Nitrate (NO_3) concentrations show a different spatial distribution more reflective of mobile source emissions with the highest concentrations in California (Figure 5-9). In the Eastern United States, the north-south gradient of nitrate concentrations may reflect the temperature dependence of the partition of nitrate into the gas or particle phase (Lee and Hopke 2006).

Figure 5-9. Mean nitrate (NO_3) concentrations as a proportion of 10 μg/m^3: 49 STN monitors, 2001-2005.

Silicon (Si) concentrations are much higher in the arid Southwestern United States and other locations impacted by wind-blown sandy soils (Figure 5-10). Since the monitor-specific means for aluminum and silicon are highly correlated (r = 0.96), the geographic pattern for aluminum concentrations was essentially identical to that for silicon.

Figure 5-10. Mean silicon (Si) concentrations as a proportion of 5 μg/m^3: 49 STN monitors, 2001-2005.

Iron (Fe) concentrations reflect the presence of iron in both sandy and clay soils and the additional contributions from manufacturing processes, such as steel production (Gildemeister et al. 2007), with the highest levels in Cleveland, OH and Birmingham, AL (Figure 5-11).

Figure 5-11. Mean iron (Fe) concentrations as a proportion of 300 ng/m^3: 49 STN monitors, 2001-2005.

Zinc (Zn) concentrations, in contrast to Fe, are almost entirely related to manufacturing processes (Kim et al. 2007), such as steel production, with the highest levels in Birmingham, AL and Cleveland, OH (Figure 5-12).

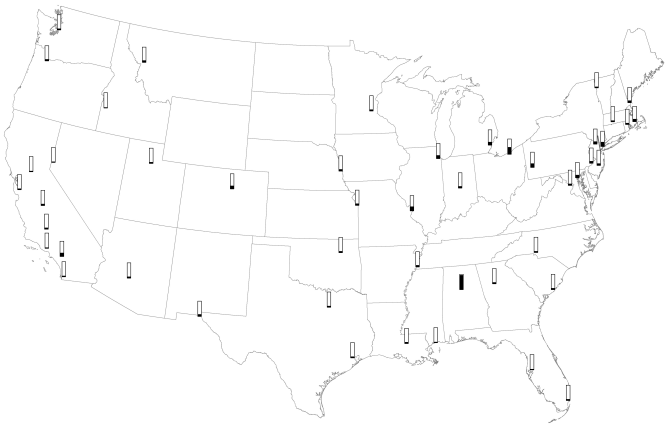

Figure 5-12. Mean zinc (Zn) concentrations as a proportion of 120 ng/m^3: 49 STN monitors, 2001-2005.

Elemental carbon (EC) concentrations show a more uniform distribution across the United States (Figure 5-13). This lack of a strong geographic pattern reflects the contributions of many combustion sources to elemental carbon concentrations.

Figure 5-13. Mean elemental carbon concentrations as a proportion of 2 μg/m^3: 49 STN monitors, 2001-2005.

Organic carbon (OC) concentrations also show a relatively uniform distribution across the United States, but with higher concentrations in California's central valley. (Figure 5-8).

Figure 5-14. Mean organic carbon concentrations as a proportion of 10 μg/m^3: 49 STN monitors, 2001-2005.

Vanadium (V) concentrations were high in seaport cities and in the northeastern metropolitan areas reflecting vanadium in residual fuel oil used for shipping and for domestic heating (Figure 5-15).

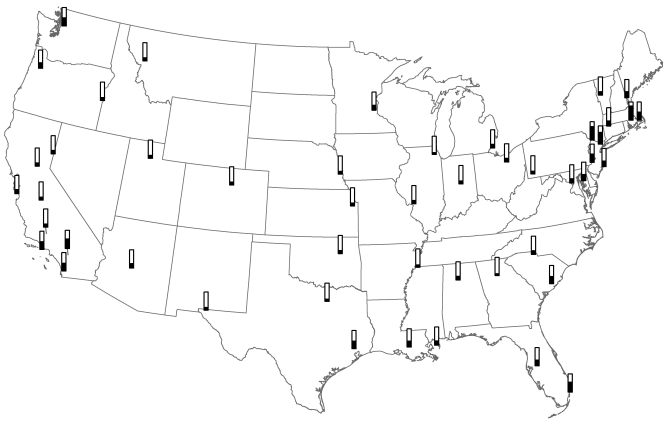

Figure 5-15. Mean vanadium (V) concentrations as a proportion of 10 ng/m^3: 49 STN monitors, 2001-2005.

Nickel (Ni) concentrations were similarly high in two major seaports: New York, NY with 22 ng/m^3 and San Jose, CA with 12 ng/m^3. Excluding these two cities, nickel concentrations were very low throughout the United States (Figure 5-16).

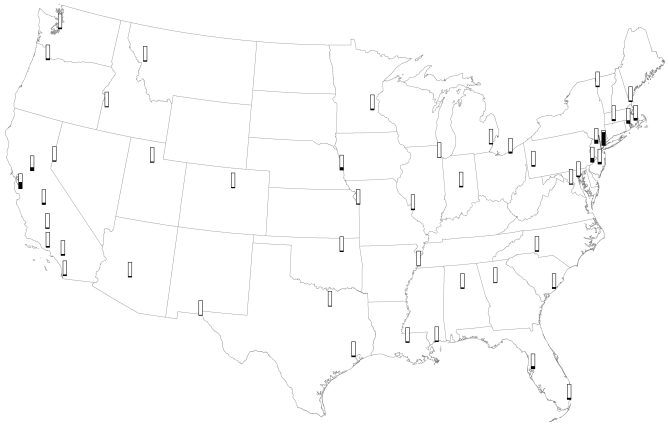

Figure 5-16. Mean nickel (Ni) concentrations as a proportion of 25 ng/m^3: 49 STN monitors, 2001-2005.

Conclusions Regarding Mean Concentrations

Through the NAAQS process, EPA has already determined unhealthy levels of total fine particle mass ($PM_{2.5}$): 35 $\mu g/m^3$ for a 24-hour average and 15 $\mu g/m^3$ for an annual average. Further studies of the adverse health effects of human exposures to ambient $PM_{2.5}$ concentrations above these levels will not address the scientific uncertainties underlying the current standards. Five CBSAs had more than 10 percent of days above 35 $\mu g/m^3$: three in Southern California (Bakersfield, Fresno, and Riverside); Edison, NJ; and Houston, TX (Figure 5-1).

Conversely, some metropolitan areas may have too few days with air quality near the current standards to address key scientific uncertainties. Seven CBSAs had both 10 percent or fewer individual days above 17.5 $\mu g/m^3$ (half the current 24-hour NAAQS for $PM_{2.5}$) and 5-year average concentrations below 10.5 $\mu g/m^3$ (75% of the annual NAAQS for $PM_{2.5}$): Phoenix, AZ; Denver, CO; Miami, FL; Tampa, FL; Rockingham, NH; El Paso, TX; and Seattle, WA (Figures 5-1 and 5-2).

$PM_{2.5}$ and most key constituents of $PM_{2.5}$ are present within a four-fold range of concentrations throughout the United States. (Figure 5-3). Elemental carbon (EC) and organic carbon (OC) were widely distributed across the U.S. with little regional pattern (Figures 5-13 and 5-14). Other constituents showed regional patterns: vanadium (V) concentrations were highest in the northeastern metropolitan corridor (Figure 5-15), nitrate (NO_3) concentrations were highest in southern California (Figure 5-9), and sulfate (SO_4) concentrations were highest in the Eastern United States. (Figure 5-7). For these constituents, epidemiologic studies would benefit from the selection of a few characteristic metropolitan areas in each of these regions.

In the extreme, some $PM_{2.5}$ constituents have distributions skewed by extreme levels in only one or two metropolitan areas (Table 5-1 and Figure 5-3). Nickel (Ni) concentrations are extraordinarily high in two major seaports: New York, NY and San Jose, CA (Figure 5-16). Silicon (Si) concentrations are extraordinarily high in the desert southwest: El Paso, TX and Phoenix, AZ (Figure 5-10). Metals, such as iron (Fe) and zinc (Zn), have extreme concentrations in manufacturing and refining centers: Cleveland, OH; Phoenix, AZ; and Birmingham, AL (Figures 5-11 and 5-12). Selenium (Se) concentrations are extraordinarily high in steel manufacturing centers (Pittsburgh, PA and Cleveland, OH), but still had considerable variation across the remaining metropolitan areas (Figure 5-8). These metropolitan areas would tend to dominate any epidemiologic analyses of these specific constituents.

Chapter 6
Variability in Concentrations of Criteria Pollutants and Selected $PM_{2.5}$ Species

Indicators of Monitor-Specific Variability

For time-series and case-crossover studies of short-term exposures, greater day-to-day variability in ambient air pollutant concentrations tends to increase the statistical power of such studies. Locations with greater variability will be more influential in the determination of concentration-response relationships. However, not all variations are equally useful; variations due to infrequent extreme events are less useful than frequent regular variation.

For total fine particle mass ($PM_{2.5}$), the city-specific interquartile ranges vary closely with the city-specific means (r^2 = 0.82), while city-specific coefficients of variation, the monitor-specific variance divided by the monitor-specific mean expressed as a percentage, are independently distributed around the grand mean of 62 percent (Figure 6-1).

Extreme variability generally reflects episodic extreme events. For example, the outlier interquartile range in Oxnard, CA, reflects the episodic impact of the Los Angeles area. The three most extreme coefficients of variation (Salt Lake City, UT; Missoula, MT; and Boise City, ID) reflect infrequent extreme pollution events in normally low particle areas.

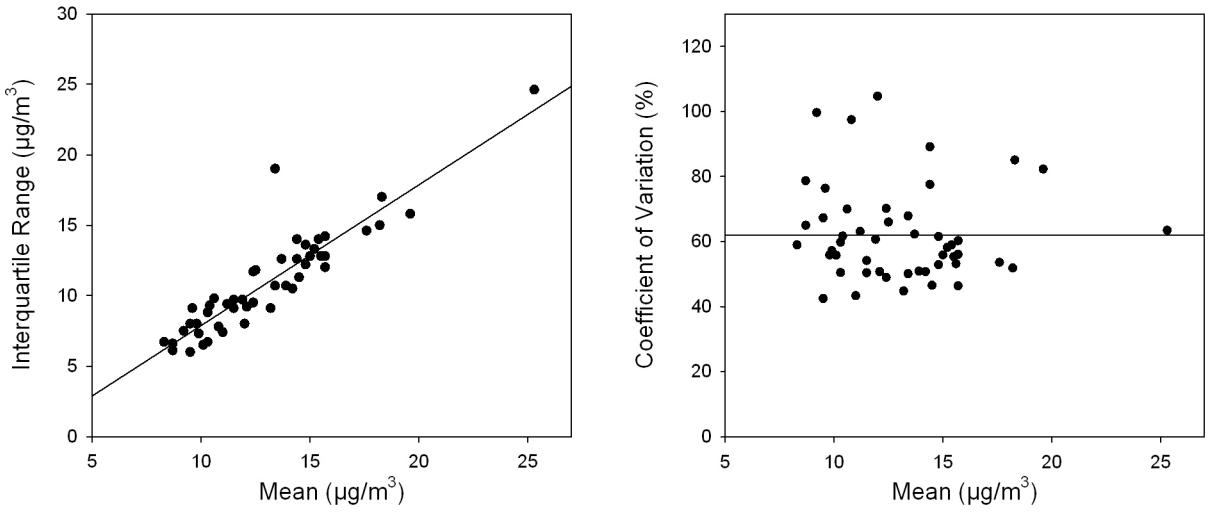

Figure 6-1. Distributions of monitor-specific interquartile range and coefficients of variation by monitor-specific means for $PM_{2.5}$: 49 STN monitors, 2001–2005.

Variability Relative to the Mean

The distributions of coefficients of variation show considerable differences across the criteria pollutants and the components of PM (Figure 6-2). Pollutants with monitor-specific coefficients of variation greater than 100 percent have substantially variability relative to their means than pollutants with monitor-specific coefficients of variations less than 100 percent. Pollutants with lower coefficients of variation generally have relatively stable source contributions, such as those pollutants related to local mobile source emissions in these urban areas.

For most pollutants, the monitor-specific coefficients of variation fell within a tight range from 40 to 100 percent of the pollutant-specific means. Even pollutants with the lowest coefficients of variation, ozone and NO_2, have shown sufficient variation for epidemiologic studies. Pollutants with higher coefficients of variation may have strong seasonal patterns, for example, nitrate (NO_3), or episodic source events, for example, chlorine (Cl). Potassium (K) is associated with seasonal burning of wood and other plant materials. Other PM constituents (Si, Al, and Ti) may be related to wind-blown dusts. Since seasonal patterns in pollutants are generally removed by modeling or by design, the useful variation for an epidemiologic study is the within season variation.

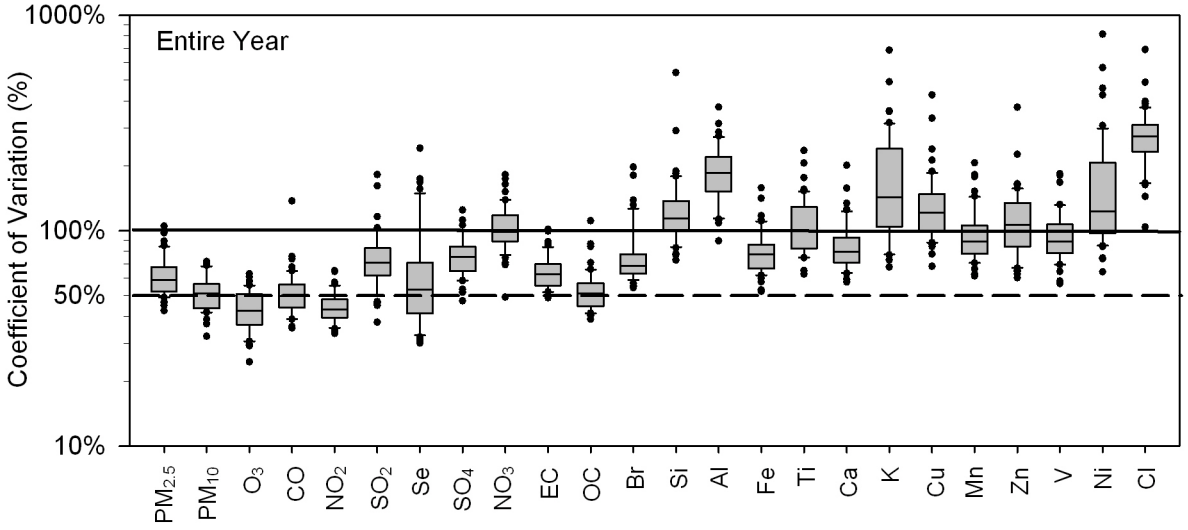

Figure 6-2. Distributions of monitor-specific coefficients of variation for various pollutants over the entire year: 49 STN monitors, 2001–2005.

Seasonal Differences in Coefficient of Variation

Season-specific distributions of the monitor-specific coefficients of variation provide insights into the sources of these pollutants (Figure 6-3). For example, chlorine (Cl) showed higher coefficients of variation in both seasons with the highest summer values in Tulsa, OK and Salt Lake City, UT suggestive of wind-blown salt and the highest winter values in Baltimore, MD; Charleston, SC, and Houston, TX. Conversely, for sulfate (SO_4), the high coefficients of variation disappeared with seasonal adjustment.

Potassium (K) had greater coefficients of variation in the summer, perhaps related to episodic controlled or uncontrolled combustion of plant materials, with high values in mid-western cities. In the summer, high values were observed for Indianapolis, IN (470%) and Minneapolis, MN (386%); while, in the winter, high values were observed for Miami, FL (622%) and Charleston, SC (379%).

Aluminum (Al) had greater summer coefficients of variation in mid-western cities, such as St. Louis, MO (405%) and Indianapolis, IN (352%), and greater winter values in the southern cities, such as Miami, FL (289%); El Paso, TX (272%); and Houston, TX (237%), perhaps related to episodic dust events involving windborne clay soils rich in kaolin.

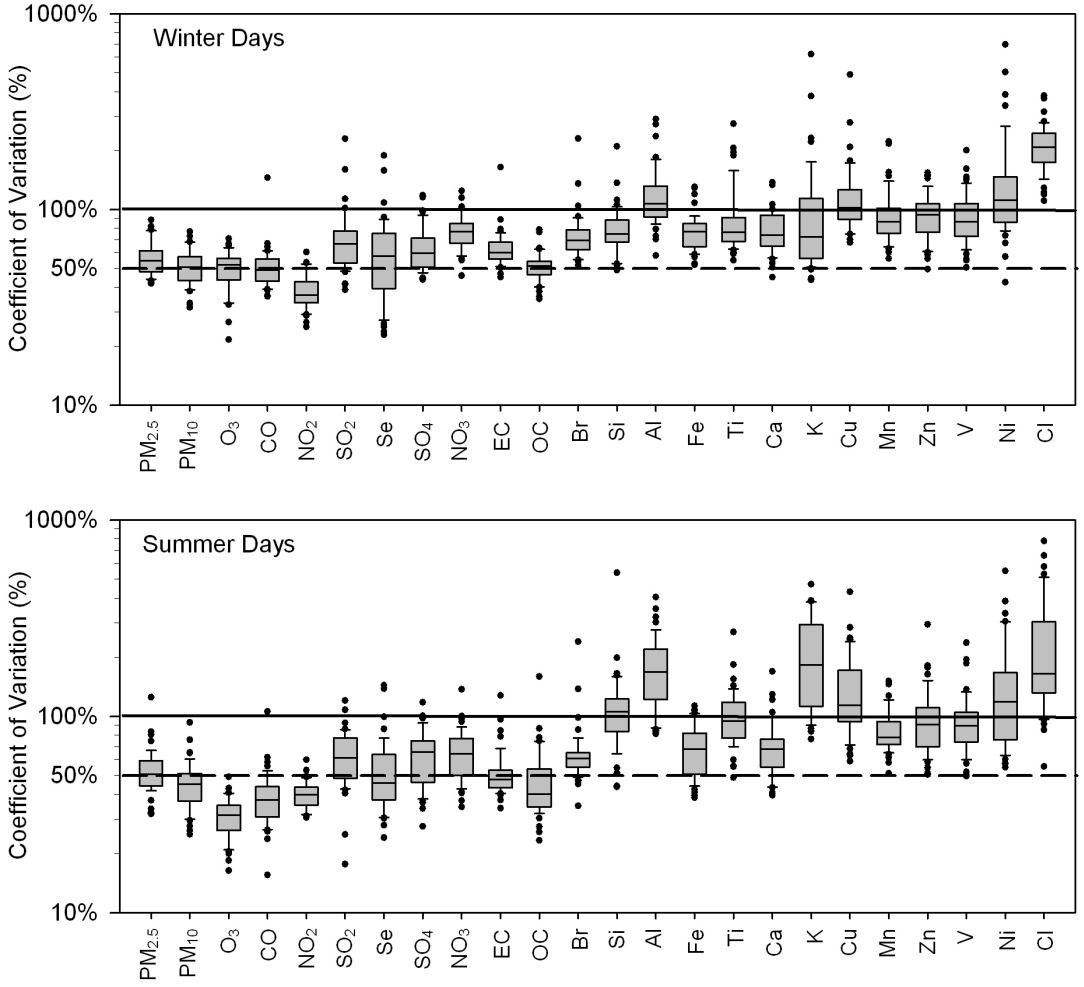

Figure 6-3. Distributions of monitor-specific coefficients of variation for various pollutants for winter and for summer days: 49 STN monitors, 2001–2005.

The increased summer-time coefficients of variation for K and Al are particularly evident with monitor-specific ratios of the summer and winter coefficients of variation (Figure 6-4).

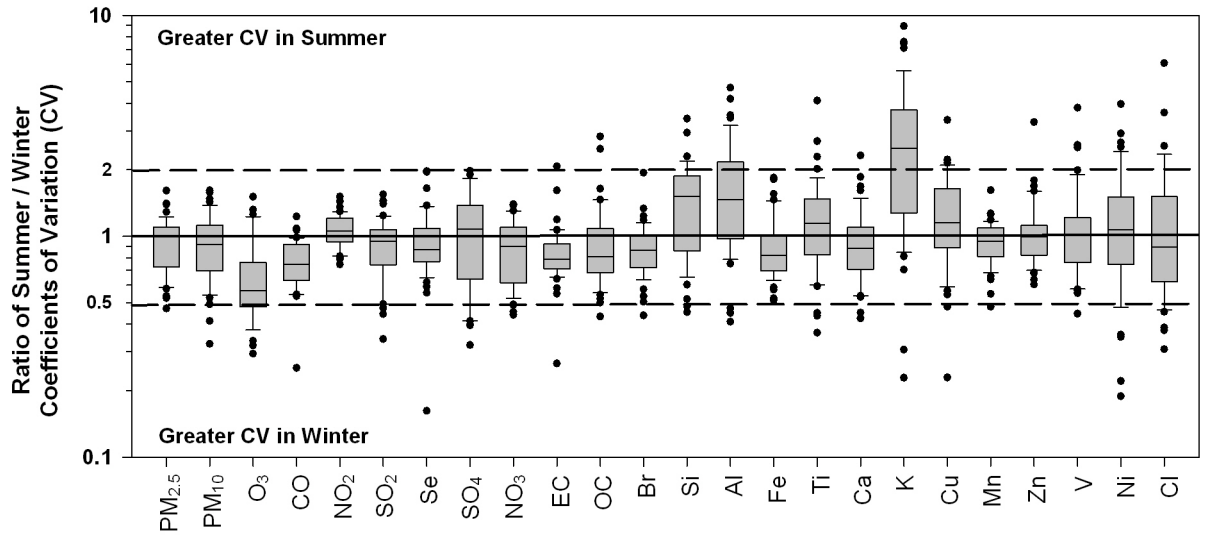

Figure 6-4. Monitor-specific ratios of summer/winter coefficients of variation for various pollutants: 49 STN monitors, 2001–2005.

Conclusions Regarding Coefficients of Variation

For most metropolitan areas with Speciation Trends Network (STN) monitors, total fine particle mass ($PM_{2.5}$) and most $PM_{2.5}$ constituents had modest coefficients of variation between 50 percent and 100 percent of the monitor specific means reflecting a regular pattern with small daily variations (Figure 6-2). This general pattern was present in both the winter and summer months (Figure 6-3).

A few constituents had skewed distributions of coefficients of variation, indicating a pattern of isolated extreme concentrations (Figure 6-1). The constituents showing the most skewed distributions were nickel (Ni), potassium (K), aluminum (Al), and chlorine (Cl).

Chapter 7
Correlations with Total Fine Particle Mass ($PM_{2.5}$)

Since the association between total fine particle mass ($PM_{2.5}$) and mortality has already been well established, the specific $PM_{2.5}$ constituents most likely to explain this association should show strong associations with total $PM_{2.5}$ mass. The monitor-specific coefficient of determinations (the square of the correlation coefficient, R^2) between $PM_{2.5}$ and other pollutants vary greatly across pollutants and seasons (Figures 7-1 to 7-15). Horizontal reference lines indicate strong correlations (R^2 greater than 49%), moderate correlations (R^2 between 25% and 49%), and weak correlations (R^2 less than 10%). City-specific correlations are presented in Tables A-5 and A-8 in the Appendix.

Particulate matter that is less than 10µm in aerodynamic diameter (PM_{10}) was strong-to-moderately correlated with total $PM_{2.5}$ mass in the majority of cities, while moderate correlations with ozone (O_3) were observed in the majority of cities (Figure 7-1). Not fully reflected by the unsigned coefficient of determination, the overall relationship between $PM_{2.5}$ and O_3 varied widely across cities, exhibiting both negative and positive correlations. Much of this variation appeared to be due to seasonal patterns. For every monitor, O_3 and $PM_{2.5}$ had weak-to-moderate inverse correlations during the winter. In contrast, during the summer the correlations were all positive or weak. In the winter the strongest inverse correlations were observed in California and in the Northeast while in the summer the strongest positive correlations were found in the Southeast.

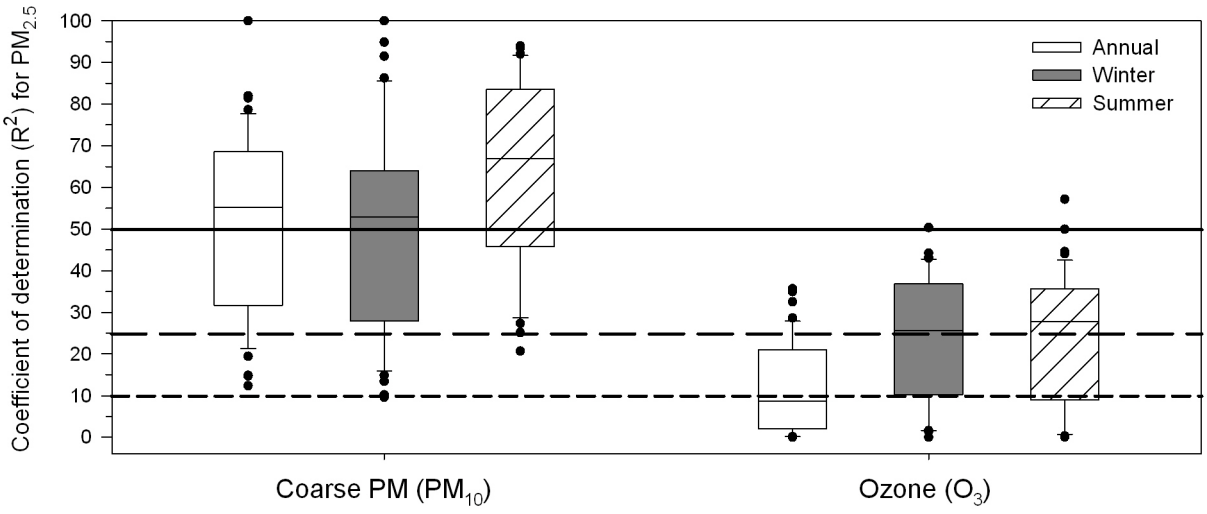

Figure 7-1. Distributions of the monitor-specific percentage of the daily variation in $PM_{2.5}$ determined by the variation PM_{10} and O_3 for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

Fossil Fuel Combustion

Sulfate (SO_4) from fossil fuel combustion had the highest R^2 with total $PM_{2.5}$ mass of any single $PM_{2.5}$ constituent, with summertime R^2 above 70 percent in most Core Based Statistical Areas (CBSAs) (Figure 7-2). For the entire year, SO_4 was highly correlated with total $PM_{2.5}$ mass for most CBSAs east of the Mississippi River (Figure 7-3). Selenium (Se), a more specific marker for coal combustion, had lower correlations in most cities, but was strongest in Washington, DC; Chicago, IL; Baton Rouge, LA; Baltimore, MD; Detroit, MI; and Pittsburgh, PA. The seasonal differences for SO_4 and SO_2 reflect seasonal differences in the conversion of gaseous SO_2 into particulate SO_4.

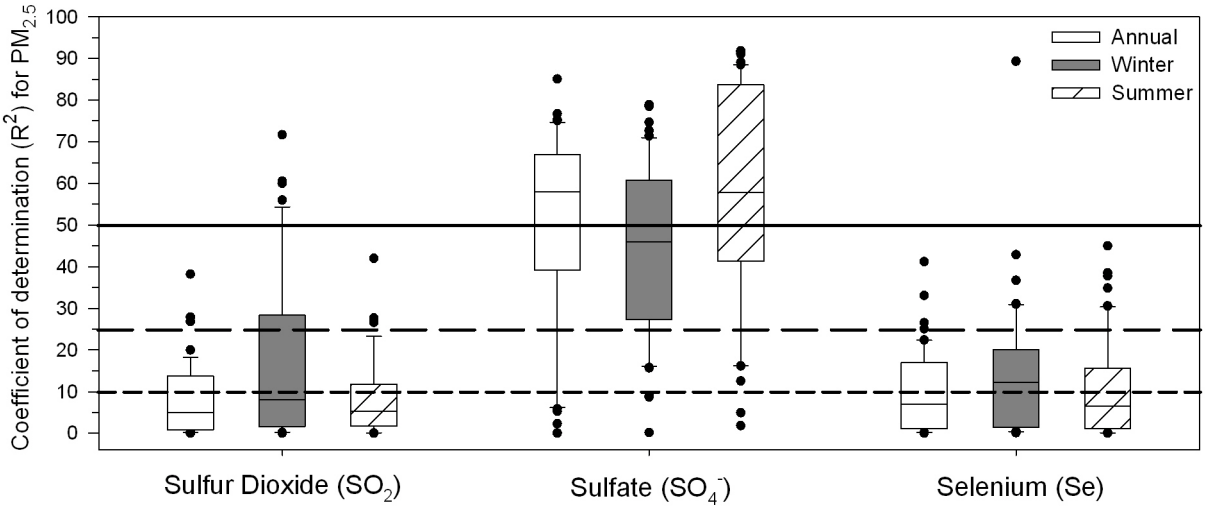

Figure 7-2. Distributions of the monitor-specific percentage of the daily variation in $PM_{2.5}$ determined by the variation in selected markers of coal combustion for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

Figure 7-3. Monitor-specific correlations between $PM_{2.5}$ and sulfate: 49 STN monitors, 2001–2005.

Mobile Sources

Potential markers of mobile sources (NO_2, NO_3, EC, OC, and CO) were moderate-to-strongly correlated with total $PM_{2.5}$ mass in many cities, especially in the winter (Figure 7-4). Total $PM_{2.5}$ mass was moderately or strongly correlated with organic carbon (OC) throughout the United States (Figure 7-5). For nitrogen dioxide (NO_2), the strongest correlations with total $PM_{2.5}$ mass were observed in the Northeast, while, for nitrate (NO_3), the strongest correlations were observed in California and the Pacific Northwest.

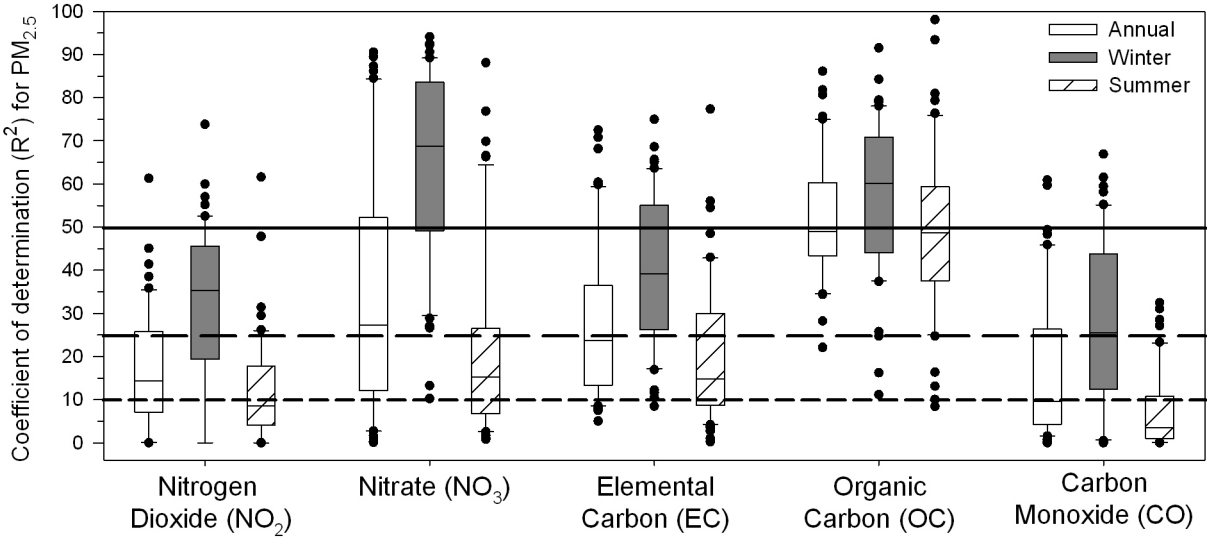

Figure 7-4. Distributions of the monitor-specific percentage of the daily variation in total $PM_{2.5}$ mass determined by the variation in selected markers of mobile sources for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

Figure 7-5. Monitor-specific correlations between total $PM_{2.5}$ mass and organic carbon: 49 STN monitors, 2001–2005.

Residual-Oil Fly Ash

Residual-oil fly ash may be indicated by manganese (Mn), nickel (Ni) or vanadium (V), depending on the source of the fuel oil . During the winter, a few cities, especially in the Northeastern United States, had moderate correlations between $PM_{2.5}$ and these three metals (Figure 7-7). During the summer, only weak correlations were observed between these pollutants and $PM_{2.5}$ in the majority of cities with no real geographic pattern (Figure 7-6).

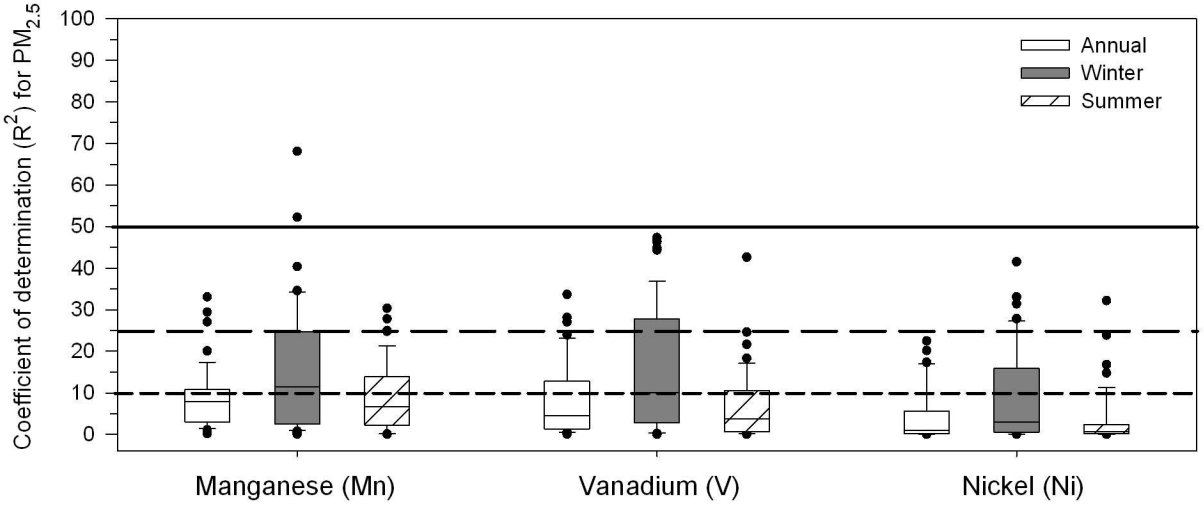

Figure 7-6. Distributions of the monitor-specific percentage of the daily variation in total $PM_{2.5}$ mass determined by the variation in selected markers of residual-oil fly ash for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

Figure 7-7. Monitor-specific correlations between total $PM_{2.5}$ mass and vanadium during the winter months (December – February): 49 STN monitors, 2001–2005.

Biomass Combustion

Potential markers for biomass combustion, potassium (K) and bromine (Br), had moderate to strong correlations with total $PM_{2.5}$ mass in many cities, especially during the winter (Figure 7-8). During the winter, the strongest correlations were for Speciation Trends Network (STN) monitors located in the western part of the United States, especially in the Northwest, but were still moderate in the Northeastern United States and California (Figure 7-9). Tampa, FL is an exception to the national pattern, perhaps due to local agricultural burning in central Florida.

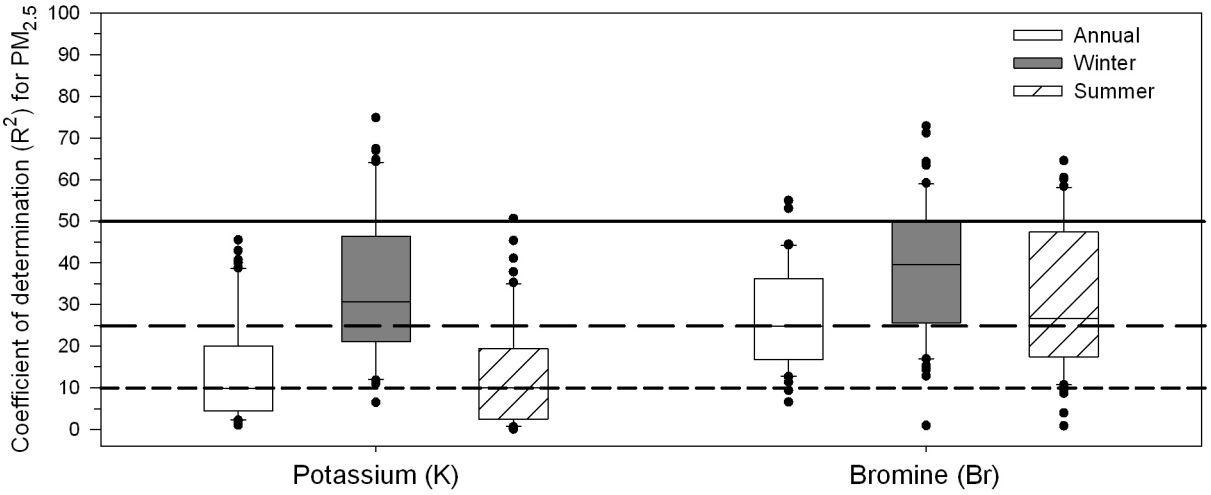

Figure 7-8. Distributions of the monitor-specific percentage of the daily variation in total $PM_{2.5}$ mass determined by the variation in selected markers of biomass combustion for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

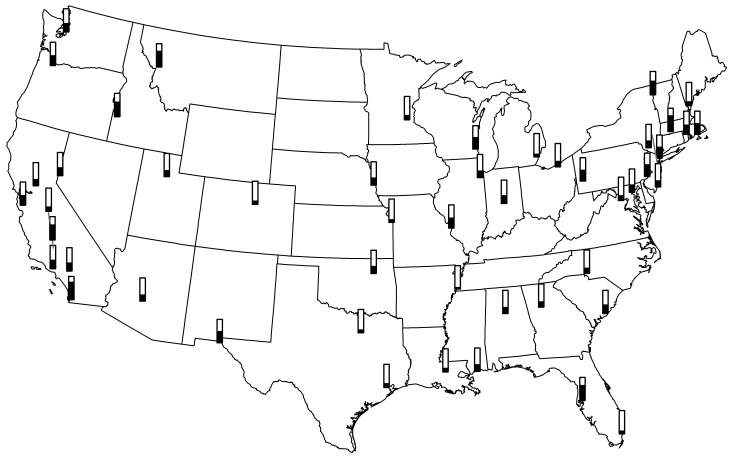

Figure 7-9. Monitor-specific correlations between total $PM_{2.5}$ mass and potassium (K) during winter: 49 STN monitors, 2001–2005.

Crustal Materials

Elements common in wind-blown crustal materials, aluminum (Al) and silicon (Si), appeared to only be weakly correlated with total $PM_{2.5}$ mass (Figure 7-10). In the summer, silicon was weakly correlated aside from the desert southwest and agricultural Midwest (Figure 7-11). The higher correlations in Seattle, WA and Tampa, FL may be related to transoceanic transportation of sand during extreme desert dust events. Overall for the other crustal elements, calcium (Ca) and titanium (Ti) produced moderate correlations in only a few cities. For Si and Ti, the monitor-specific averages were similar regardless of season or region.

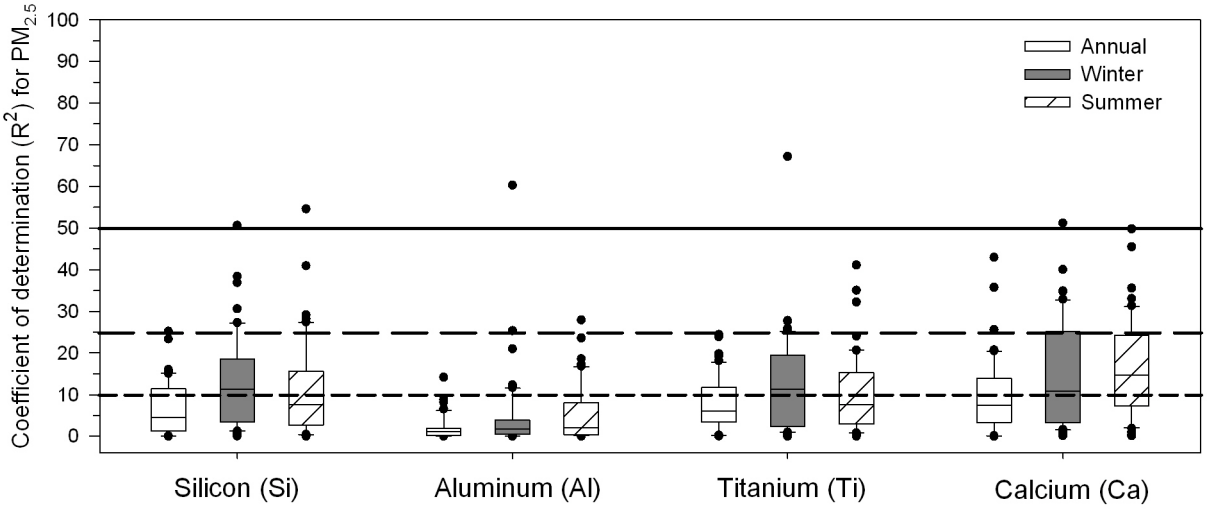

Figure 7-10. Distributions of the monitor-specific percentage of the daily variation in total $PM_{2.5}$ mass determined by the variation in selected markers of crustal elements for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

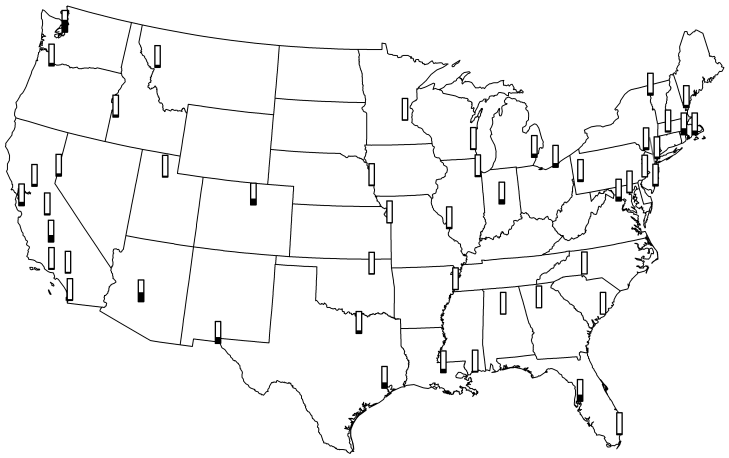

Figure 7-11. Monitor-specific correlations between total $PM_{2.5}$ mass and silicon (Si) during summer: 49 STN monitors, 2001–2005.

Metallic Elements

Metallic elements represent a complex pattern due to the wide variety of sources: windblown crustal materials, emissions from manufacturing processes, and emission from the use of manufactured products. Iron (Fe) was moderately correlated with total $PM_{2.5}$ mass in the majority of cities, with stronger correlations in the winter (Figure 7-12). The strongest year-round correlations were observed in Northern California, the Northwest, and the Northeast (Figure 7-13).

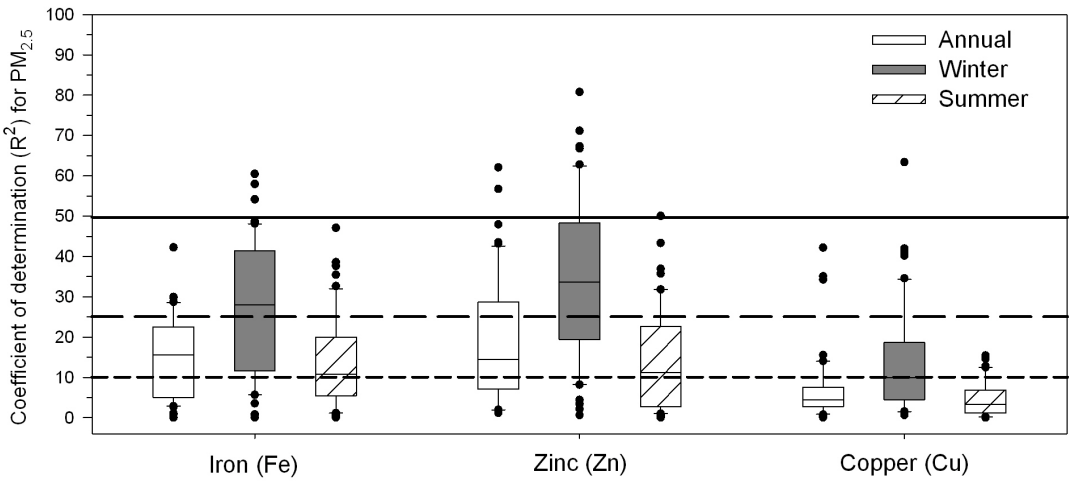

Figure 7-12. Distributions of the monitor-specific percentage of the daily variation in total $PM_{2.5}$ mass determined by the variation in selected metals for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

Figure 7-13. Monitor-specific correlations between total $PM_{2.5}$ mass and iron (Fe): 49 STN monitors, 2001–2005.

Sea Spray/Road Salt

Potential markers of sea spray/road salt, sodium (Na) and chlorine (Cl), had only weak to moderate correlations with total $PM_{2.5}$ mass regardless of location or season (Figure 7-14). During the winter, chlorine (Cl) was moderately correlated in some cities in the Western and Northern parts of the country (Figure 7-15).

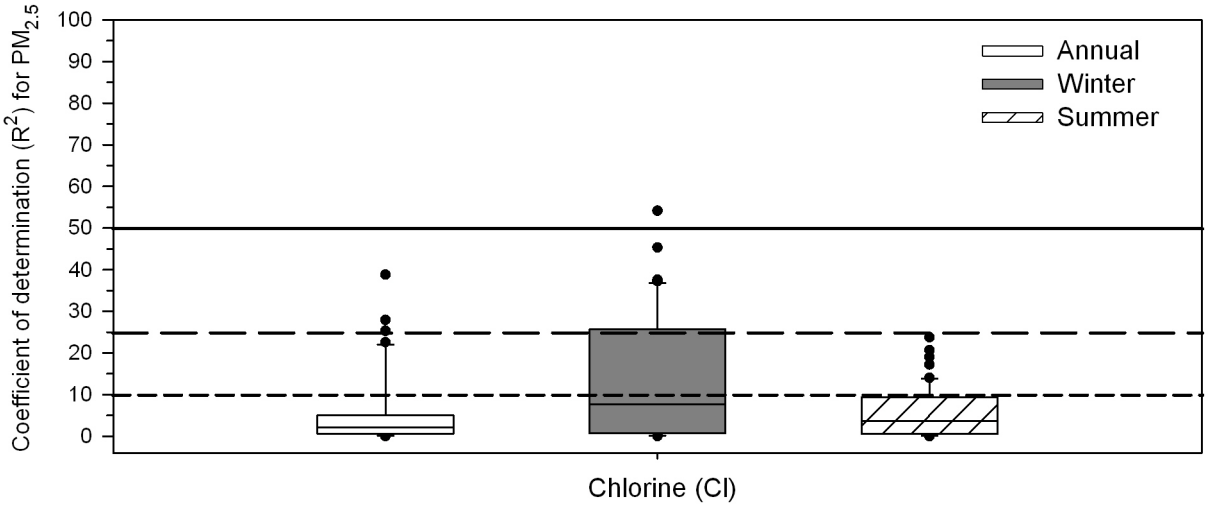

Figure 7-14. Distributions of the monitor-specific percentage of the daily variation in $PM_{2.5}$ determined by the variation in selected markers of sea spray/road salt for all days, winter days, and summer days: 49 STN monitors, 2001–2005.

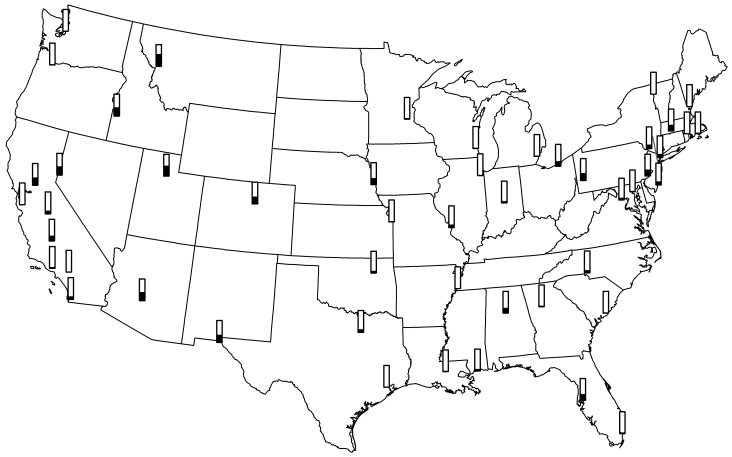

Figure 7-15. Monitor-specific correlations between total $PM_{2.5}$ mass and chlorine (Cl) during winter: 49 STN monitors, 2001–2005.

Conclusions Regarding Correlations with $PM_{2.5}$

Since previous epidemiologic studies in many metropolitan areas throughout the world have already well-established the association between total fine particle mass ($PM_{2.5}$) and increased mortality and hospitalizations, the examination of associations with specific PM constituents should begin with those constituents that covary with $PM_{2.5}$. For this report, the key PM constituents associated with total $PM_{2.5}$ mass are sulfate, nitrate during the winter, elemental carbon, and organic carbon. These constituents tend to be ones that are associated with significant health effects estimates possibly explaining the association of total $PM_{2.5}$ mass with mortality and hospitalization. However, previous studies have observed associations between copper, potassium, zinc, titanium, aluminum, chlorine, iron, nickel, silicon, and vanadium and adverse health effects (United States Environmental Protection Agency 2009a). Aluminum, chlorine, silicon, and titanium had low correlations with total $PM_{2.5}$ mass while correlations between $PM_{2.5}$ mass and the other constituents were source and therefore city dependent.

Chapter 8
Inter-Correlations Between Selected PM Constituents

Recent and future epidemiologic and clinical studies are focused on resolving the relative independent and joint effects of PM constituents and co-pollutants in a multipollutant context. Observational studies cannot resolve the independent effects of highly correlated air pollutants. Therefore, the ability to distinguish between various PM constituents would be enhanced by independent variation in these constituents as characterized by low pair-wise correlations.

In this chapter correlation within previously defined source categories is first examined to determine the effectiveness of these commonly used groupings. In the second section of this chapter correlations between selected constituents from different source categories are presented. The constituents selected are those shown to be associated with adverse health effects.

Within-Source Category

The monitor-specific coefficient of determinations (the square of the correlation coefficient, R^2) between $PM_{2.5}$ constituents vary greatly across pollutants and seasons. Figures 8-1 through 8-3 present within source categories, as identified in Chapter 7, annually and in the winter and summer seasons. Horizontal reference lines indicate strong correlations (R^2 greater than 49%), moderate correlations (R^2 between 25% and 49%), and weak correlations (R^2 less than 10%). City-specific coefficients of determination are presented in Tables A-9 to A-17 in the Appendix.

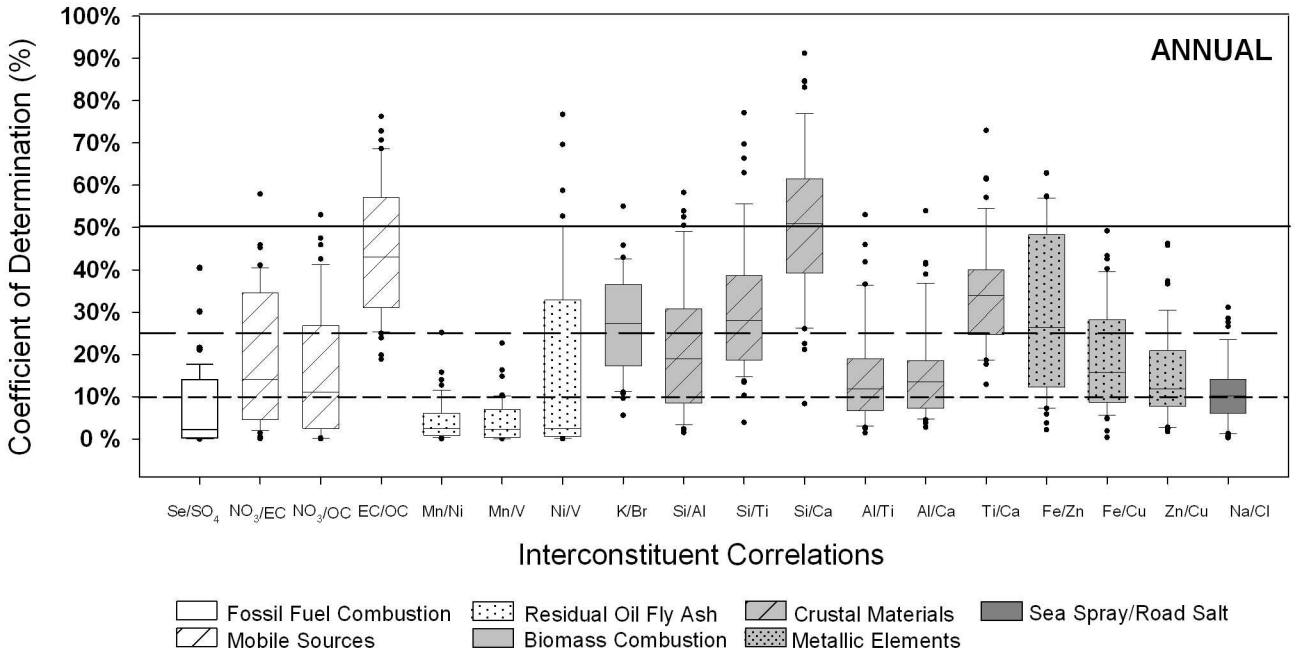

Figure 8-1. Distributions of monitor-specific of coefficients of determination for constituents within-source category for all days: 49 STN monitors, 2001–2005.

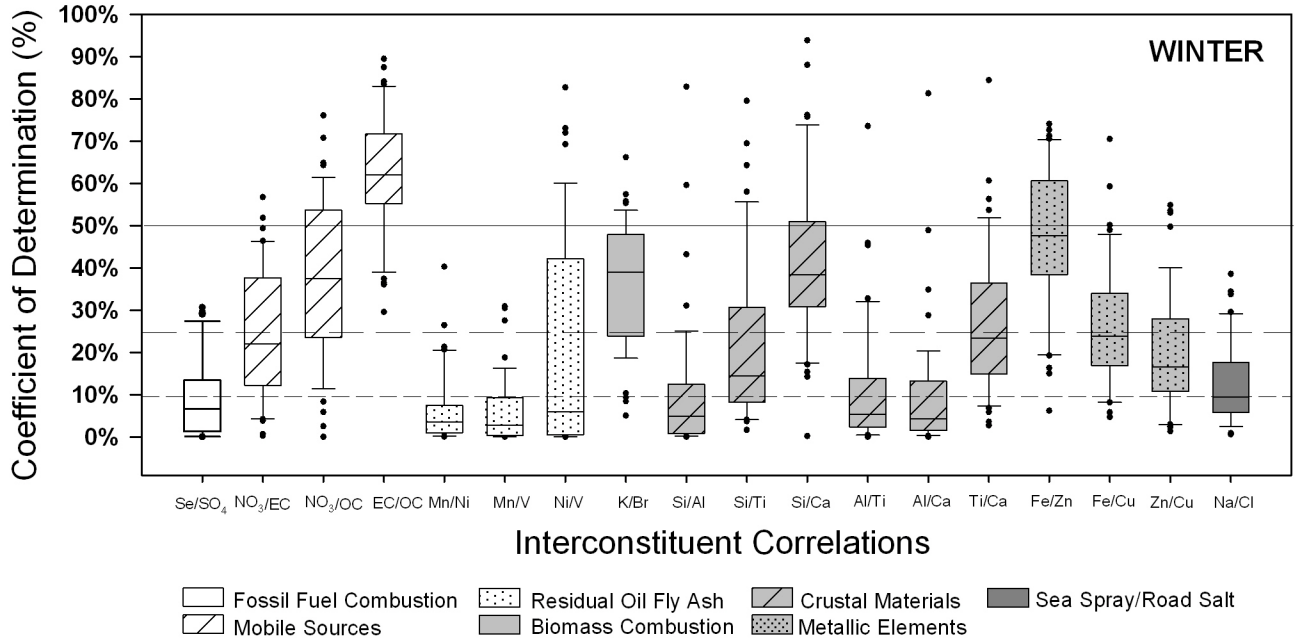

Figure 8-2. Distributions of monitor-specific of coefficients of determination for constituents within-source category for winter days: 49 STN monitors, 2001–2005.

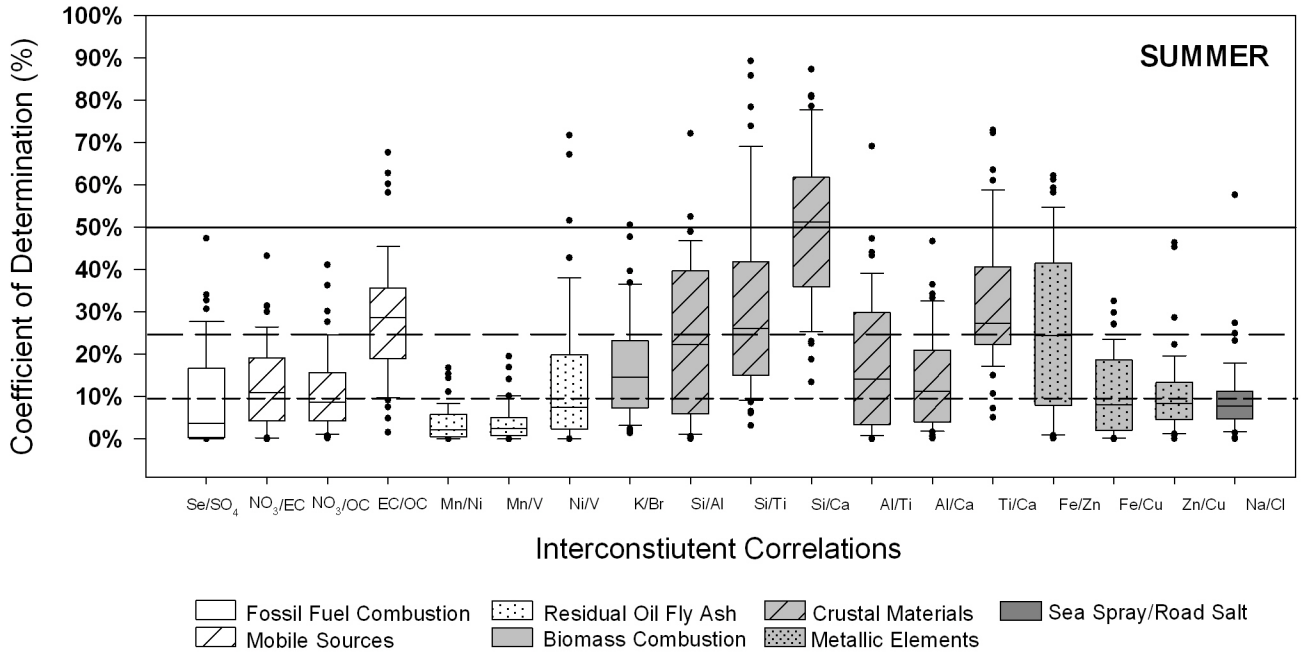

Figure 8-3. Distributions of monitor-specific of coefficients of determination for constituents within-source category for summer days: 49 STN monitors, 2001–2005.

Fossil Fuel Combustion

Sulfate and selenium were moderately correlated in only Baltimore, MD, Cleveland, OH, Detroit, MI, and Chicago, IL. These correlations are higher in the summer than in the winter.

Mobile Sources

Correlations between nitrate and elemental carbon, nitrate and organic carbon, and elemental carbon and organic carbon were on average higher in the winter than during the summer. The lower summer correlations possibly reflect the independent sources of secondary organic aerosols, while the higher winter correlations reflect the common combustion sources of both elemental carbon and primary organic aerosols. In the majority of cities moderate to strong correaltions were observed between elemental carbon and organic carbon regardless of season, and between nitrate and organic carbon during the winter. During the winter nitrate and elemental carbon had strong correlations in San Jose, CA and Springfield, MA.

Residual-Oil Fly Ash

Nickel and vanadium had high correlations during the winter in the Northeastern metropolitan corridor; Minneapolis, MN; and Seattle, WA; but low in other metropolitan areas. These correlations also appeared to be higher in the winter compared with the summer. Similarly, there were higher correlations between manganese and nickel and manganese and vanadium during the winter. The higher winter correlations are possibly due to the higher use of oil heating during the colder months.

Biomass Combustion

Potassium and bromine exhibited higher winter correlations with the strongest correlations occurring in Seattle, WA and Northeastern metropolitan areas.

Crustal Materials

Correlations between all of the crustal materials were higher in the summer than during the winter. Silicon and aluminum, the major elemental constituents of sandy and clay soils (United States Environmental Protection Agency 2009c), are highly correlated in the summer throughout the Eastern United States, but in the winter the monitor-specific correlations are high only in the desert Southwest. The influence of wind-blown dust arising from arid conditions and agricultural operations is apparent from these seasonal correlations. Silicon is also highly correlated with calcium in the majority of cities, and with titanium during the summer. In the winter high correlations between titanium and silicon were only observed in the Western United States. Aluminum was poorly correlated with both titanium and calcium in the majority of cities regardless of season. The exceptions were a few cities in California; Denver, CO; and Phoenix, AZ.

Metallic Elements

High correlations in the winter for zinc and iron, especially in northern metropolitan areas, may reflect a common source. Although metropolitan areas with steel manufacturing do show moderate winter-time correlations, the common source for zinc and iron appears to be present in less industrialized metropolitan such as Portland, OR, Chicago, IL, and New York City, NY. One possible common source might be dusts from mobile sources, such as tire and engine wear (Majestic et al. 2009; Sanders et al. 2003). Zinc and copper had low seasonal correlations in most metropolitan areas, but rose to moderate correlations in the densely populated, and high traffic, mid-Atlantic areas during the winter months. Similarly, iron and copper had higher correlations during the winter; however, there did not appear to be a spatial pattern to these correlations.

Sea Spray/Road Salt

Sodium and chlorine were not well correlated in the majority of metropolitan areas. The exceptions were those cities along bodies of water such as San Jose, CA, Providence, RI, Miami, FL, and Gulfport, MS.

Between-Source Categories

In order to summarize the results, cities were grouped into four regions (Northeast, South, Midwest, and West) as defined by the U.S. Census Bureau. In Figures 8-4 through 8-7 horizontal reference lines indicate strong correlations (R^2 greater than 49%), moderate correlations (R^2 between 25% and 49%), and weak correlations (R^2 less than 10%). The constituents were selected based on their significant associations with morbidity and mortality as well as their significant modification of these health effects. The selected elements include elemental carbon, organic carbon, copper, potassium, zinc, titanium, aluminum, chlorine, nitrate, sulfate, iron, nickel, silicon, and vanadium (United States Environmental Protection Agency 2009a). Inter-constituent correlations presented in the previous section (i.e., within-source category) will not be discussed in this section. City-specific correlations are presented in Tables A-9 to A-17 in the Appendix.

Midwest

Elemental carbon was moderately correlated with potassium, zinc, and iron. Average correlations between these constituents were mainly driven by Indianapolis, IN, and Milwaukee, WI. Organic carbon was moderately correlated with potassium in all Midwestern cities. Overall organic carbon was moderately correlated with iron except in Detroit, MI, Omaha, NE, and St. Louis, MO. Moderate correlations were observed between potassium and iron in all Midwestern cities except St. Louis, MO. Potassium was moderately correlated with silicon except in Indianapolis, IN, Kansas City, MO, and Minneapolis, MN.

Seasonal differences were noted in the above correlations. Moderate correlations between elemental carbon and potassium were observed in the winter but not the summer. Some correlations were not significant overall but were significant during certain seasons. During the winter organic carbon was moderately correlated with zinc and nitrate, potassium was moderately correlated with zinc, and nitrate was moderately correlated with sulfate. Nitrate was also moderated correlated with sulfate during the summer months. Moderate correlations were observed between organic carbon and sulfate, zinc, and nitrate; and iron and titanium in the summer. Only weak correlations were observed for the other inter-constituent correlation combinations

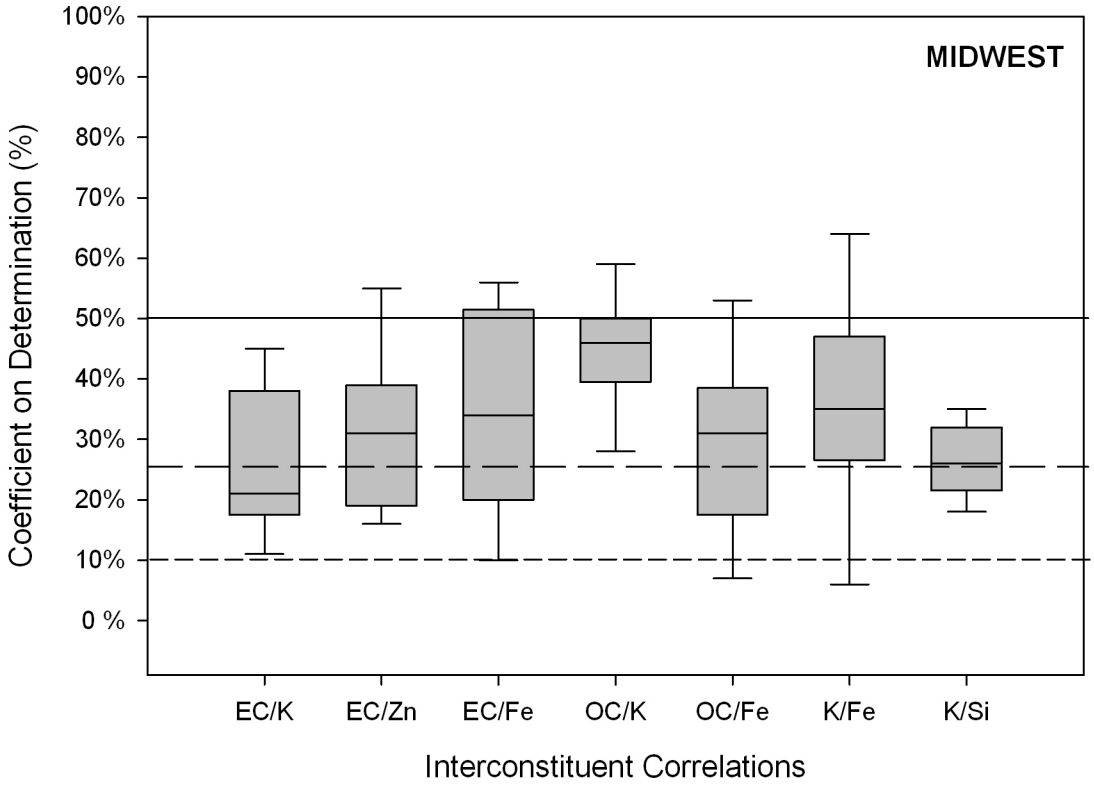

Figure 8-4. Distributions of monitor-specific of coefficients of determination for constituents between-source categories in Midwestern cities for all days: 49 STN monitors, 2001–2005.

Northeast

Moderate correlations between elemental carbon and zinc, organic carbon and iron, and organic carbon and potassium were observed in all Northeastern cities. Elemental carbon was also moderately correlated with iron except in Burlington, VT. Over this region, elemental carbon was moderately correlated with nickel; however, the average correlation was mainly driven by New York City, NY and Philadelphia, PA. Moderate correlations with iron were also observed with titanium, sulfate, nickel, silicon, and zinc. Iron was moderately correlated with titanium in all Northeastern cities except Burlington, VT whereas the moderate correlations observed between iron and sulfate were driven by New York City, NY, Rockingham, NH, and Springfield, MA. Iron and silicon were moderately correlated in most cities in this region with the exception of Edison, NJ and Springfield, MA.

The moderate correlations observed between nickel and zinc, and organic carbon and sulfate were heavily influenced by the strong correlations in New York City, NY (R^2 = .59). Overall, nitrate had moderate correlations with zinc and sulfate, but was poorly correlated in Edison, NJ, Philadelphia, PA, and Pittsburgh, PA. Potassium was moderately correlated with zinc in all Northeastern cities except Boston, MA. Seasonally, correlations of elemental carbon, iron, and zinc with nickel were only observed in the winter. The overall moderate correlations observed between potassium and zinc also were driven by the winter months. Significant correlations between organic carbon and iron, organic carbon and sulfate, and nitrate and zinc were seen during the summer but not during the winter. Only weak correlations were observed for the other inter-constituent correlation combinations.

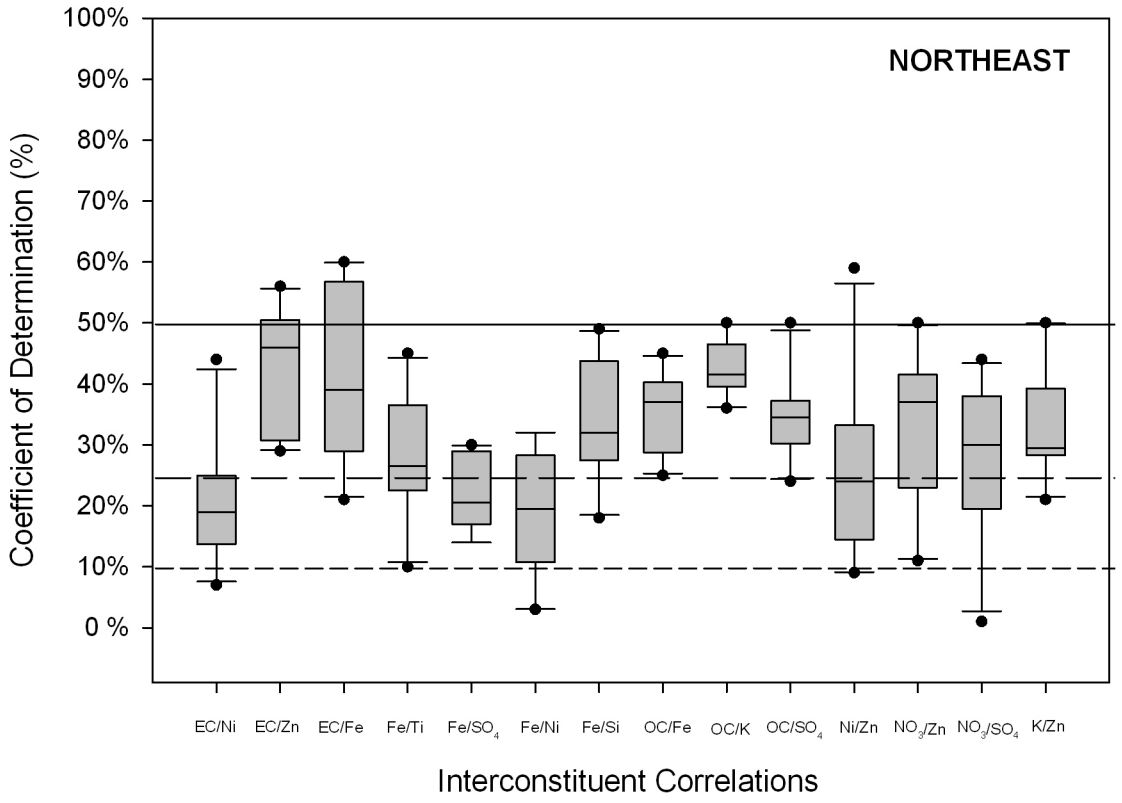

Figure 8-5. Distributions of monitor-specific of coefficients of determination for constituents between-source categories in Northeastern cities for all days: 49 STN monitors, 2001–2005.

South

Moderate correlations were observed between elemental carbon and iron and titanium. Strong correlations between these constituents were observed in Atlanta, GA. Iron was also moderately correlated with silicon except in Baltimore, MD and Memphis, TN. The overall moderate correlations observed between iron and zinc were driven by the strong correlation seen in Atlanta, GA, Baltimore, MD, Birmingham, AL, and Washington, DC.

For the region zinc was moderately correlated with organic carbon, with strong correlations in Houston, TX. Moderate correlations between elemental carbon and iron and iron and potassium were observed during the winter but not during the summer. In contrast, significant correlations between iron and silicon were seen during the summer and not the winter. Only weak correlations were observed for the other inter-constituent correlation combinations.

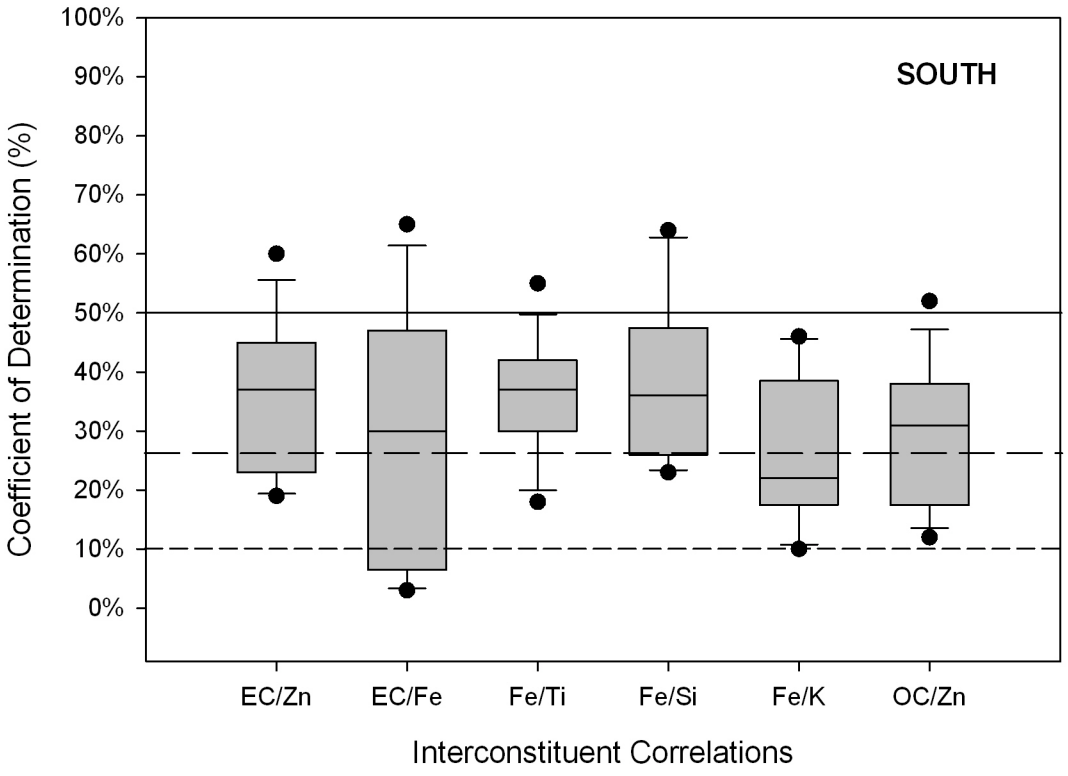

Figure 8-6. Distributions of monitor-specific of coefficients of determination for constituents between-source categories in Southern cities for all days: 49 STN monitors, 2001–2005.

West

As with the other regions, elemental carbon was moderately correlated with iron and zinc. The overall moderate correlations between elemental carbon and iron were heavily influenced by the strong correlations observed in Denver, CO, Riverside, CA, and Seattle, WA. Strong-to-moderate correlations between elemental carbon and zinc were observed in all western cities as well as between iron and silicon, and iron and titanium. Iron was also moderately correlated with aluminum and organic carbon. Overall iron was moderately correlated with potassium, with strong correlations in Boise City, ID, Denver, CO, and Phoenix, AZ.

Moderate correlations were observed between potassium and organic carbon, with the strongest correlation seen along the west coast cities. The overall moderate correlations between potassium and zinc were mainly driven by a few cities: Fresno, CA; Missoula, MT; Portland, OR; and Sacramento, CA. As with the other regions the correlation between elemental carbon and zinc were driven by the winter months. Stronger correlations between iron and aluminum, organic carbon and iron, and potassium and zinc were observed in the winter compared to the summer. Only weak correlations were observed for the other inter-constituent correlation combinations.

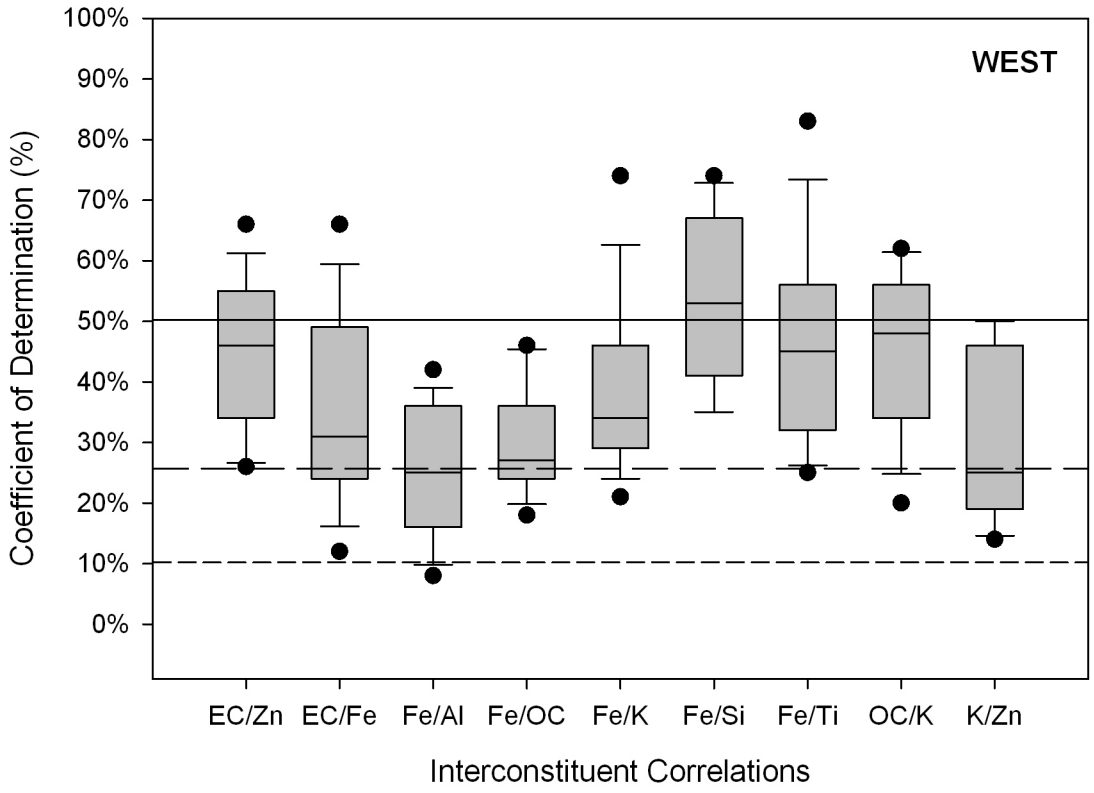

Figure 8-7. Distributions of monitor-specific of coefficients of determination for constituents between-source categories in Western cities for all days: 49 STN monitors, 2001–2005.

Conclusions Regarding Interconstituent Correlations

A common approach for estimating the total health effect of multiple pollutant exposures is to use the indicator approach where the concentration of one pollutant is used to represent the combined exposure to several pollutants or to an emissions source (Dominici et al. 2010). In the first section of this chapter, the strength of the intercorrelations among constituents assigned to the same source category was examined. Selenium, a constituent of coal combustion, is not well correlated with the other constituents typically associated with fossil fuel combustion. For mobile sources and crustal materials, elemental carbon and silicon could serve as indicators, respectively. Potential indicators of residual oil fly ash, biomass combustion, and sea spray/road salt appear to be geographically and seasonal dependent. The correlations between the metallic elements appear to be source dependent, varying based on either manufacturing or dusts from mobile sources.

The second section of this chapter focused on between-source categories. Previous studies have identified associations between several $PM_{2.5}$ constituents and adverse health effects. In metropolitan areas where these constituents are highly correlated, observational studies will have difficulties in determining the causal constituent. More significant interconstituent correlations were seen in Northeastern cities compared to the other regions. Across all regions strong-to-moderate correlations were observed between elemental carbon and zinc, and elemental carbon and iron. Significant correlations were seen between organic carbon and potassium in the Midwest, Northeast, and West. Iron and potassium had strong-to-moderate correlations in all regions except the Northeast. In Midwestern and Northeastern cities, iron was strongly-to-moderately correlated with organic carbon. Iron was also strongly-to-moderately correlated with both titanium and silicon on all regions except the Midwest.

Chapter 9
Relationships Between Coefficients of Variation and Correlations

Low correlation between a pair of air pollutants may be caused by a lack of temporal variability in one or both pollutants rather than simply an absence of a relationship. The purpose of this chapter is to determine whether low correlations between pairs of pollutants were due to low coefficient of variations of either pollutant. Only a few pollutants had monitor-specific coefficients of variations less than 50 percent. These include $PM_{10,}$ ozone, carbon monoxide, nitrogen dioxide, sulfur dioxide, selenium, sulfate, elemental carbon, and organic carbon.

Table 9-1 lists those cities with both correlations between the pollutant and $PM_{2.5}$ in lowest 10^{th} percentile and coefficient of variations less than 50 percent for either pollutant. PM_{10} was strong-to-moderately correlated with $PM_{2.5}$ in all of the cities therefore it was not included. For the majority of these cities the low correlations between the pollutants and $PM_{2.5}$ may have been caused by the low coefficients of variation of that pollutant. However, in Baton Rouge, LA, Charlotte, NC, and Dallas, TX low coefficients of variation for $PM_{2.5}$ were also observed.

Low Correlations with $PM_{2.5}$

Table 9-1. Cities with $PM_{2.5}$ Correlations in the Lowest 10^{th} Percentile and Coefficients of Variations Less Than 50 Percent for Either $PM_{2.5}$ or the Other Pollutant

O_3	CO	NO_2	SO_2	Se	SO_4	EC	OC
Portland, OR	Omaha, NE Houston, TX	Charlotte, NC Dallas, TX	San Jose, CA Miami, FL	San Diego, CA Reno, NV Oxnard, CA Kansas City, MO	Phoenix, AZ	Baton Rouge, LA Charlotte, NC	Omaha, NE

Low Interconstituent Correlations

Those pollutants with high interconstituent correlations in all cities were not further examined since these relationships are unlikely to have been greatly affected by low coefficient of variations. In the previous section those cities were presented in which both the correlations between the pollutants were in the lowest 10th percentile and the coefficients of variation were less than 50 percent for either pollutant. Elemental carbon was poorly correlated with organic carbon, potassium, and zinc in Baton Rouge, LA possibly due to low coefficients of variation of elemental carbon. Other poor correlations potentially due to low coefficients of variation of elemental carbon include correlation between elemental carbon and zinc in Detroit, MI and between elemental carbon and vanadium in Tulsa, OK. In Phoenix, AZ, we observed low coefficients of variation for sulfate possibly leading to weak correlations between sulfate and vanadium and sulfate and organic carbon. For other correlations with organic carbon, Table 9-2 list those cities with both correlations between the pollutants lowest 10th percentile and coefficient of variations less than 50 percent for either pollutant. In all cities weak correlations between organic carbon and the other constituent may have been caused by a low coefficient of variation of organic carbon.

Table 9-2. Cities with Between Pollutant Correlations in the Lowest 10^{th} Percentile and Coefficients of Variations Less Than 50 Percent for Either $PM_{2.5}$ Constituent

Cu	K	Zn	Cl	NO_3	Fe	Ni	Si	V
Omaha, NE Tulsa, OK	Miami, FL Tampa, FL Memphis, TN	Omaha, NE Charleston, SC	St. Louis, MO Omaha, NE Memphis, TN	Indianapolis, ID Kansas City, MO St. Louis, MO Tulsa, OK Memphis, TN	Tampa, FL Charleston, SC	Miami, FL St Louis, MO Kansas City, MO Memphis, TN	Miami, FL Tampa, FL	Tampa, FL Indianapolis, IN Minneapolis, MN Tulsa, OK

Conclusions Regarding Relationships Between Coefficients of Variations and Correlations

Only $PM_{10,}$ ozone, carbon monoxide, nitrogen dioxide, sulfur dioxide, selenium, sulfate, elemental carbon and organic carbon had monitor-specific coefficients of variations less than 50 percent. Low correlations with $PM_{2.5}$ due to low coefficients of variations were only possible in a handful of cities. Many of these cities were also identified when examining interconstituent correlations. Cities that were listed for multiple pollutants include Omaha, NE, Miami, FL, Tampa, FL, Charlotte, NC, Charleston, SC, Tulsa, OK, Memphis, TN, St Louis, MO; Kansas City, MO, and Indianapolis, IN.

Chapter 10
Potential Monitors for Increased Frequency

Summary of Results

This analysis illustrates that criteria pollutants and $PM_{2.5}$ species concentrations, variability, and relationships with $PM_{2.5}$ can differ both temporally and spatially. For those pollutants exhibiting a geographic pattern, the highest concentrations were observed in the Northeast (S, SO_4, and SO_2), California (NO_3), or both ($PM_{2.5}$ and V). Examining all of the selected sites, PM_{10}, O_3, Ca, K, Zn, V, Al, Si, Ti, Na, and SO_4 were significantly higher in the summer compared to the winter, whereas the opposite was true for CO, NO_2, SO_2, Br, NO_3, EC, OC, Mn, Ni, and Cl. A few of the pollutants ($PM_{2.5}$, PM_{10}, OC, V, and NO_3) also exhibited seasonal patterns that differed regionally.

In comparison with the $PM_{2.5}$ NAAQS for all cities, the majority of days were below the daily standard of 35 $\mu g/m^3$ while a few cities had annual concentrations either well above or well below the annual standard of 15 $\mu g/m^3$. Additionally, in approximately half of the cities, coefficients of variation of less than 49 percent for PM_{10}, Se, CO, NO_2, and OC were observed whereas only a few are below 49 percent for $PM_{2.5}$, EC, NO_3, SO_4, and SO_2.

Regardless of season or region PM_{10} and SO_4 were strong-to-moderately correlated with $PM_{2.5}$ while Se showed weak correlations and SO_2 exhibited both seasonal and regional patterns. The correlations between $PM_{2.5}$ and O_3 were negative in the winter and positive in the summer, and showed some regional differences. Strong correlations with markers of wood smoke (K, Br, and Ca) were seen in some cities, usually in the winter in the Western part of the United States. Elements associated with mobile sources were well correlated with $PM_{2.5}$ in most cities, with the exception of Cu, which exhibited different geographic and seasonal patterns. Of the crustal elements (Al, Fe, Si, and Ti) only Fe was moderately correlated with $PM_{2.5}$ in the majority of cities, with the strongest correlations observed in the winter. Finally, markers of residual-oil fly ash and sea spray were at best moderately correlated with $PM_{2.5}$ in a small percentage of cities.

Next, correlations within and between selected source groups were examined. Although used as a marker of coal combustion, selenium was not well correlated with the other fossil fuel constituents (i.e., sulfur and sulfate). For mobile sources, elemental carbon and organic were moderately correlated and correlations with nitrate were stronger in the winter. Correlations between nickel and vanadium were only significant in a few cities with the strongest correlations observed in the winter. Similarly, potassium and bromine (markers for biomass combustion) were also more highly correlated in the winter. Correlations between all of the crustal materials were higher in the summer than during the winter. Silicon was significantly correlated with other crustal materials in the majority of the cities whereas correlations between aluminum and the other crustal elements were generally weak. Elements used as markers for seas spray/road salt were not well correlated and correlations between the metallic elements appear to be source dependent, varying based on either manufacturing or dusts from mobile sources.

To examine between source groups, elements that have previously been associated with adverse health effects were selected. Strong-to-moderate correlations were observed across all regions between elemental carbon and zinc, and elemental carbon and iron. Iron was also correlated with elements associated with mobile sources, crustal materials, and biomass combustion in all regions. Correlations such as those between elemental carbon and zinc in the Northeast were mainly driven by a few cities (e.g., New York City, NY) while others such as iron and silicon were present in all cities across a region.

Some of the observed low correlations between pollutants in certain cities may have been due to a low coefficient of variation for either one or both pollutants. Cities that appeared frequently were Omaha, NE; Miami, FL; Tampa, FL; Charlotte, NC; Charleston, SC; Tulsa, OK; Memphis, TN; St Louis, MO; Kansas City, MO; and Indianapolis, IN.

Selection of Cities

An increasing number of studies are examining the associations between gaseous pollutants and PM species and health effects, as well as their role as potential confounders in the PM-health effects associations. A meta-analysis of PM and gaseous pollutants showed that PM, NO_2, CO, and SO_2, all showed a positive and significant mortality risk estimate (Stieb et al. 2002). Among the pollutants from the Speciation Trends Network (STN), the strongest associations with mortality have been observed for $PM_{2.5}$ mass, EC, NO_3, Cl, Cu, Fe, K, Ti, V, and Zn (Ostro et al. 2007). Studies have also shown that certain chemical species significantly modify the association between $PM_{2.5}$ and mortality (Franklin and Schwartz 2008), while others have examined the role of gaseous pollutants such as O_3 as either potential confounders or surrogates to $PM_{2.5}$ exposures (Sarnat et al. 2001).

Although these studies have identified serious health risks, there is still uncertainty as to which components are the most harmful. Furthermore, the pollutant itself may not be the causal agent but rather a surrogate for a particular source. For example, previous time-series analyses indicate that, of the sources of $PM_{2.5}$, motor vehicle exhaust usually has the strongest associations with all non-accidental mortality and with cardiovascular mortality (Mar et al. 2000; Laden et al. 2000; Janssen et al. 2002).

Various studies have documented significant heterogeneity among community-specific health effect estimates of PM_{10} on mortality (Peng et al. 2005; Dominici et al. 2003), $PM_{2.5}$ and mortality (Franklin et al. 2007), and $PM_{2.5}$ on hospital admissions (Bell et al. 2006). Heterogeneity in city-specific and seasonal-specific estimates have also been demonstrated in ozone mortality studies (Bell et al. 2005; Bell et al. 2004; Levy et al. 2005; Ito et al. 2005). Differences in city-specific health effect estimates may reflect variations in the air pollution mixture and its sources. These variations may be city-specific, adding to the complexity of focusing on a particular pollutant.

Daily monitoring data would aid an epidemiologist in determining the health effect of a particular pollutant. The criteria for selecting cities for daily speciation monitoring includes sufficient population size, sufficient concentration levels and variability, and low correlations between pollutants independent of low variability.

Using the analyses of air quality measurements from 49 STN monitors for 2001-2005 of the previous chapters, daily monitoring would be of greatest epidemiologic interest for metropolitan areas that meet the following five criteria:

1. Population of the metropolitan area;
2. Mean levels of criteria air pollutants;
3. Variation in levels of criteria air pollutants and particulate matter constituents;
4. Correlation among criteria air pollutants and particulate matter constituents; and
5. Relationship of correlations to the coefficient of variation.

In Table 10-1, those cities that do not meet each criteria are indicated by a bar shaded in gray.

Criterion 1: Adequate Population Levels

In Table 10-1, those cities with populations less than 500,000 and greater than 5,000,000 are highlighted. Cities with smaller populations may not have enough daily deaths to perform a time-series analysis. Conversely, very large cities will have an adequate number of daily deaths but the monitor may not represent well the population's exposure. City-specific populations can be found on Table 4-1.

Criterion 2: Mean Levels of $PM_{2.5}$

The city-specific daily and/or annual $PM_{2.5}$ in comparison to NAAQS was examined. Concentrations were deemed too high if more than 10 percent of days were above the daily standard ($35\mu g/m^3$) and/or the 5-year annual average was more than 10 percent above the annual standard ($15\mu g/m^3$). Cities were identified as having too low $PM_{2.5}$ concentrations when 10 percent or fewer days above 17.5 $\mu g/m^3$ (50% of the daily standard) and/or 5-year annual average concentrations below $10\mu g/m^3$ (75% of the annual standard). These concentrations are presented in Figures 5-1 and 5-2.

Criterion 3: Adequate Level of Variability

Only 10 pollutants ($PM_{2.5}$, PM_{10}, CO, NO_2, O_3, SO_2, Se, SO_4, EC, and OC) had coefficients of variation less than 50 percent in any of the cities. NO_2, O_3, and Se had coefficients of variations less than 50 percent in most cities so it would be difficult to eliminate a city based on one low coefficient of variation. Therefore, those cities with coefficients of variation less than 50 percent for five or more pollutants were identified as having inadequate levels of variation. City- and pollutant-specific coefficients of variation are presented in Tables A-4 and A-7 of the Appendix.

Criterion 4: Correlation Among Criteria Air Pollutants and Particulate Matter Constituents

Ideally in a selected city correlations between $PM_{2.5}$ and constituents and the other criteria pollutants would be strong. In contrast, correlations among the particulate matter constituents should be low. Some of pollutants (e.g., selenium and chlorine) will have low correlations with $PM_{2.5}$ in the majority of cities or be highly correlated (e.g., elemental carbon and organic carbon) in the majority of cities. Therefore the following exclusion criteria were developed: Strong correlations ($R^2 > 0.49$) with $PM_{2.5}$ must be present with over 50 percent of the other pollutants; and $R^2 > 0.49$ can only occur between less than 10 percent of the potential constituent combinations described in Chapter 8. These coefficients of determination are shown in Tables A-8 through A-17 of the Appendix.

Criterion 5: Relationship of Correlations to the Coefficient of Variation

As described in Chapter 9, low correlations between pollutants may be caused by low coefficients of variation of either pollutant. In Table 10-1, those cities where low correlations could be due to low variability are indicated by bars shaded in gray.

Cities Meeting Criteria 1-5

According to Table 10-1, the 11 metropolitan areas meeting the above criteria are:

Atlanta, GA	Baltimore, MD
Boston, MA	Springfield, MA
Edison, NJ	Newark, NJ
Cleveland, OH	Pittsburgh, PA
Providence, RI	Salt Lake City, UT
Milwaukee, WI	

Table 10-1. Selection Criteria for Identifying Metropolitan Areas for Enhanced PM Speciation Monitoring, 49 STN Monitors, 2001-2005

City	Too High/Low Population Size	Too High/Low $PM_{2.5}$ Levels	Coefficient of Variation < 50%	$R^2 < 0.49$ between $PM_{2.5}$ and other pollutants	$R^2 > 0.49$ between constituents	Low R^2 due to low coefficient of variation
Birmingham, AL		High				
Phoenix, AZ		Low				
Bakersfield, CA		High				
Fresno, CA		High				
Oxnard, CA						
Riverside, CA		High				
Sacramento, CA						
San Diego, CA						
San Jose, CA						
Denver, CO		Low				
Washington, DC						
Miami, FL		Low				
Tampa, FL		Low				
Atlanta, GA						
Boise City, ID		Low				
Chicago, IL						
Indianapolis, IN						
Baton Rouge, LA						
Baltimore, MD						
Boston, MA						
Springfield, MA						
Detroit, MI						
Minneapolis, MN						
Gulfport, MS						

Table 10-1. Continued

City	Population Size	Too High/Low $PM_{2.5}$ Levels	Coefficient of Variation < 50%	R^2 < 0.49 between $PM_{2.5}$ and other pollutants	R^2 > 0.49 between constituents	Low R^2 due to low coefficient of variation
Kansas City, MO						
St. Louis, MO						
Missoula, MT						
Omaha, NE						
Reno, NV		Low				
Rockingham, NH		Low				
Edison, NJ						
Newark, NJ						
New York, NY						
Charlotte, NC						
Cleveland, OH						
Tulsa, OK						
Portland, OR		Low				
Philadelphia, PA						
Pittsburgh, PA						
Providence, RI						
Charleston, SC						
Memphis, TN						
Dallas, TX						
El Paso, TX		Low				
Houston, TX		Low				
Salt Lake City, UT						
Burlington, VT		Low				
Seattle, WA		Low				
Milwaukee, WI						

Geographical Distribution

An additional consideration not previously discussed is the geographical distribution of selected cities. The above list of cities does not represent well the Western part of the United States while Northeastern cities appear to be overly represented. In the West, San Diego, CA and Sacramento, CA met four out of five criteria and was excluded only by the low correlation between $PM_{2.5}$ and selenium possibly due to the minor contribution of coal combustion. In the Northeast, Springfield, MA and Edison, NJ may be dropped in preference to their larger neighbors.

Conclusion

In this report we developed a methodology for selecting cities for daily speciation monitoring. As an illustration of this methodology we applied the 5 design criteria to the ambient air quality data from 2001-2005 of 49 meteropolitan areas. Based on these results and in combination with consideration to geographical distribution we generated the following list of potential candidatemetropolitan areas for enhanced air quality monitoring;

San Diego, CA	Atlanta, GA
Baltimore, MD	Boston, MA
Newark, NJ	Cleveland, OH
Pittsburgh, PA	Providence, RI
Salt Lake City, UT	Milwaukee, WI

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