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Spatial and temporal variability of fine particle composition and source types in five cities of Connecticut and Massachusetts

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Abstract

To protect public health from PM_{2.5} air pollution, it is critical to identify the source types of PM_{2.5} mass and chemical components associated with higher risks of adverse health outcomes. Source apportionment modeling using Positive Matrix Factorization (PMF), was used to identify PM_2 . source types and quantify the source contributions to $PM_{2.5}$ in five cities of Connecticut and Massachusetts. Spatial and temporal variability of $PM_{2.5}$ mass, components and source contributions were investigated. PMF analysis identified five source types: regional pollution as traced by sulfur, motor vehicle, road dust, oil combustion and sea salt. The sulfur-related regional pollution and traffic source type were major contributors to $PM_{2.5}$. Due to sparse ground-level $PM_{2.5}$ monitoring sites, current epidemiological studies are susceptible to exposure measurement errors. The higher correlations in concentrations and source contributions between different locations suggest less spatial variability, resulting in less exposure measurement errors. When concentrations and/or contributions were compared to regional averages, correlations were generally higher than between-site correlations. This suggests that for assigning exposures for health effects studies, using regional average concentrations or contributions from several PM_{2.5} monitors is more reliable than using data from the nearest central monitor.

Keywords

 $PM_{2.5}$; source apportionment; Positive Matrix Factorization (PMF); spatial variability; temporal variability; exposure measurement errors

1. Introduction

Fine particles (particles of an aerodynamic diameter ≤ 2.5 µm; PM_{2.5}) originate from local and regional anthropogenic and natural sources. The adverse respiratory health effects of PM2.5 mass in children have been investigated by numerous epidemiological studies (Gent

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et al. 2003; Delfino et al. 2004; Lewis et al. 2005). In addition, several studies have linked health effects to individual particle components (Franklin et al. 2008; Ostro et al. 2009; Gent et al. 2009). For the purpose of establishing regulatory standards, it is important to identify source types of $PM_{2.5}$ mass and chemical components that may pose risks to public health. Source apportionment modeling techniques have been used to link a variety of health outcomes to specific source types (Ozkaynak and Thurston, 1987; Laden et al. 2000; Mar et al. 2000; Gent et al. 2009).

 $PM_{2.5}$ mass and component concentrations as well as $PM_{2.5}$ source contributions can vary spatially (Kim et al. 2005). However, because of the sparsity of ground-level $PM_{2.5}$ monitoring sites, measurements from a central monitoring site have been often used in epidemiological studies. This approach may not be appropriate considering the potential spatial heterogeneity of the source contributions. If concentrations and/or source contributions are not highly correlated between different locations, the resulting exposure misclassification is likely to introduce bias in the health effects risk estimates. Therefore, it is critical to examine the spatial variability of the $PM_{2.5}$ mass concentrations, composition, and the source contributions on a regional scale in order to investigate the potential exposure measurement error and to assess the plausibility of using a central monitoring site to determine the temporal variability of exposures within a region.

The objectives of our study are to identify $PM_{2.5}$ source types and quantify their contributions using data collected at five U.S. Environmental Protection Agency (EPA) monitoring sites: four in Connecticut and one in Massachusetts. To the best of our knowledge, no studies on the spatial and temporal variability of $PM_{2.5}$ mass, components, and source contributions have been conducted in the study region.

2. Methods

2.1. Sampling and chemical analysis of fine particle filters

 $PM_{2.5}$ samples were collected on Teflon filters at five EPA monitoring sites located in Bridgeport, Danbury, Hartford, and New Haven, CT and Springfield, MA between August, 2000 and February, 2004 (U.S. EPA 2010). Daily 24-hr (midnight to midnight) PM_{2.5} integrated samples were collected in Hartford (1,206 samples) and New Haven (1,181 samples) during the monitoring period. 24-hr $PM_{2.5}$ integrated samples were collected every third day in Bridgeport (418 samples) and Danbury (386 samples). Daily 24-hr sampling was performed in Springfield (744 samples) with intermittent missing periods. Available data from alternate sites near the originally selected sites were used for days when data were missing: i.e., 3.3% of the Bridgeport samples; 3.0% of the Hartford samples; 1.9% of the New Haven samples; and 20.3% of the Springfield samples. All the locations of the primary and alternate sites are presented in Figure 1.

The filter samples obtained from the Connecticut and Massachusetts Departments of Environmental Protection were analyzed using X-ray fluorescence (XRF) for 51 elements (Desert Research Institute, Reno, NV) and optical reflectance technique for elemental carbon (EC) (Harvard University). The analytical uncertainty for each elemental concentration was determined. The minimum detection limit (MDL) for the 51 elements was defined as 3 times the analytical uncertainty. The MDL for EC was set as 3 times the standard deviation of the values from the optical reflectance analysis of field blanks (Gent et al. 2009). We excluded elements from the source apportionment modeling with more than 90% of the samples below the MDL unless they were important tracer elements for potential source types. Although only a small fraction of samples are above the tracer element MDL, these samples can provide information about the maximum daily impact of the corresponding source type at the receptor. However, we did not include tracer elements if all

the samples were below the MDL. In our study, the remaining elements for the source apportionment analysis were not very sensitive to the selected threshold for exclusion.

2.2. Positive matrix factorization (PMF)

PMF is based on a multivariate factor analysis and results in factor contributions and factor profiles (Paatero and Tapper 1994). In PMF, the matrix $X(n \times m)$ includes measured mass concentrations, and is represented as the sum of the product of G ($n \times p$) and F ($p \times m$) matrices and the residual matrix $E(n \times m)$, where *n* is the number of samples, *m* is the number of chemical species, and *p* is the number of independent source types.

$$
X{=}GF{+}E
$$

The object function Q that is to be minimized is defined as

$$
Q=\sum_{i=1}^n\sum_{j=1}^m(\varepsilon_{ij}/u_{ij})^2
$$

where u_{ij} is the uncertainty of the species j in a sample i. In addition to minimizing the object function *Q*, non-negativity constraints need to be met, meaning that all the elements in *G* and *F* are to be non-negative (Paatero and Tapper 1994; Paatero 1997).

In our PMF analysis, the concentration and sample-specific uncertainty fields were entered as reported by analytical laboratories, and those days with at least one missing value were excluded from the analysis. We included the $PM₂$ mass concentrations as a total variable (weak species), and all the species were categorized as strong (signal-to-noise ratio≥2), weak (0.2≤signal-to-noise ratio<2), or bad (signal-to-noise ratio<0.2) species (Paatero and Hopke 2003). The weak species were downweighted by tripling the uncertainty, and the bad species were excluded from the analysis. PMF analyses were conducted using the robust mode to mitigate the effects of extreme values on the PMF solution (Paatero 1997). However, there were recognized extreme values from fireworks and the 2002 Quebec forest fires during our study period, and we excluded those samples to completely prevent them from affecting the PMF solutions. In addition, we examined the concentration scatter plots and time-series plots of all analyzed species to check for extreme values. After the PMF runs, we selected converged solutions with the minimal Q value in the robust mode for each run with the different number of factors, ranging from 3 to 10, and compared the Q values with those in the non-robust mode to make sure that any remaining extreme values did not excessively affect the model fit. A PMF solution was determined as the most physically reasonable and interpretable one among all PMF solutions. We named the source types based on the association of a given factor with tracer elements and considered additional information regarding the factor contributions such as seasonality and day of week variation. The PMF solution has rotational ambiguity since non-negativity constraints generally do not guarantee a unique solution (Paatero et al. 2002). To further reduce the rotational ambiguity in a solution and find a final PMF solution, the parameter FPEAK can be used, and the resulting rotations from the parameter FPEAK runs can be examined in a G space plot, which is a graphical procedure showing a distribution between two different factor contributions (Paatero et al. 2005). In our study, the parameter FPEAK values from −2 to 2 were explored.

The goodness of fit for the PMF model was assessed by comparing the predicted concentrations with the measured concentrations based on % mean relative error (MRE) defined as $100 \times$ | (predicted concentrations) – (measured concentrations) | / (measured concentrations). Model fit was also examined on a scatter plot of predicted by measured concentrations and coefficient of determination (R^2) was calculated.

2.3. Analyses of fine particle spatial and temporal variability

To make comparisons between sites a dataset was created that included days during which measurements were available for all five sites. The inter-site relationships of the $PM_{2.5}$ mass and elemental concentrations as well as source contributions were assessed using Spearman correlation coefficients for non-normally distributed data to minimize the influence of outliers. Relationships between individual sites and regional average concentrations (defined as the mean daily concentrations from four sites excluding the fifth to be compared) were also examined.

For the analyses of seasonal differences, each year was separated into warm season (April to October) and cold season (November to March). Days of the week were categorized into weekdays (Monday to Friday) and weekends (Saturday and Sunday). Finally, two-sample ttest and one-way analysis of variance (ANOVA) were used to examine differences between seasons, day of week and sites (alpha set to 0.05).

3. Results and Discussion

3.1. PM2.5 concentrations

Mean $PM_{2.5}$ mass concentrations are summarized in Table 1 by site and season. Mean (SE) PM_{2.5} mass concentrations ranged from 11.9 (0.2) μ g/m³ in Hartford to 17.0 (0.3) μ g/m³ in New Haven. Mean $PM_{2.5}$ levels in Bridgeport, Danbury, and Springfield were not significantly different (F-value=0.33 and $p=0.719$). PM_{2.5} concentrations in the cold season were higher than those observed during the warm season in all sites except Bridgeport. Mean $PM_{2.5}$ concentrations in the cold and warm seasons are shown in Table 1. $PM_{2.5}$ concentrations between the two seasons were significantly different only in New Haven (tvalue=4.07 and $p<0.0001$).

PMF analysis identified five source types: regional pollution (as traced by sulfur), motor vehicle, road dust, oil combustion and sea salt. The concentrations of chemical components used for PMF analysis are presented in Table 2, and the source contributions resolved are shown in Tables 3 and A. Although source apportionment analysis was conducted for each site separately, the same five source types were identified for all sites. The sulfur-related pollution was the major contributor to the PM_{2.5} mass concentrations followed by the traffic source types. Sea salt contributed the least. The contributions of the sulfur-related pollution, motor vehicle and sea salt were similar at all sites, whereas the contributions of road dust and oil combustion varied by site. It is noted that an additional PMF analysis was performed for Springfield after excluding the samples from an alternate site of Chicopee (approximately 20%) to make sure that the replacement did not bias the results. The source contributions without the samples from the Chicopee site were comparable to those with the samples from both the primary and alternate sites. Because only a relatively small amount of data from alternate sites for Bridgeport, Hartford, and New Haven were used and the distance from the primary sites to their alternate sites was reasonably short (i.e., 2.5 km on average), it was unlikely that use of the alternate sites would have significant effects on the results. Therefore, all the subsequent results for the Bridgeport, Hartford, New Haven, and Springfield sites are based on the samples collected from both the primary and alternate sites.

The regional sulfur-related pollution contributed to $PM_{2.5}$ mass from 4.7 μ g/m³ (39%) in Springfield to 7.1 μ g/m³ (42%) in New Haven. These contributions displayed a strong seasonal pattern with significantly higher contributions in warm season at all sites (tvalue=6.24, p<0.0001 in Bridgeport; t-value=5.64, p<0.0001 in Danbury; t-value=10.17, p<0.0001 in Hartford; t-value=10.60, p<0.0001 in New Haven; t-value=6.99, p<0.0001 in Springfield). Atmospheric photochemical activity is greater during the warm season, thus a larger fraction of emitted sulfur dioxide is oxidized to sulfate. No significant differences were found between weekdays and weekends in sulfur source contributions (Table 4). The element Na appears to be associated with the regional sulfur-related pollution, and the following may explain the association between them. Acid sulfate particles collected on the Teflon filter such as H_2SO_4 and NH_4HSO_4 can react with NaCl also collected on the same filter, producing $Na₂SO₄$ and HCl. Because HCl is a gas, it can escape from the filter, and only $Na₂SO₄$ may be measured. In addition, air masses carrying emissions from coal-fired power plants can be transported over the Atlantic Ocean before they reach the receptors. NaCl and sulfate can be collected on the Teflon filter and produce the discussed reaction above.

The motor vehicle source type was characterized by its high loadings of EC, Zn, Pb, Cu, and Br. Its contribution to PM_{2.5} concentrations varied by site from 3.1 μ g/m³ (28%; Hartford) to 5.0 μg/m³ (30%; New Haven). During the cold season, source contributions varied between 4.3 μ g/m³ (37%; Hartford) and 7.6 μ g/m³ (42%; New Haven). In the warm season, they ranged from 2.1 μ g/m³ (17%; Danbury) to 3.1 μ g/m³ (19%; New Haven). These contributions depend in part on the local traffic volume and vehicle type (e.g., heavy trucks/ buses or light trucks/passenger cars). High traffic volume congestion can increase the amount of "creep-phased" (e.g., stop and go) vehicles on the road and result in more particles per vehicle than steadily operating vehicles (Shah et al. 2004). Heavy-duty trucks and buses primarily use diesel fuel and generally emit more particles than light-duty trucks and passenger cars using gasoline. A higher proportion of heavy-duty trucks and buses in traffic is likely to cause higher source contributions of motor vehicle, and the proximity to industrial areas may be reflected in the proportion. The contributions of motor vehicles exhibited seasonal variability and were significantly higher during the cold season at all sites (t-value=5.90, p<0.0001 in Bridgeport; t-value=6.21, p<0.0001 in Danbury; t-value=10.85, p<0.0001 in Hartford; t-value=14.72, p<0.0001 in New Haven; t-value=9.37, p<0.0001 in Springfield). Lower height of boundary mixing layer and more stable air conditions in cold season account for this difference. In addition, incomplete combustion during cold starts results in an increased particle formation due to incremented nucleation of vehicular exhaust during the cold season (Grieshop et al. 2006; Kuhn et al. 2005). Motor vehicle source contributions varied by the day of the week. For all the locations, source contributions were significantly higher on the weekdays compared to weekends (t-value= 3.77 , p= 0.0002 in Bridgeport; t-value=2.91, p=0.0039 in Danbury; t-value=5.88, p<0.0001 in Hartford; tvalue=6.99, p<0.0001 in New Haven; t-value=3.78, p=0.0002 in Springfield). Traffic volume is higher during weekdays and our findings suggest that a large fraction of vehicular emissions is of local origin. If a large fraction of vehicular emissions is transported to our study region, the day of week variation is likely to be much less pronounced. This is because the transported vehicular emissions tend to dilute the day of week variation depending on the distance from the source area to our study region and wind direction and speed.

The road dust source type was responsible for a large fraction of Si, Fe, Al, Ca, Ba, and Ti concentrations. The contribution of this source type to the PM_{2.5} mass varied from 0.8 μ g/ m^3 (7%; Hartford) to 2.9 μg/m³ (18%; New Haven). In the warm season, the contributions of road dust ranged from $\frac{0.9 \text{ }\mu\text{g/m}^3}{0.8 \text{ }\mu\text{s}}$; Hartford) to 3.0 $\mu\text{g/m}^3$ (19%; New Haven), and between 0.6 μ g/m³ (4%; Hartford) and 2.8 μ g/m³ (16%; New Haven) in the cold season. Resuspended road dust contains a mixture of soil particles, abraded asphalt, and tire-, brake-,

and engine-wear particles (Rogge et al. 1993). The amount of re-suspended road dust depends on the surface loading, dryness of the road, vehicle speed and weight, and wind speed, all of which vary by site. Source contributions were higher during the warm season in all five sites, but the seasonal differences were statistically significant only for Bridgeport (tvalue=5.82, p<0.0001), Hartford (t-value=5.95, p<0.0001), and Springfield (t-value=4.51, p<0.0001). This seasonal pattern is likely due to more soil particles transported and deposited on the road and less frequent washouts due to the decreased precipitation during the warm season. In addition, frequent snow and its subsequent melting in the cold season may contribute to wetness of the road surface, reducing re-suspension of road dust. Source contributions on weekdays were significantly higher than weekends in Bridgeport (tvalue=3.15, p=0.0018), New Haven (t-value=16.94, p<0.0001), and Springfield (tvalue=5.49, p<0.0001). This source type was found to have significant respiratory health effects in our previous study (Gent et al. 2009).

V and Ni are tracers of oil combustion source type. The contribution of this source type to total PM_{2.5} mass ranged from 0.6 μg/m³ (5%) in Danbury to 2.2 μg/m³ (19%) in Hartford. There are many oil combustion sources such as fuel oil-fired power plants, ships and ferries, and homes and buildings using heating oil. The former two sources use residual oil which is known to emit more particles than the latter which usually uses distillate oil (U.S. EPA, 1998). Fuel oil-fired power plants located along the East Coast affect Northeastern cities downwind of the plants. There is a high demand for distillate oil for home heating in the Northeast (from 1999 to 2003 54% of total demand) compared to the rest of the country (11%) (CT DEP, 2005; EIA, 2003). Many commercial buildings heated by high capacity boilers that use residual oil may be also responsible for the source contributions, emitting more particles than homes. Harbor traffic and airports in Bridgeport, Hartford, and New Haven also contribute to particles measured at the respective monitoring sites. Oil source contributions in all five sites were significantly higher in cold season compared to warm season (t-value=4.36, p<0.0001 in Bridgeport; t-value=4.83, p<0.0001 in Danbury; tvalue=9.64, p<0.0001 in Hartford; t-value=12.77, p<0.0001 in New Haven; t-value=10.72, p<0.0001 in Springfield). It is possibly due to seasonal differences in space heating by oil boilers since emissions from oil-fired power plants, vehicles, ships and ferries are considered to be relatively more uniform throughout the year. However, emissions from ships carrying heating oil for New England might be higher in the cold season, since the New Haven port is known as an oil seaport and the volume of oil transportation is approximately 35% higher during the cold season (EIA, 2008). Day of week variation was significant only for Hartford and Springfield where source contributions were higher on the weekdays (t-value=2.07, p=0.0390; t-value=2.94, p=0.0034, respectively). The higher source contributions on the weekdays for these sites were found to be primarily in the warm season.

Sea salt particles are composed of Cl and Na. The contribution of these particles to PM_{2.5} mass ranged from 0.1 μ g/m³ (1%) in Springfield to 0.4 μ g/m³ (2%) in New Haven. At all five monitoring sites, source contributions were higher during the cold season than in warm season. All differences were statistically significant, except for Bridgeport (t-value=1.66, p=0.098 in Bridgeport; t-value=4.51, p<0.0001 in Danbury; t-value=5.99, p<0.0001 in Hartford; t-value=6.56, p<0.0001 in New Haven; t-value=5.36, p<0.0001 in Springfield). The higher contributions in cold season may be due to the use of sea salt to reduce ice formation on the road and street surfaces (Lee et al. 2003; Gertler et al. 2006). In addition, higher wind speed during the cold season may also increase the airborne sea salt particles. Only Springfield had statistically significant difference in the source contributions between weekdays and weekends (t-value=2.27 and p=0.0236).

Scatter plots showing the relationship between the measured and PMF-predicted PM_2 . concentrations are presented by Figure A. \mathbb{R}^2 coefficients (Figure A) and % MRE (Tables 3 and A) were estimated. The predicted and measured concentrations were in a good agreement.

3.2. Spatial relationships

 $PM_{2.5}$ mass concentration relationships among all five sites were investigated with correlations. The average correlation coefficient was 0.84 (SD=0.05; range=0.75–0.93). Correlation coefficients were determined for the five individual source types. The sulfurrelated pollution showed the highest between-site average correlation $(r=0.81; SD=0.04)$ followed by motor vehicle (r=0.64; SD=0.06), road dust (r=0.60; SD=0.12), oil combustion $(r=0.48; SD=0.23)$, and sea salt $(r=0.44; SD=0.11)$. In terms of between-site correlations for particle components, the concentrations of S ($r=0.87$; SD=0.04), K ($r=0.79$; SD=0.05), and Zn ($r=0.70$; SD=0.08) were highly correlated. In contrast, the Pb ($r=0.33$; SD=0.07) and Mn (r=0.29; SD=0.07) were poorly correlated. Since the study region is impacted by regional sources, pollutants such as S or sulfur-related compounds are expected to be highly correlated (Liu et al. 1996; Suh et al. 1997). The concentrations/contributions from local source types such as traffic (e.g., a large fraction of EC or motor vehicle) are highly correlated because their impacts vary in the same way at each of the sites. For example, the day of week patterns are similar at all sites. Because these sites are not far apart they are impacted by similar meteorological conditions such as atmospheric stability, wind speed and rain that can affect the levels of locally emitted pollutants. Correlations between Bridgeport, Danbury, and Hartford were generally higher than those between New Haven or Springfield and those three sites of Bridgeport, Danbury, and Hartford. Concentrations/contributions in New Haven were less likely to be correlated with the ones at other monitoring sites overall. New Haven monitoring site is impacted by many local sources including two interstates (I-91 and I-95), an active commercial harbor, and an oil-fired power plant. For all sites except for New Haven, the Spearman correlations decrease with between-site distance. The between-site correlations are summarized in Tables 5 and B.

In addition to the correlations between sites, correlations between the regional averages and the respective site concentration were examined (Tables 6 and C). The average correlation coefficient of PM_{2.5} for the five sites was 0.89 (SD=0.05), ranging from 0.83 to 0.94. This high correlation is in agreement with previous studies (Burton et al. 1996). The regional sulfur-related pollution was found to have the highest correlation coefficient $(r=0.87;$ $SD=0.03$) among the five source types, followed by motor vehicle ($r=0.73$; $SD=0.05$), road dust (r=0.68; SD=0.08), oil combustion (r=0.62; SD=0.20), and sea salt (r=0.53; SD=0.08). For many of the chemical components, correlations between the regional averages and site concentrations were high e.g., S (r=0.91; SD=0.03), K (r=0.85; SD=0.04), Zn (r=0.77; SD=0.07), EC (r=0.76; SD=0.09), Si (r=0.74; SD=0.09), and Na (r=0.71; SD=0.10). Low correlations were found for Ba ($r=0.26$; SD=0.06) and Mn ($r=0.28$; SD=0.09). Correlations between regional averages and each site s concentrations were generally higher than the between-site correlations. Variability was lower when the regional averages were used for estimating correlations. This suggests that for exposure assessments and health effects studies, it may be preferable to average concentration data from multiple sites within a study region rather than use data from one monitoring site. Averages of several monitors are less likely to be affected by local sources, making the regional averages more representative exposure estimates.

4. Conclusions

Source types of PM_{2.5} in five cities (four in Connecticut, one in Massachusetts) were identified and quantified using PMF. Although analysis was conducted by individual

monitoring site, all sites in this Northeastern coastal region were impacted by similar source types: sulfur-related pollution, motor vehicle, road dust, oil combustion and sea salt. The sulfur-related pollution and motor vehicle were the major contributors to $PM_{2.5}$ at all five monitoring sites. Among the five cities, Bridgeport, Danbury, and Hartford data were most highly correlated. New Haven site correlation coefficients were generally lower possibly due to the impact of local sources. Correlations varied by component and source type but tended to be high. Correlations between chemical components or source contributions and regional averages were high. This suggests that average concentrations or contributions from several $PM₂$, monitors would be more reliable estimates of exposure for health effects studies compared to estimates from individual air quality monitors.

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Abbreviations

References

- Burton RM, Suh HH, Koutrakis P. Spatial variation in particulate concentrations within metropolitan Philadelphia. Environ Science Tech. 1996; 30:400–407.
- Delfino RJ, Quintana PJE, Floro J, Gastanaga VM, Samimi BS, Kleinman MT, Liu LJS, Bufalino C, Wu CF, McLaren CE. Association of $FEV₁$ in asthmatic children with personal and microenvironmental exposure to airborne particulate matter. Environ Health Perspect. 2004; 112:932–941. [PubMed: 15175185]
- Franklin M, Koutrakis P, Schwartz J. The role of particle composition on the association between PM2.5 and mortality. Epidemiology. 2008; 19:680–689. [PubMed: 18714438]
- Gent JF, Triche EW, Holford TR, Belanger K, Bracken MB, Beckett WS, Leaderer BP. Association of low-level ozone and fine particles with respiratory symptoms in children with asthma. JAMA. 2003; 290:1859–1867. [PubMed: 14532314]
- Gent JF, Koutrakis P, Belanger K, Triche E, Holford TR, Bracken MB, Leaderer BP. Symptoms and medication use in children with asthma and traffic-related sources of fine particle pollution. Environ Health Perspect. 2009; 117:1168–1174. [PubMed: 19654929]
- Gertler A, Kuhns H, Abu-Allaban M, Damm C, Gillies J, Etyemezian V, Clayton R, Proffitt D. A case study of the impact of winter road sand/salt and street sweeping on road dust re-entrainment. Atmos Environ. 2006; 40:5976–5985.
- Grieshop AP, Lipsky EM, Pekney NJ, Takahama S, Robinson AL. Fine particle emission factors from vehicles in a highway tunnel: Effects of fleet composition and season. Atmos Environ. 2006; 40:S287–S298.
- Kim E, Hopke PK, Pinto JP, Wilson WE. Spatial variability of fine particle mass, components, and source contributions during the regional air pollution study in St. Louis Environ Science Tech. 2005; 39:4172–4179.
- Kuhn T, Biswas S, Sioutas C. Diurnal and seasonal characteristics of particle volatility and chemical composition in the vicinity of a light-duty vehicle freeway. Atmos Environ. 2005; 39:7154–7166.
- Laden F, Neas LM, Dockery DW, Schwartz J. Association of fine particulate matter from different sources with daily mortality in six U.S cities. Environ Health Perspect. 2000; 108:941–947. [PubMed: 11049813]
- Lee PKH, Brook JR, Dabek-Zlotorzynska E, Mabury SA. Identification of the major sources contributing to PM2.5 observed in Toronto. Environ Science Tech. 2003; 37:4831–4840.
- Lewis TC, Robins TG, Dvonch JT, Keeler GJ, Yip FY, Mentz GB, Lin X, Parker EA, Israel BA, Gonzalez L, Hill Y. Air pollution-associated changes in lung function among asthmatic children in Detroit. Environ Health Perspect. 2005; 113:1068–1075. [PubMed: 16079081]
- Liu LJS, Burton R, Wilson WE, Koutrakis P. Comparison of aerosol acidity in urban and semi-rural environments. Atmos Environ. 1996; 30:1237–1245.
- Mar TF, Norris GA, Koenig JQ, Larson TV. Associations between air pollution and mortality in Phoenix, 1995 to 1997. Environ Health Perspect. 2000; 108:347–353. [PubMed: 10753094]
- Ostro B, Roth L, Malig B, Marty M. The effects of fine particle components on respiratory hospital admissions in children. Environ Health Perspect. 2009; 117:475–480. [PubMed: 19337525]
- Ozkaynak H, Thurston GD. Associations between 1980 United States mortality rates and alternative measures of airborne particle concentration. Risk Anal. 1987; 7:449–461. [PubMed: 3444932]
- Paatero P, Tapper U. Positive Matrix Factorization: a non-negative factor model with optimal utilization of error estimates of data values. Environmetrics. 1994; 5:111–126.
- Paatero P. Least squares formulation of robust non-negative factor analysis. Chemometr Intell Lab Systems. 1997; 37:23–35.
- Paatero P, Hopke PK, Song XH, Ramadan Z. Understanding and controlling rotations in factor analytic models. Chemometr Intell Lab Systems. 2002; 60:253–264.
- Paatero P, Hopke PK. Discarding or downweighting high-noise variables in factor analytic models. Anal Chim Acta. 2003; 490:277–289.
- Paatero P, Hopke PK, Begum BA, Biswas SK. Graphical diagnostic method for assessing the rotation in factor analytic models of atmospheric pollution. Atmos Environ. 2005; 39:193–201.
- Rogge WF, Hildemann LM, Mazurek MA, Cass GR. Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: Roads as sources and sinks. Environ Science Tech. 1993; 27:1892–1904.
- Shah SD, Cocker DR, Miller JW, Norbeck JM. Emission rates of particulate matter and elemental and organic carbon from in-use diesel engines. Environ Science Tech. 2004; 38:2544–2550.
- Suh HH, Nishioka Y, Allen GA, Koutrakis P, Burton RM. The metropolitan acid aerosol characterization study: Results from the summer 1994 Washington, D.C. field study. Environ Health Perspect. 1997; 105:826–834. [PubMed: 9347898]
- Connecticut Department of Environmental Protection. [Accessed 26 Apr 2009] Low sulfur heating oil in the northeast states: an overview of benefits, costs, and implementation issues. 2005. Available at:

[http://www.ct.gov/dep/lib/dep/air/regulations/sip/regionalhaze/aa_report060101heatingoil\[1\].pdf](http://www.ct.gov/dep/lib/dep/air/regulations/sip/regionalhaze/aa_report060101heatingoil[1].pdf)

- Energy Information Administration (EIA). [Accessed 19 Apr 2009] Fuel oil and kerosene sales. 2003. Available at:
	- [http://www.eia.doe.gov/pub/oil_gas/petroleum/data_publications/fuel_oil_and_kerosene_sales/](http://www.eia.doe.gov/pub/oil_gas/petroleum/data_publications/fuel_oil_and_kerosene_sales/historical/2003/pdf/table1.pdf) [historical/2003/pdf/table1.pdf](http://www.eia.doe.gov/pub/oil_gas/petroleum/data_publications/fuel_oil_and_kerosene_sales/historical/2003/pdf/table1.pdf)

Energy Information Administration (EIA). [Accessed 28 May 2009] Oil imports by month. 2008. Available at:

[http://www.eia.doe.gov/oil_gas/petroleum/data_publications/company_level_imports/](http://www.eia.doe.gov/oil_gas/petroleum/data_publications/company_level_imports/cli_historical.html) [cli_historical.html](http://www.eia.doe.gov/oil_gas/petroleum/data_publications/company_level_imports/cli_historical.html)

- U.S. Environmental Protection Agency (U.S. EPA). [Accessed 29 May 2009] AP-42: Fuel oil combustion. 1998. Available at:<http://www.epa.gov/ttn/chief/ap42/ch01/index.html>
- U.S. Environmental Protection Agency (U.S. EPA). [Accessed 20 Jul 2010] Ambient PM_{2.5} monitoring. 2010. Available at: www.epa.gov/ttn/amtic

Appendices

Danbury

Hartford

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Springfield

Figure A.

Scatter plots between measured concentrations and predicted concentrations Note: The R^2 is increased to 0.84 (slope=0.95; intercept=0.27) without an outlying measured concentration of 105 μ g/m³.

Table A
Source profile of PM_{2.5} in Bridgeport, Danbury, New Haven, and Springfield (Unit: µg/m³ for PM_{2.5} and ng/m³ for all elements) Source profile of PM_{2.5} in Bridgeport, Danbury, New Haven, and Springfield (Unit: µg/m³ for PM_{2.5} and ng/m³ for all elements)

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New Haven

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Correlations between sites for PM_{2.5} mass, source contributions, and selected PM_{2.5} chemical components Correlations between sites for PM_{2.5} mass, source contributions, and selected PM_{2.5} chemical components

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Table C
Average and seasonal correlations between the regional averages and the concentrations in Bridgeport, Danbury, Hartford, New Haven, or Springfield for
PM_{2.5} mass concentrations, source contributions, and selected Average and seasonal correlations between the regional averages and the concentrations in Bridgeport, Danbury, Hartford, New Haven, or Springfield for $PM_{2.5}$ mass concentrations, source contributions, and selected $PM_{2.5}$ chemical components

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Research Highlights

- PMF analysis identified five source types of PM_{2.5}.
- The regional sulfur and traffic were major contributors to PM_{2.5}.
- Regional averages from several PM_{2.5} monitors are more reliable than data from the nearest central monitor for health effects studies.

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Summary of PM_{2.5} mass concentrations (μ g/m³) for the five monitoring sites (Mean (SE)) μ g/m³) for the five monitoring sites (Mean (SE)) Summary of PM2.5 mass concentrations (

Note: Warm-Season (April-October) and Cold-Season (November-March) Note: Warm-Season (April-October) and Cold-Season (November-March)

Summary of $PM_{2.5}$ mass (μ g/m³), chemical component concentrations (ng/m³), and source contributions (μ g /m³)

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for comparisons between sites.

for comparisons between sites.

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The seasonal and the day of week variations of the source contributions (The seasonal and the day of week variations of the source contributions $(\mu g/m^3)$ by site

Correlations between the concentrations in Hartford and the ones in all other sites for PM_{2.5} mass, source contributions, and selected PM_{2.5} chemical Correlations between the concentrations in Hartford and the ones in all other sites for PM_{2.5} mass, source contributions, and selected PM_{2.5} chemical components

5-site average correlations between the regional averages and the respective site concentrations for PM_{2.5} mass concentrations, source contributions, and 5-site average correlations between the regional averages and the respective site concentrations for $PM_{2.5}$ mass concentrations, source contributions, and selected PM_{2.5} chemical components selected PM_{2.5} chemical components

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