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Spatial Variation in Particulate Concentrations within Metropolitan Philadelphia

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During the summers of 1992 and 1993, particle mass concentrations ($PM_{2.5}$ and PM_{10}) were measured at eight sites located within metropolitan Philadelphia. Particle sampling was performed simultaneously at these sites on alternate days during the summer of 1992 and every day at seven of these sites during the summer of 1993. Sampling was conducted over 24-h periods beginning at 9 AM (EDT) during both summers. All $PM_{2.5}$ and PM_{10} samples were collected using 10 L/min inertial impactors with particle cutpoints of 2.5 and 10 μm, respectively. In this paper, we examine the relationship among $PM_{2.5}$, coarse particulate ($2.5 < d_a < 10$ μm), and PM_{10} concentrations. In addition, we analyze their spatial variation and compare our findings with those made in an earlier study of sulfate (SO_4^{2-}) concentrations. $PM_{2.5}$ and PM_{10} concentrations were found to be relatively uniform across Philadelphia, suggesting that concentrations measured at a single monitoring site are able to characterize particulate concentrations across Philadelphia and other similar urban areas well. Coarse particulate concentrations were found to vary spatially within Philadelphia, with its variation related to population density. Coarse particulate levels were also shown to vary by day of week as weekday levels were higher than weekend levels. Variability in PM_{10} concentrations was driven primarily by variability in $PM_{2.5}$ concentrations, which in Philadelphia comprised approximately 75% of PM_{10}. SO_4^{2-} related species in Philadelphia were, in turn, responsible for variability in $PM_{2.5}$ and, as a result, in PM_{10} as well. SO_4^{2-}-associated species were the largest component of both $PM_{2.5}$ and PM_{10} concentrations, comprising approximately 65 and 50% of their concentrations, respectively.

Introduction

Perhaps the clearest indication of the danger posed by urban pollution was provided by the severe 1952 London, England, air pollution episode in which approximately 4000 excess deaths occurred (*1*). The cause of these excess deaths was attributed primarily to particulate matter (*1*). Further evidence of the potential adverse effects of particles and their constituents was provided by similar, although less devastating, air pollution episodes that occurred in Donora, PA, and other communities in the mid-1900s.

Today, due to improved pollution control technology and environmental protection laws, such catastrophic air pollution episodes no longer occur in the United States. Despite this, concerns about the effects of particles and their constituents persist. Recent findings from epidemiologic studies have consistently demonstrated associations between currently observed particle concentrations and daily mortality for communities across the United States (*2*−*6*). These associations have been demonstrated primarily for total suspended particulate and PM_{10} concentrations, in large part because these air pollution data have been historically available. Results from other studies, however, suggest that fine particles ($PM_{2.5}$) and particle components, such as sulfate (SO_4^{2-}), and aerosol strong acidity (H^+) also may be associated with increased incidence of adverse health impacts (*7*−*12*).

The consistency and significance of these findings has resulted in increased efforts to examine the effects of specific constituents of particulate matter on community morbidity and mortality. The success of these efforts will depend on the relative contributions of these constituents to overall particulate levels and on the ability of monitoring data to represent the exposures of entire study populations. To investigate these issues, we have undertaken the Metropolitan Aerosol Acidity Characterization Study (MAACS). This study is intended to (1) characterize fine ($PM_{2.5}$), coarse ($2.5 < d_a < 10$ μm), and inhalable (PM_{10}) concentrations in urban environments and (2) identify the particulate size fraction responsible for the observed variability in outdoor PM_{10} concentrations. This study will be conducted in five eastern United States cities: Philadelphia, PA, Washington, DC, Nashville, TN, Boston, MA, and Atlanta, GA.

This paper presents findings from Philadelphia, the first of the five MAACS cities. In Philadelphia, $PM_{2.5}$ and PM_{10} concentrations were measured simultaneously during the summers of 1992 and 1993 at six sites located throughout the metropolitan Philadelphia area, at a neighboring urban site, and at an upwind, background site. Population density and relative location information were obtained for each site. At one of the metropolitan Philadelphia sites, wind direction, relative humidity, and temperature were measured hourly. These air pollutant, meteorologic, and site characteristics data were used to determine the extent of spatial variation in $PM_{2.5}$, coarse particulate, and PM_{10} concentrations within the metropolitan Philadelphia area and to assess the relative contributions of the various particulate measures.

Methods

Particle mass concentrations ($PM_{2.5}$ and PM_{10}) were measured in the metropolitan Philadelphia area during the

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TABLE 1

Summary of Site-Specific Characteristics[a]

site	population density (persons/mi^2)	distance from city center (km)	direction (from city center in deg from North)
Valley Forge (VAL)	2 191	28.8	315
Roxborough (ROX)	6 037	13.2	315
Northeast Airport (NE)	7 281	18.3	45
Camden, NJ (CAM)	9 721	7.2	90
City Laboratory (LAB)	12 052	8.9	45
Presbyterian (PBY)	15 822	6.1	225
City Center (CEN)	18 323	0.6	0
Temple University (TEM)	18 369	3.4	0

[a] Population densities obtained using City of Philadelphia (1990) census data.

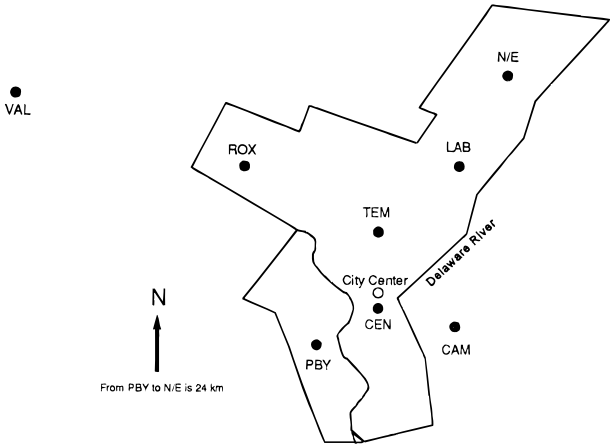

FIGURE 1. Monitoring sites for the study conducted in the metropolitan Philadelphia area during the summers of 1992 and 1993. Note that the ROX site was monitored only during the summer of 1992.

summers of 1992 and 1993. Sampling was conducted simultaneously at eight stationary ambient monitoring (SAM) sites during the summer of 1992 and at seven of these sites during the summer of 1993 (Figure 1). Sites were selected based on their population density and on their distance and direction from downtown Philadelphia (Table 1). Five of the sites within Philadelphia were positioned in a southwest to northeast line, spanning the longest direction of the city and corresponding to the hypothesized prevailing summer wind direction. The sixth Philadelphia site, Roxborough (ROX), was monitored only during the first summer and was located to the west of the city center. Other monitoring sites were located in Camden, NJ (CAM), a neighboring city east of the Delaware River, and in Valley Forge, PA (VAL), a rural site approximately 29 km northwest of the city center (defined as City Hall). The VAL site was located upwind from Philadelphia and thus was selected as the "background" site. The site elevations were comparable with the exception of that for the ROX site, which was approximately 100 m higher than the other site elevations. None of the selected sites were impacted by large local stationary particle sources. Population densities, which were obtained with the Planning Analysis Section using Philadelphia Census Track (*13*) figures, were highest at the TEM and CEN sites and were lowest at the VAL, ROX, and N/E sites.

Particle sampling was performed on alternate days during the summer of 1992 and every day during the summer of 1993. All samples were collected for 24-h periods beginning at 9 AM (EDT). Samplers generally were placed on the roof of a trailer or low-story building, with their inlets approximately 6 ft above the roof. At the rural VAL site, however, samplers were placed on a 10-ft high pole, with their inlets approximately 6 ft above ground level. $PM_{2.5}$ and PM_{10} samples were collected at flow rates of 10 L/min using inertial impactors. The impactors consisted of an inlet-impactor section to remove particles above either 2.5 or 10 μm in diameter followed by a Teflon filter mounted in a plastic holder to collect $PM_{2.5}$ or PM_{10}, respectively (*14*, *15*). The impactor plate was oiled to minimize particle bounce and to provide a sharp particle cutpoint.

Particle concentrations were determined using pre- and post-sampling filter weights. All filter weights were measured using electronic microbalances (Cahn Models 21 and C-31). In order to ensure consistent values for mass, the filters were equilibrated prior to weighing under controlled temperature (65−75 F) and relative humidity (40 ± 5%) conditions. In order to eliminate the effects of static charge, filters were passed over polonium sources (α-rays) prior to weighing. LODs for $PM_{2.5}$ and PM_{10} samples were estimated to equal three times the root mean square error (RMSE) of the blank filter measurements or 4.0 and 4.2 μg m^{-3}, respectively. The precision of the measurements were determined using collocated samples (28 pairs) and was found to equal 11% for $PM_{2.5}$ and 9.2% for PM_{10}. Sampling, preparation, and analysis procedures for $PM_{2.5}$ and PM_{10} have been described previously (*14*).

Coarse particle ($2.5 < d_a < 10\ \mu$m) concentrations were determined using the difference between measured PM_{10} and $PM_{2.5}$ concentrations. The precision of coarse particle concentrations was determined using collocated $PM_{2.5}$ and PM_{10} data, where two $PM_{2.5}$ ($1_{PM2.5}$ and $2_{PM2.5}$) and two PM_{10} (1_{PM10} and 2_{PM10}) samples were collocated and collected for identical monitoring periods. For each monitoring period, coarse particle concentrations were calculated for each of the four $PM_{10}-PM_{2.5}$ combinations: $1_{PM10}-1_{PM2.5}$, $1_{PM10}-2_{PM2.5}$, $2_{PM10}-1_{PM2.5}$, and $2_{PM10}-2_{PM2.5}$. The difference of the coarse particulate concentration from the mean coarse particulate concentration for its monitoring period was determined and used to calculate the RMSE of coarse particulate measurements. The precision was estimated to equal the RMSE of the coarse particulate concentrations divided by the overall mean coarse particle concentration or 40%.

Wind direction, wind speed, relative humidity, and temperature were measured hourly at the North Philadelphia Airport (N/E). Although meteorologic data were collected only at the N/E site, meteorologic conditions are likely to be similar for all sampling sites, since the distances separating the sites were relatively small. Except for the background VAL site, all sites were within a radius of 10 mi from the city center. The most prevalent wind direction

TABLE 2

Summary Statistics for $PM_{2.5}$, Coarse Particles, and PM_{10}[a]

pollutant/site	1992 N	1992 mean ± SD	1992 max	1993 N	1993 mean ± SD	1993 max
		$PM_{2.5}$				
Valley Forge (VAL)	35	19.3 ± 8.3	36.1	76	22.2 ± 11.6	61.7
Roxborough (ROX)	32	17.7 ± 8.3	35.0			
North Airport (N/E)	34	19.2 ± 9.2	38.4	75	21.3 ± 11.8	65.9
Camden, NJ (CAM)	32	17.8 ± 7.9	35.1	74	22.4 ± 11.4	54.8
Laboratory (LAB)	34	20.6 ± 9.7	43.4	73	23.6 ± 12.0	68.2
Presbyterian (PBY)	33	21.0 ± 9.6	40.8	74	23.1 ± 12.3	72.2
500 S. Broad St. (CEN)	34	20.1 ± 10.5	40.6	71	25.1 ± 12.3	64.6
Temple Univer. (TEM)	28	19.6 ± 10.0	37.4	76	23.5 ± 11.3	70.0
		Coarse Particles				
Valley Forge (VAL)	35	5.2 ± 3.5	14.5	75	6.3 ± 3.0	14.5
Roxborough (ROX)	29	6.9 ± 2.3	14.4			
North Airport (N/E)	33	6.3 ± 4.0	16.3	74	7.0 ± 3.6	22.3
Camden, NJ (CAM)	29	6.2 ± 3.9	17.6	73	7.0 ± 3.2	19.2
Laboratory (LAB)	34	8.1 ± 3.9	21.4	70	7.8 ± 4.9	25.8
Presbyterian (PBY)	32	5.6 ± 4.6	14.4	73	8.8 ± 4.1	24.2
500 S. Broad St. (CEN)	32	5.1 ± 4.4	16.5	71	8.1 ± 4.0	20.9
Temple Univer. (TEM)	24	6.1 ± 6.5	19.9	75	7.9 ± 4.2	23.1
		PM_{10}				
Valley Forge (VAL)	35	24.5 ± 8.5	44.1	76	28.7 ± 13.3	67.3
Roxborough (ROX)	30	25.3 ± 9.9	45.7			
North Airport (N/E)	34	26.2 ± 11.1	48.1	74	28.2 ± 13.8	73.7
Camden, NJ (CAM)	32	24.5 ± 10.6	48.6	76	29.4 ± 13.4	74.0
Laboratory (LAB)	36	28.4 ± 11.8	53.3	70	31.9 ± 14.4	72.7
Presbyterian (PBY)	35	27.0 ± 11.4	50.8	73	32.0 ± 13.2	72.3
500 S. Broad St. (CEN)	32	24.8 ± 12.6	55.6	77	32.8 ± 12.9	70.9
Temple Univer. (TEM)	26	26.2 ± 11.8	53.1	76	31.9 ± 13.5	76.8

[a] Samples were collected on alternate days at eight sites within the metropolitan Philadelphia area during the summer of 1992 and at seven of these sites (except ROX) during the summer of 1993. All concentrations expressed as $\mu g\ m^{-3}$). *N* indicates the sample size.

as well as the mean temperatures, relative humidities, and wind speeds for the 24-h sampling period were used to represent the weather conditions for each period. Prevalent wind directions were categorized as north, south, east, or west. Data were omitted from data analyses when field or laboratory problems justified their exclusion. Data capture was 95% of 826 possible samples for both $PM_{2.5}$ and PM_{10}. All statistical analyses were performed using the Statistical Analysis System (SAS) (*16*).

$PM_{2.5}$, coarse particulate, and PM_{10} concentrations at each site were characterized and compared using Pearson correlation coefficients, one-way analysis of variance (ANOVA), and univariate regression procedures. The influences of population density, distance from the city center, and wind direction on outdoor $PM_{2.5}$, coarse particulate, and PM_{10} levels were examined using Pearson correlation coefficients and regression techniques. The relative contributions of fine and coarse particles to overall PM_{10} concentrations also are investigated. Statistical significance was based on *p*-values of 0.05.

Results

Summary results of $PM_{2.5}$, coarse particle, and PM_{10} concentrations are presented by site in Table 2. $PM_{2.5}$, PM_{10}, and coarse particle concentrations were higher during the summer of 1993 as compared to the summer of 1992 (Table 2). Correspondingly, episodes of elevated $PM_{2.5}$ and PM_{10} concentrations were also more frequent during the summer of 1993. The disparity in the concentrations was most pronounced for $PM_{2.5}$ and PM_{10} as maximum concentrations differed by nearly a factor of 2. Maximum coarse particle concentrations generally were comparable across the two summers, as were mean and maximum temperatures and wind speeds. The incidence of northern, southern, eastern, and western wind directions were similar for both summers. Summary meteorological data and site-specific characteristics have been described previously (*17*).

Location and Meteorological Parameters. Although the distances separating the sites were small, the sites were characterized by a wide range of population densities, from approximately 2000 to 18 000 persons/mi^2 (Table 1). Population density was negatively associated with distance from the city center ($r = -0.91$). The most prevalent wind direction for the 24-h sampling periods during both 1992 and 1993 was from the west. In 1992, 44, 29, 15, and 12% of the sample days had prevalent winds from the west, north, south, and east, respectively. In 1993, 39% of the sample days were characterized by westerly winds, 31% by southerly winds, 16% by easterly winds, and 14% by northerly winds. Wind speed and temperature were found to differ by wind direction using ANOVA and Tukey HSD procedures. Wind speeds were highest when winds were from the eastern direction, followed by those from the northern direction, and finally by those from the southern and western directions. Temperature showed the opposite pattern as it was highest when the wind direction was from the southern and western directions and lowest when the wind was from the eastern direction.

Relationship among $PM_{2.5}$, Coarse Particles, and PM_{10}. $PM_{2.5}$ concentrations tended to be substantially higher than corresponding coarse particle levels, comprising the largest fraction of PM_{10} concentrations. Coarse particle concentrations generally were low, reaching a maximum of only 21 and 26 $\mu g\ m^{-3}$ during the summers of 1992 and 1993,

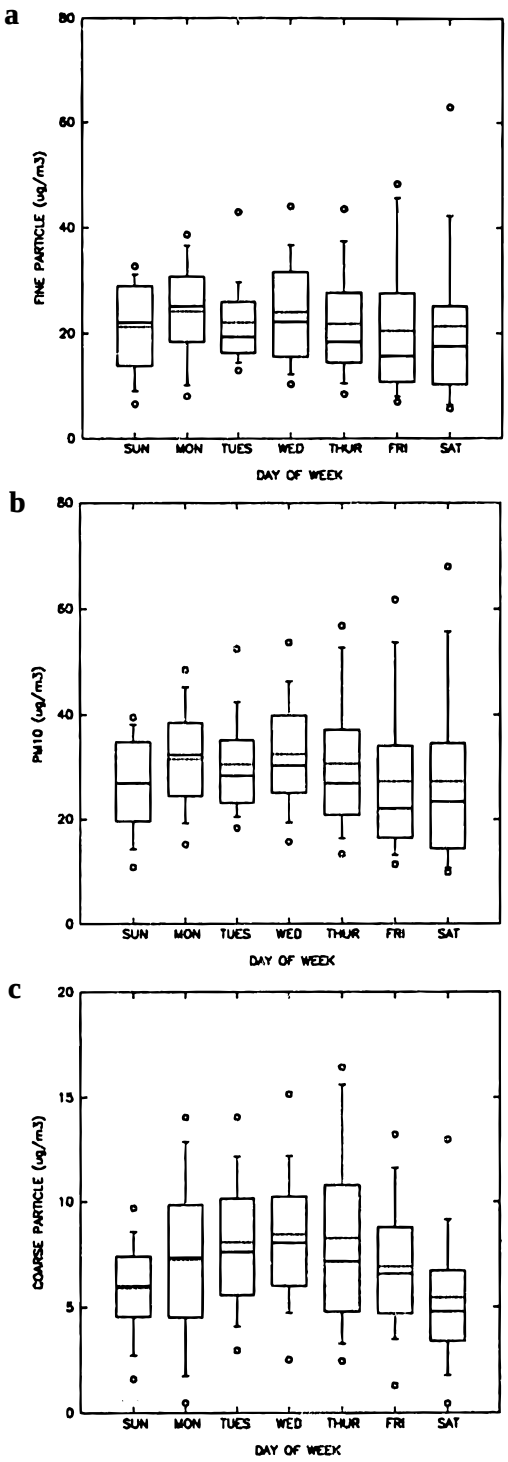

FIGURE 2. (a) $PM_{2.5}$, (b) PM_{10}, and (c) coarse particulate concentrations stratified by day of week, respectively. Samples collected at eight sites located in Philadelphia, PA, during the summer of 1992 and at seven of these sites during the summer of 1993.

respectively. Neither $PM_{2.5}$ nor PM_{10} concentrations were found to vary significantly by day of week (Figure 2a,b); however, coarse particle concentrations were found to be significantly higher on weekdays as compared to weekends using a two-sample *t*-test (Figure 2c). These differences were more pronounced at the metropolitan Philadelphia sites as compared to the background site VAL.

$PM_{2.5}$ and PM_{10} concentrations measured during the summers of 1992 and 1993 were strongly correlated at all sites (Table 3). Correlations between PM_{10} and coarse particle concentrations were weaker but remained significant (Table 3). The relative contributions of fine and coarse particles to PM_{10} were compared for the eight sites using ratios of $PM_{2.5}/PM_{10}$ concentrations. At all sites, $PM_{2.5}$ comprised the largest fraction of PM_{10} concentrations. The mean $PM_{2.5}/PM_{10}$ ratios at the eight sites were similar, ranging between 0.71 and 0.77. Ratios increased slightly with PM_{10} levels, as evidenced by the significant positive correlations between the $PM_{2.5}/PM_{10}$ ratio and PM_{10} concentrations ($r = 0.17$). When data were stratified by site, however, ratios at the PBY and TEM sites were found to be independent of PM_{10} concentrations.

Spatial Variation in Particle Concentrations. Significant correlations generally were found for $PM_{2.5}$, PM_{10}, and coarse particle concentrations measured at the eight sites (Tables 4−6). Correlations between the sites were strongest for $PM_{2.5}$, with most correlation coefficients near unity. Correlation coefficients for pairwise comparisons of site PM_{10} concentrations were slightly lower than those for $PM_{2.5}$ but remained high. For coarse particles, correlation coefficients were substantially lower, reaching a maximum of only 0.61. $PM_{2.5}$ and PM_{10} concentrations were found to be uniform across the eight sites using one-way analysis of variance (ANOVA) procedures. Coarse particle concentrations, on the other hand, exhibited significant spatial variation, as VAL site concentrations were significantly lower than those measured at the LAB and PBY sites.

Neither population density nor distance from the city center were correlated to $PM_{2.5}$, coarse particulate, or PM_{10} concentrations (Table 3). As discussed above, coarse particulate concentrations were lower in the less populated communities (VAL) as compared to the more densely populated communities (LAB, PBY). This trend was consistent across communities, as VAL, ROX, N/E, and CAM coarse particle concentrations were lower than those for LAB, PBY, TEM, and CEN. All particulate measures were positively correlated to temperature, while all measures except for coarse particles were negatively correlated to wind speed.

Particle concentrations were found to differ significantly by wind direction using ANOVA and Tukey HSD procedures. Both $PM_{2.5}$ and PM_{10} concentrations were significantly higher when winds were from the west and south as compared to the north and east (Figures 3 and 4). Higher $PM_{2.5}$ concentrations from the west and south were expected since major sources of $PM_{2.5}$ and sulfur dioxide (SO_2), the SO_4^{2-} precursor, were located west of Philadelphia, in areas such as Pittsburgh and the Ohio River Valley.

When stratified by wind direction, mean $PM_{2.5}$ concentrations did not vary by site or population density, as regression of mean $PM_{2.5}$ concentrations on population density showed insignificant slopes and low values of R^2 (<0.02) for all wind directions. Similar results were found for PM_{10}. Regression of mean PM_{10} concentrations on population density resulted in low R^2 values (<0.04) for all wind directions and insignificant slopes for southerly and westerly winds. Regression of mean PM_{10} concentrations on population density, however, yielded significant slopes for winds from the east (0.0004 ± 0.0002) and north (0.0003 ± 0.0001). For these wind directions, these results suggest that PM_{10} concentrations increase by 4 μg m^{-3} with every 10 000 persons/mi^2.

Coarse particle concentrations were found to differ significantly by wind direction using ANOVA procedures,

TABLE 3

Correlation Coefficients for Pairwise Comparisons between Particulate Measures and Site and Meteorlogical Variables[a]

	$PM_{2.5}$	PM_{10}	coarse particles	population density	temp	wind speed
PM_{10}	0.95*					
coarse particles	0.29*	0.57*				
population density	0.08*	0.11*	0.12*			
temp	0.61*	0.64*	0.37*	NA		
wind speed	−0.24*	−0.21*	−0.05	NA	−0.12*	

[a] Samples were collected on alternate days at eight sites within the metropolitan Philadelphia area during the summer of 1992 and at seven of these sites (except ROX) during the summer of 1993. Asterisks (*) indicate significant correlations (p-values < 0.05). NA indicates not available.

TABLE 4

Pearson Correlation Coefficients for Pairwise Comparisons of Outdoor $PM_{2.5}$ Concentrations Measured in Metropolitan Philadelphia[a]

	VAL	ROX	NE	CAM	LAB	PBY	CEN	TEM
ROX	0.96							
NE	0.90	0.94						
CAM	0.92	0.93	0.94					
LAB	0.91	0.90	0.94	0.94				
PBY	0.90	0.79	0.94	0.89	0.89			
CEN	0.84	0.70	0.80	0.87	0.86	0.85		
TEM	0.92	0.92	0.94	0.93	0.93	0.93	0.87	

[a] Data were collected over 24-h periods on alternate days at eight sites during the summer of 1992 and at seven of these sites during the summer of 1993. Measurements were not made at the ROX site during 1993. All correlations were significant (p-values < 0.05).

TABLE 5

Pearson Correlation Coefficients for Pairwise Comparisons of Outdoor PM_{10} Concentrations Measured in Metropolitan Philadelphia[a]

	VAL	ROX	NE	CAM	LAB	PBY	CEN	TEM
ROX	0.96							
NE	0.88	0.86						
CAM	0.89	0.91	0.92					
LAB	0.87	0.92	0.90	0.90				
PBY	0.90	0.72	0.82	0.85	0.81			
CEN	0.83	0.62	0.81	0.83	0.79	0.81		
TEM	0.88	0.79	0.89	0.91	0.87	0.91	0.80	

[a] Data were collected over 24-h periods on alternate days at eight sites during the summer of 1992 and at seven sites during the summer of 1993. Samples were not collected at the ROX site during 1993. All correlations were significant (p-values < 0.05).

as concentrations were highest when winds were from the south and west (Figure 5). When stratified by wind direction, coarse particle concentrations did not vary by site or population density for westerly wind directions, as regression of mean coarse particle concentrations on population density showed insignificant slopes and low R^2 values. For easterly, northerly, and southerly winds, coarse particle levels were found to increase by $1-2\ \mu g\ m^{-3}$ with every 10 000 persons/mi^2. [Regression of mean coarse particle concentrations on population density yielded significant slopes of 0.0001 (±0.00006), 0.0002 (±0.00006), and 0.0001 (±0.00005), respectively.] Population density, however, explained little of the variation in mean coarse particle concentrations, resulting in low R^2 values (<0.07) when mean concentrations were regressed on population density.

TABLE 6

Pearson Correlation Coefficients for Pairwise Comparisons of Outdoor Coarse Particle Concentrations Measured in Philadelphia, PA[a]

	VAL	ROX	NE	CAM	LAB	PBY	CEN	TEM
ROX	0.61*							
NE	0.34*	0.33						
CAM	0.38*	0.39*	0.52*					
LAB	0.32*	0.67*	0.38*	0.50*				
PBY	0.41*	0.30	0.26*	0.40*	0.36*			
CEN	0.28*	0.41*	0.32*	0.18	0.14	0.38*		
TEM	0.46*	0.47*	0.48*	0.63*	0.38*	0.42*	0.22*	

[a] Data were collected over 24-h periods on alternate days at eight sites during the summer of 1992 and at seven sites during the summer of 1993. Measurements were not obtained for the ROX site during 1993. Asterisks (*) indicate significant correlations (p-values < 0.05).

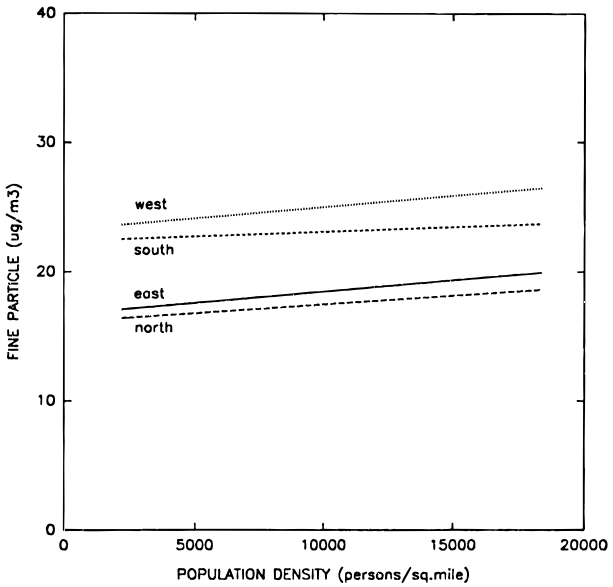

FIGURE 3. Regression lines of mean summertime fine particle (d_a < 2.5 μm) concentrations on population density. Mean concentrations stratified by wind direction. Data collected in Philadelphia, PA, during the summers of 1992 and 1993.

Discussion

From these findings, it is clear that PM_{10} concentrations were driven primarily by $PM_{2.5}$ concentrations. Not only did $PM_{2.5}$ comprise approximately 75% of PM_{10}, but as evidenced by the strong correlations between PM_{10} and $PM_{2.5}$, its variability also was the primary determinant of the variability in PM_{10} concentrations. Coarse particle concentrations, which comprised the remaining 25% of

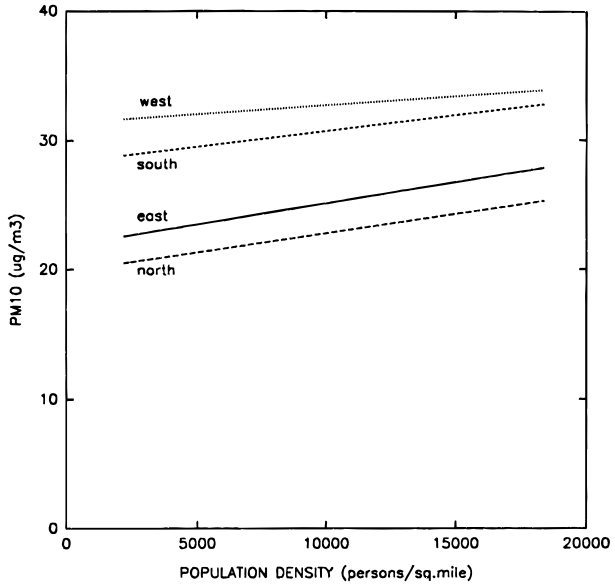

FIGURE 4. Regression lines of mean summertime PM_{10} ($d_a < 10\ \mu m$) concentrations on population density. Mean concentrations stratified by wind direction. Data collected in Philadelphia, PA, during the summers of 1992 and 1993.

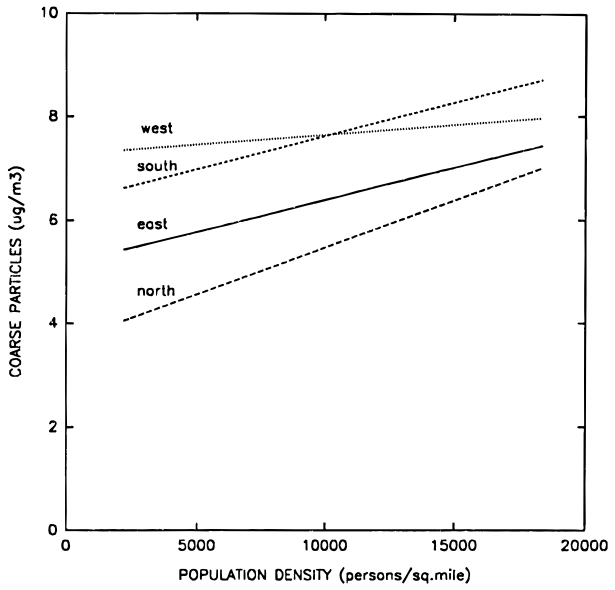

FIGURE 5. Regression lines of mean summertime coarse particle ($2.5 < d_a < 10\ \mu m$) concentrations on population density. Mean concentrations stratified by wind direction. Data collected in Philadelphia, PA, during the summers of 1992 and 1993.

PM_{10}, tended to be lower and less strongly correlated with PM_{10} concentrations.

Local factors, such as the day of the week, population density, and location, did not affect $PM_{2.5}$ concentrations, suggesting that local sources contributed little to its concentrations. Further support for this suggestion is provided by the observed uniformity in $PM_{2.5}$ concentrations within metropolitan Philadelphia and also by the significant correlations in the sites' $PM_{2.5}$ concentrations. These findings provide convincing evidence that sources for $PM_{2.5}$ are regional in nature, with fine particles being transported into Philadelphia from sources located to the west and south of the city. In addition, results from our analysis show that $PM_{2.5}$ concentrations within the city are unaffected by the local phenomena examined in our analysis, as no major local $PM_{2.5}$ sources exist and deposition, the primary fine particulate loss mechanism, occurs uniformly throughout the city.

Coarse particle concentrations, on the other hand, were dependent on local factors. Coarse particle concentrations were highest on weekdays, which may result from possible increases in population densities on these days. Correspondingly, weekday/weekend differences in coarse particle concentrations were most pronounced at the urban sites, where the discrepancy between weekday and weekend population densities may be more prominent. In addition, areas with higher population densities tended to have higher coarse particle concentrations as compared to less densely populated areas. These results indicate the importance of local sources to coarse particulate levels. Since coarse particulate sources are varied and scattered across the city, they cause coarse particle concentrations to vary spatially within Philadelphia.

When coarse particles are combined with $PM_{2.5}$ into a single-particle measure PM_{10}, the contribution of local sources is diminished. However, PM_{10} concentrations measured on weekends remained significantly lower than those measured on weekdays. As was the case for $PM_{2.5}$, PM_{10} concentrations were found to be uniform within Philadelphia, with PM_{10} concentrations also significantly correlated between all sites. Significant correlations among the sites are consistent with findings from an earlier study of inhalable particles conducted in Philadelphia by Suggs and Burton (*18*). Significant site correlations were due to the fact that PM_{10} was composed primarily of $PM_{2.5}$ at all eight sites. Correlation coefficients for pairwise comparisons between the sites' PM_{10} concentrations were lower than those for $PM_{2.5}$ as the result of the contribution of coarse particles to PM_{10} levels.

Concentrations of all particle measures were influenced by wind direction. For $PM_{2.5}$, concentrations were highest when winds were from the west and south, which was expected since fine particle sources were located to the west and south of Philadelphia. Wind direction, however, had no affect on the spatial variation in $PM_{2.5}$ concentrations, as $PM_{2.5}$ concentrations were uniform across Philadelphia irrespective of wind direction. Coarse particulate concentrations also were highest for westerly and southerly wind directions. Since coarse particles are produced locally, it is possible that the observed lower wind speeds for westerly and southerly winds resulted in decreased particulate dispersion and thus higher coarse particulate concentrations. This theory is contradicted, however, by the nonsignificant correlations found between coarse particle concentrations and wind speed. These nonsignificant correlations may be an artifact of the 24-h sampling times, which may obscure the relationship between coarse particle levels and wind speed. Higher coarse particle levels may also be due to regional transport of coarse particles into Philadelphia from regional sources located to the west and south of the city. Since the precision in coarse particle measurements was relatively low (≈40%), however, definitive conclusions about coarse particulate sources cannot be made, demonstrating the need for more precise coarse particulate measurements. Nonetheless, human activity-related sources are likely to be major sources of coarse particles, as coarse particulate concentrations were found to increase by approximately 1 $\mu g\ m^{-3}$ for every 10 000 persons/mi^2 when winds were from the east, north, and south wind directions.

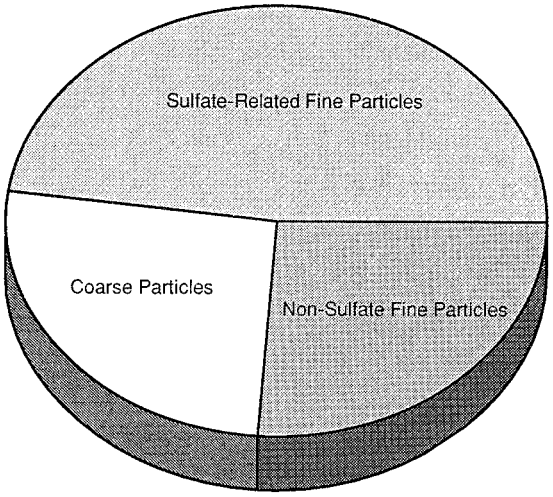

FIGURE 6. Composition of PM_{10} in Philadelphia, PA, during the summers of 1992 and 1993. Sulfate-related fine particles represents the fraction of PM_{10} that is comprised of SO_4^{2-}, NH_4^+, and H^+ or 48%, while nonsulfate fine particles represents the fraction of PM_{10} that is comprised of non-SO_4^{2-} fine particles or 26%. Coarse particles comprise 26% of PM_{10}. Data represent averages for the two summers. SO_4^{2-} concentrations reported in companion study (*17*).

As would be expected from these findings, PM_{10} concentrations also were highest when winds were from the west and the south. For these west and south winds, no spatial variation in PM_{10} concentrations was evident. For east and north winds, however, PM_{10} concentrations were found to vary spatially, increasing by approximately 2 $\mu g\ m^{-3}$ with every 10 000 persons/mi^2. This spatial variation probably resulted from the low $PM_{2.5}$ concentrations observed for east and north wind conditions, allowing coarse particles to have a greater influence on overall PM_{10} concentrations.

These results are consistent with those from earlier studies, which found that $PM_{2.5}$ is comprised primarily of SO_4^{2-} and SO_4^{2-}-associated species, such as ammonium ion (NH_4^+) and aerosol strong acidity (H^+) (*19*). Sources of $PM_{2.5}$ thus are comprised primarily of sources of sulfate precursors (e.g., SO_2), the largest of which are coal-fired utility plants located to the west and the south of Philadelphia along the Ohio River Valley. It was not surprising then to find that $PM_{2.5}$ concentrations exhibited characteristics similar to those of SO_4^{2-}. When considered in conjunction with results from a companion study conducted by Suh *et al.* (*17*), results show that both $PM_{2.5}$ and SO_4^{2-} are (1) highest in Philadelphia when winds were from the west and south, (2) uniform within the metropolitan Philadelphia area, and (3) relatively unaffected by local factors. This similarity between $PM_{2.5}$ and SO_4^{2-} was due to the fact that approximately 65% of the $PM_{2.5}$ in Philadelphia was composed of SO_4^{2-}-related species.

The importance of SO_4^{2-}-associated species to $PM_{2.5}$ suggests SO_4^{2-}-associated species also to be a significant determinant of PM_{10} concentrations. In fact, SO_4^{2-}-associated species were the largest component of PM_{10}, comprising in Philadelphia approximately 50% of PM_{10} as compared to approximately 25% each for non-SO_4^{2-} fine particles and coarse particles (Figure 6). These results indicate that the variability in the PM_{10} and $PM_{2.5}$ concentrations was driven in large part by the variability in SO_4^{2-}-associated concentrations. This variability in SO_4^{2-}-associated concentrations was, in turn, driven by changes in wind direction and other meteorological parameters (*17*). In other areas within the northeastern United States, SO_4^{2-}-associated species also are likely to be primary determinants of both $PM_{2.5}$ and PM_{10}. For areas in the western United States, however, other particulate components such as nitrates are likely to be more influential.

Conclusions

$PM_{2.5}$ was found to comprise a substantial fraction of PM_{10}. As a result, $PM_{2.5}$ and PM_{10} concentrations were highly correlated. Coarse particle concentrations also were significantly correlated with PM_{10} and to a lesser degree with $PM_{2.5}$. Correlation coefficients for pairwise comparisons showed PM_{10} and $PM_{2.5}$ concentrations to be highly correlated for all eight sites. Correlations between the sites' coarse particle concentrations were weaker but remained significant. $PM_{2.5}$, PM_{10}, and coarse particle concentrations were found to be uniform within metropolitan Philadelphia, but were found to vary with wind direction. Results indicate that $PM_{2.5}$ particles were transported into Philadelphia from regional sources located to the west and south, while coarse particle concentrations were generated primarily by local sources. Results also suggest that variability in PM_{10} concentrations is driven by corresponding variability in $PM_{2.5}$ levels. When compared with results from a companion study by Suh et al. (*17*), results also showed that $PM_{2.5}$, and thus PM_{10}, concentrations were driven by variability in SO_4^{2-}-associated concentrations. SO_4^{2-}-associated species were found to comprise approximately 65% of $PM_{2.5}$ and 50% of PM_{10} concentrations. A single stationary ambient monitor may be sufficient to estimate outdoor PM_{10} and $PM_{2.5}$ concentrations for urban populations living in the northeastern United States.

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