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The Denver Aerosol Sources and Health (DASH) Study: Overview and Early Findings

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Abstract

Improved understanding of the sources of air pollution that are most harmful could aid in developing more effective measures for protecting human health. The Denver Aerosol Sources and Health (DASH) study was designed to identify the sources of ambient fine particulate matter $(PM_{2.5})$ that are most responsible for the adverse health effects of short-term exposure to PM $_{2.5}$. Daily 24-hour PM_{2.5} sampling began in July 2002 at a residential monitoring site in Denver, Colorado, using both Teflon and quartz filter samplers. Sampling is planned to continue through 2008. Chemical speciation is being carried out for mass, inorganic ionic compounds (sulfate, nitrate and ammonium), and carbonaceous components, including elemental carbon, organic carbon, temperature-resolved organic carbon fractions and a large array of organic compounds. In addition, water soluble metals were measured daily for 12 months in 2003. A receptor-based source apportionment approach utilizing positive matrix factorization (PMF) will be used to identify PM 2.5 source contributions for each 24-hour period. Based on a preliminary assessment using synthetic data, the proposed source apportionment should be able to identify many important sources on a daily basis, including secondary ammonium nitrate and ammonium sulfate, diesel vehicle exhaust, road dust, wood combustion and vegetative debris. Meat cooking, gasoline vehicle exhaust and natural gas combustion were more challenging for PMF to accurately identify due to high detection limits for certain organic molecular marker compounds. Measurements of these compounds are being improved and supplemented with additional organic molecular marker compounds. The health study will investigate associations between daily source contributions and an array of health endpoints, including daily mortality and hospitalizations and measures of asthma control in asthmatic children. Findings from the DASH study, in addition to being of interest to policymakers, by identifying harmful PM2.5 sources may provide insights into mechanisms of PM effect.

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Keywords

air pollution; particulate matter; health effects; mortality; source apportionment

1. Introduction

Findings from observational epidemiology studies on the adverse health effects of ambient particulate matter (PM) and other pollutants indicate effects of short-term concentration changes on mortality and morbidity (Dominici et al., 2003; Pope and Dockery, 2006). As a surrogate for exposure, these studies have primarily relied on outdoor PM mass concentrations measured at one or more monitoring sites distributed over large urban areas. An important finding in recent observational studies is that there appears to be heterogeneity between urban areas in the magnitude of the observed acute effect of particulate air pollution (Samet et al., 2000; Dominici et al., 2003). The causes of this heterogeneity are not known. The PM contribution from various sources, and hence the chemical composition of PM, varies by location (US-EPA, 2004). If PM toxicity is dependent on its chemical composition, variability of PM source contributions could be one explanation for the heterogeneity of effects. Evaluating associations between source contributions and health effects may thus provide insight into the heterogeneity between urban areas.

The National Research Council has stressed the need to understand the comparative toxicity of PM from different sources and recommended the use of source apportionment methods (NRC, 2004). To date, a limited number of air pollution health studies have used source apportionment to investigate mortality effects (Laden et al., 2000; Mar et al., 2000, 2006; Tsai, Apte and Daisey, 2000; Ito et al., 2006), as well as effects on hospitalizations (Anderson et al., 2007) and emergency room visits (Schreuder et al., 2006; Sarnat et al., 2008). The Denver Aerosol Sources and Health (DASH) study is designed to investigate effects of exposure to sources of fine particulate matter ($PM_{2.5}$) on an array of health endpoints by incorporating detailed daily $PM_{2.5}$ chemical speciation data and state-of-the-art source apportionment into a health study.

Several options are available to identify the role of time-varying pollutant sources in the context of a health study. Concentrations of specific source markers have been used for this purpose (Laden et al., 2000), but valid markers for many sources of interest are not available. Deterministic air quality models can estimate contributions of sources at specific locations at 4 km or finer grid resolution (Held et al., 2005), but their ability to reproduce short-term variations is limited, due in part to the lack of temporally resolved emissions information (Marmur et al., 2006). Receptor-based source apportionment approaches use PM component concentrations measured at a monitoring site to estimate site-specific source contributions to PM concentrations. Receptor models have been used to help identify PM sources in numerous geographical regions (Maykut et al., 2003; Hopke et al., 2003; Schauer et al., 1996; Long et al., 2005; Lewis et al., 2003). These models either use a chemical mass balance (CMB) approach, which requires detailed information about the chemical composition of sources in a region, or use multivariate factor analysis methods, for example, positive matrix factorization (PMF) or UNMIX, to identify sources based on the temporal patterns among chemical components measured at the receptor(s). These multivariate approaches have the advantage of not requiring prior information on static source profiles, although source profile information is still needed to identify factors extracted from multivariate methods and unambiguous factor identification is not always possible. An important objective of the DASH study is to evaluate the utility of PMF for identifying short-term pollution source contributions to the ambient PM2.5 in a large urban area.

This paper provides an overview and introduction to the DASH study, focusing on the $PM_{2.5}$ characterization methods used and the source apportionment techniques being employed. Interim 4.5-year $PM_{2.5}$ species concentrations are also presented. In addition, model validation efforts using synthetic data are presented and discussed. Model uncertainty will be explored using a novel method based on a bootstrap approach and neural networks (Hemann et al., 2008). Companion papers describe in more detail the $PM_{2.5}$ monitoring and chemical analysis protocols, as well as the methodology for point-wise uncertainty estimation (Dutton et al., 2008a, 2008b). Subsequent papers will detail results of $PM_{2.5}$ species trends, source apportionment and epidemiological analyses.

2. Methods

2.1 PM_{2.5} Collection and Characterization

2.1.1 Core Receptor Site Monitoring—A residential site on the roof of Palmer Elementary School located 5.3 km east of downtown Denver was selected as the receptor site for detailed $PM_{2.5}$ chemical speciation (Figure 1). This site was selected for its central location in a large residential community and for its distance from point and mobile sources. The school is 5.2 km from the closest large highway and at least 0.6 km from the closest commuter street. Collection of daily 24-hour $PM_{2.5}$ samples from this site has been ongoing since July 1, 2002.

 $PM_{2.5}$ samples were collected daily from midnight to midnight on both Teflon and quartz fiber filters with sampling equipment that has been used extensively in air quality studies in the past (Lough, Schauer and Lawson, 2006; Christoforou et al., 2000). The sampling protocol and the bulk chemical analysis methods briefly described below are presented in more detail in Dutton et al., (2008a). A list of all species being analyzed is contained in Table S1 in the online supplementary data.

2.1.2 Bulk Chemical Analyses—Bulk chemical speciation of all samples includes the quantification of $PM_{2.5}$ mass, inorganic ionic compounds (sulfate, nitrate, and ammonium), elemental carbon (EC) and organic carbon (OC). Gravitational mass measurements are made using a microbalance housed in a temperature and humidity controlled chamber with filter weighing methods patterned after those approved by the EPA for $PM_{2.5}$ mass determination (US-EPA, 1998). Inorganic ionic compounds (sulfate, nitrate and ammonium) are quantified using an ion chromatograph following traditional methodology for airborne particulate matter (Mueller et al., 1978). Bulk EC and OC concentrations are determined by NIOSH Method 5040; a 1.5 cm² punch is taken from each quartz fiber filter and sent to the Wisconsin State Laboratory of Hygiene for analysis.

2.1.3 Organic Compound Chemical Analysis—Methods for the detailed organic chemical analysis of quartz fiber filter samples using solvent extraction and gas chromatography/mass spectrometry (GC/MS) have been developed previously (Mazurek et al., 1987; Fraser et al., 1996; Schauer et al., 1998; Hannigan et al., 1998) and similar methods were used in this study. High volume injection (50 µl) was incorporated by way of a programmable temperature vaporization inlet, thereby improving detection limits for trace level organic compounds (Crimmins and Baker, 2006). Detailed description of the organic speciation methods is included in Dutton et al. (2008b).

2.1.4 Quality Control and Uncertainty Estimation—Proper quality assurance and quality control are essential for prevention of contamination and for ensuring consistent chemical quantification. Consistency is especially important for a time series study. To assess contamination, weekly field blank filters are collected, extracted, and analyzed

following the same procedures used for the daily sample filters. The field blanks were used to detect contamination coming from the filter substrates, handling or contaminated solvents and supplies and were used to perform blank subtraction on the daily measurements to adjust for these and other sampling artifacts.

To assure consistent and robust chemical quantification and to assist in uncertainty estimation, bi-weekly duplicate samples were collected on collocated samplers. These duplicate samples were analyzed in parallel with the main samples allowing for a direct comparison of duplicated measurements over all chemical analyses performed. Consistent and robust chemical quantification also involves instrument and equipment calibration, which is achieved by routinely calibrating all chemical analysis equipment with authentic standards. Complete details on quality control and uncertainty estimation are presented in Dutton et al. (2008a).

2.1.5 Supplemental Monitoring Data—Speciation for trace metals was done for one year on the 2003 filters. Water soluble trace metal chemical analysis was performed using an Inductively-Coupled Plasma Mass Spectrometer (ICP/MS) housed in a dedicated laboratory at the Wisconsin State Laboratory of Hygiene. Analytical details and results from this supplemental analysis will be presented elsewhere.

In addition to the data collected at the DASH core receptor site, the Air Pollution Control Division of the Colorado Department of Public Health and Environment (CDPHE) operates and maintains a network of ambient pollutant monitoring stations for the Denver metropolitan region. The network in this region consists of 14 community monitoring stations providing hourly measurements of the following: PM_{10} at 3 sites, $PM_{2.5}$ at 4 sites, ozone at 9 sites, carbon monoxide at 6 sites, sulfur dioxide at 2 sites, nitrogen oxides at 2 sites, and meteorology at 11 sites. In addition to the hourly monitors, there are 7 PM_{10} and 6 $PM_{2.5}$ sites collecting 24-hour samples and one Speciation Trends Network (STN) site providing speciated $PM_{2.5}$ measurements every 6th day for inorganic ions, total elemental and organic carbon and trace metals. These data will be utilized in the PMF modeling effort.

2.2 PM_{2.5} Source Apportionment Model

PMF multivariate receptor modeling was chosen to evaluate the source contributions of the time series of receptor samples. PMF incorporates uncertainty estimates of the data to solve matrix factorization of a linear model as a constrained, weighted least-squares problem. For this study, point-wise uncertainty estimates incorporating sampling uncertainty, detection limits, missing data observations, and outliers are used to weight each data point. The least-squares approach used in PMF also permits the inclusion of additional constraints as a key part of the analysis, such as nonnegativity constraints.

2.2.1 The PMF Model—In the context of air quality source apportionment, PMF attempts to apportion a matrix of observations, *X*, composed of individual observations x_{ij} (*i*=1...*n*, *j*=1...*m*) into *p* distinct factors according to the model:

$$x_{ij} = \sum_{k=1}^{r} g_{ik} f_{kj} + \varepsilon_{ij}.$$
 Equation 1

In Equation 1, g_{ik} represents the absolute contribution ($\mu g/m^3$) of factor k to sample i, f_{kj} represents the relative fraction ($\mu g/\mu g$) of species j in factor k, and e_{ij} is the residual of the model. The x_{ij} consist of measurements of m chemical components in n different samples.

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PMF solves Equation 1 by minimizing Q, the sum of the squared, scaled residuals, defined by:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{\varepsilon_{ij}}{s_{ij}}\right)^2$$
Equation 2

where s_{ij} is an estimate of the uncertainty in the measurement x_{ij} . A positivity constraint is applied to g_{ik} and f_{kj} , thus providing a more physically interpretable result. Written in matrix notation, Equation 1 simplifies to:

$$X=GF+E$$
 Equation 3

where G is subsequently referred to as the factor contribution matrix and F the factor profile matrix.

With some prior knowledge and experience, the PMF factors can often be linked to pollution sources. Because source emissions are spatially and temporally variable, the factor profiles should be interpreted as a qualitative guide to the types of sources to which people are exposed and are qualitatively related to profiles published in the literature. The inclusion of molecular markers unique to specific source types assists in identifying the association between factor and source type.

2.2.2 PMF Model Validation—Similar to the PM_{2.5} characterization efforts, there is substantial investment in exploring the quality and validity of the PMF results. One approach to exploring the performance of a source apportionment model is to use synthetic data (Miller et al., 2002; Brinkman et al., 2006). A synthetic data set was developed to represent the PM2.5 data that were being gathered at the receptor site in Denver and analyzed using the PMF2 algorithm (Paatero et al., 1997). The creation of the synthetic data set was based on the methods employed in Brinkman et al. (2006); differences are highlighted. Briefly, synthetic estimates of factor contributions (G^{syn}) and factor profiles (F^{syn}) for several PM2.5 sources were created and combined into a synthetic observation matrix $X^{syn} = G^{syn} F^{syn}$. One year of daily observations were simulated, comprised of 39 chemical species from nine predefined sources (secondary ammonium nitrate and sulfate, gasoline and diesel vehicle exhaust, road dust, wood combustion, meat cooking, natural gas combustion and vegetative debris). These sources were chosen based on their importance for the region as identified by the 1996–1997 Northern Front Range Air Quality Study (NFRAQS) (Watson et al., 1998). Source profiles containing bulk species and organic molecular markers were obtained from the literature as cited in Brinkman et al. (2006). Limitations on the number of species included in the synthetic data set were imposed by source profile availability; Table S2 in the online supplementary data identifies the 39 species included in the synthetic data exercise. Temporal patterns such as seasonality and weekday/weekend differences were accounted for in the factor contributions. To incorporate temporal correlations between the synthetic source contributions resulting from meteorology, the source contributions representing secondary ammonium nitrate and ammonium sulfate, wood combustion and natural gas combustion were scaled by temperature. Further correlations between sources arose from the hypothesized temporal patterns (e.g., gasoline and diesel powered motor vehicles, paved road dust, wood combustion and meat cooking all contained a weekday/weekend dependence). Table S3 in the online supplementary data shows the correlations between the synthetic source contributions. Finally, the source contributions were scaled such that their daily sum added up to the actual daily PM_{2.5} mass measured during 2003. Normally distributed random noise was added to the synthetic observations x_{ii} based on measurement uncertainty, detection limits and detection limit uncertainty associated with the analytical techniques typically used

to speciate PM_{2.5} filter samples (Brinkman et al., 2006). The synthetic observations and estimated measurement uncertainties were input into PMF, which in turn provided modeled estimates of the factor contributions and profiles (G^{pmf} and F^{pmf} , respectively). These were compared (see Results) with the known synthetic input matrices, G^{syn} and F^{syn} , to evaluate the performance of the PMF model with this synthetic dataset.

2.2.3 PMF Model Uncertainty—There are several sources of uncertainty in a PMF model solution that can affect the validity of the source estimates. One source of uncertainty results from measurement error and random sampling error in the data. Uncertainty can also come from the PMF model itself due to rotational ambiguity in the solutions. In addition, solutions can vary depending on the values selected for model parameters including the number of factors and the parameters used to control rotations and down-weighting of outliers. As part of this study, Hemann et al. (2008) developed a novel method to estimate daily factor contribution uncertainties that are due to random sampling error. A balanced bootstrap technique will be used to create replicate datasets from the PM_{2.5} time series, each of which will be modeled independently using PMF. Neural networks will be trained to classify the replicate factors based upon chemical profiles so factor orderings can be aligned across results associated with the replicate datasets. Factor contribution uncertainty will be assessed from the distribution of contributions associated with each factor.

2.3 Health Endpoints

Only a brief overview of the health endpoints that will be included in the initial DASH health analyses will be presented here. Endpoints include counts of daily mortality and daily hospitalizations, and measures of asthma control in a panel of asthmatic school children. Daily total and cause-specific mortality counts for 5-county metropolitan Denver are obtained from the Colorado Health Information Dataset compiled by the CDPHE. Medicare data on daily non-elective (i.e., emergency and urgent) hospitalizations for the population age 65 years and older are obtained from the Medicare Provider Analysis and Review (MedPAR) file and purchased from the Centers for Medicare and Medicaid Services (CMS; formerly Hospital Care Financing Administration [HCFA]). Specific admission diagnoses of interest will be respiratory hospitalization, including the subcategories of asthma and chronic obstructive pulmonary disease, and cardiovascular hospitalization, including the subcategories of myocardial infarction and congestive heart failure

An ongoing panel study of grade school children with asthma in Denver is used to provide daily asthma health endpoint data (Rabinovitch et al., 2004; Rabinovitch, Strand and Gelfand, 2006). Children aged 6–13 who attended the Kunsberg School at the National Jewish Medical and Research Center (NJMRC) and had physician-diagnosed asthma were studied over 3 consecutive winters. In the first winter (2002–2003), 57 schoolchildren participated, 43 participated the next year and 45 children participated in the final year. Endpoint measures include recording of daily asthma medication use and daily measurement of level of lung function.

3. Results

3.1 PM_{2.5} Characterization

Data on $PM_{2.5}$ mass concentration and an abbreviated set of speciated components from the start of monitoring in July 2002 through December 2006 are available at this time. Table 1 shows descriptive data on 24-hour average concentrations of $PM_{2.5}$, inorganic ions and bulk carbon. These data, together with the 2003 supplemental metals data, were used to generate the annual average $PM_{2.5}$ chemical composition chart shown in Figure 2. In this figure, OC has been converted to organic carbon mass by multiplying by a factor of 1.41 required for

mass closure. This factor agrees well with the 1.2 - 1.4 range frequently reported in the literature, originally proposed by White and Roberts (1977) and is within the range of 1.6 ± 0.2 reported for urban fresh aerosols (Turpin and Lim, 2001; Pang, Turpin and Gundel, 2006).

To assess the adequacy of our residential monitoring site in reflecting temporal patterns in $PM_{2.5}$ components across the Denver metropolitan area, $PM_{2.5}$ component time series data were examined from the only other site in Denver in which at least a subset of these data is currently available. This monitoring site is an EPA Speciation Trends Network (STN) site and is located in the most heavily-industrialized area in Denver (the Commerce City area in northeast Denver) which also has a dense network of interstate highways. Of any area in Denver, temporal variability in PM components in Commerce City might be anticipated to be most different from that at our residential monitoring site. Because the STN analysis uses a different ECOC analysis method than the DASH core site analysis, and these have well documented differences (Chow et al., 2004, 2005), conclusions regarding comparisons between the two sites must be made guardedly. While correlations between PM components over time at the Commerce City site and our residential site were relatively good (lowest r=0.70), concentrations of all components were generally higher in Commerce City, especially so for EC and OC (Figures 3 and 4). Also, the absolute differences in concentrations for some components varied over time. For example, EC concentrations were dramatically higher in the winter in Commerce City compared to those at our residential site, but only modestly higher in other seasons (Figure 4).

3.2 Source Apportionment Model Assessment

To evaluate the performance of the PMF model using the synthetic data set, PMF modeled estimates of factor contributions and profiles (G^{pmf} and F^{pmf} , respectively) were compared with the known synthetic matrices, G^{syn} and F^{syn} . In all the model runs, PMF2 was run in robust mode. Different numbers of factors and values of the rotational factor (called FPEAK) were investigated to obtain an optimal solution based on inspection of the scaled residuals and the rotational uncertainty matrix (ROTMAT) output by PMF. This approach is described further in Brinkman et al. (2006). The optimum model was able to discern all nine of the original input factors and was relatively insensitive to selection of FPEAK values ranging from 0.0 to 0.4.

Table 2 lists the coefficient of determination (\mathbb{R}^2) between the synthetic and modeled factor contributions, G^{syn} and G^{pmf} , for a nine factor solution with an FPEAK value of zero. The PMF source contributions correlated well for six of the nine sources, all with \mathbb{R}^2 values greater than 0.79. The correlation was best for ammonium nitrate, wood combustion and vegetative debris (all with \mathbb{R}^2 greater than 0.98). The worst correlation was for meat cooking ($\mathbb{R}^2 = 0.14$). This is a result of a relatively high detection limit for cholesterol, the primary organic marker for meat cooking. Gasoline exhaust and natural gas combustion only correlated marginally well with \mathbb{R}^2 around 0.5. Table 2 also lists the normalized mean bias (NMB) for the contributions of each of the k sources defined by:

 $NMB_{k} = \frac{\sum_{i=1}^{n} \left(g_{i,k}^{syn} - g_{i,k}^{pmf} \right)}{\sum_{i=1}^{n} g_{i,k}^{syn}}$

Equation 7

In Equation 7, g^{syn} and g^{pmf} are the individual components of factor contribution matrices G^{syn} and G^{pmf} defined above.

Source contributions estimated by PMF matched those used to develop the synthetic data for paved road dust, ammonium sulfate and diesel exhaust. PMF overestimated source contributions for ammonium nitrate, natural gas combustion, vegetative debris, wood combustion, and meat cooking, and underestimated source contributions for gasoline vehicle exhaust.

4. Discussion

The DASH study was designed primarily to allow investigation of the effect of short-term variation in ambient source contributions of $PM_{2.5}$ on several health endpoints. To this end, daily concentrations of an array of $PM_{2.5}$ components are being measured at a centrally located residential site chosen to be distant from large emission sources. A concerted effort is made to measure a large number of organic components, with a commensurate lesser effort made to measure trace metal components. This detailed $PM_{2.5}$ characterization provides the raw data for source apportionment and should provide sufficient information to resolve multiple source types. A multivariate, receptor-based approach (PMF) was chosen for the daily source apportionment and a novel method was developed to estimate uncertainties based on a bootstrap approach (Hemann et al., 2008).

The ability to identify $PM_{2.5}$ source contributions depends critically on the $PM_{2.5}$ components that make up the daily multivariate measurement. The characterization of $PM_{2.5}$ in this study is more detailed than has previously been carried out for this purpose. The more fully $PM_{2.5}$ is characterized (i.e., the more source tracer compounds that are quantified), the more likely PMF will be able to identify unique factors that are interpretable as individual pollution sources. The synthetic data exercise demonstrated that PMF could discern the sources of interest using realistic estimates of source variability and profiles, though not all source contributions were accurately estimated. The sources exhibiting the most bias (based on the normalized mean bias estimates in Table 2) suffered from either poor detection limits for important tracer species (e.g., meat cooking identified by the fatty acids listed in Table S2) or highly correlated source contributions (e.g., ammonium nitrate and wood combustion, R=0.82). This exercise motivated us to improve detection limits and incorporate more organic molecular marker compounds, thereby providing additional information to assist PMF in distinguishing different, highly correlated sources. Table S1 in the online supplementary data includes the expanded list of species currently being quantified.

The synthetic data exercise also allowed us to compare our chemical speciation scenario which included organic markers with other scenarios that lacked these compounds. Replacing the organic markers with trace metals did not provide sufficient information for PMF to successfully pick out all nine of the original factors, often combining several of the important sources into one factor. The STN typically generates speciated $PM_{2.5}$ data for only one out of every three or six days and includes trace metals, but no organic markers. Data from the DASH study therefore have distinct advantages over existing STN data for longitudinal studies of daily health endpoints, both in regards to the frequency of $PM_{2.5}$ speciation measurements and the array of species identified.

Because source apportionment models are complex and subject to user discretion, critical evaluation of the results is important. Each of the PMF model source contributions will be scrutinized to be sure they match hypothesized temporal and spatial expectations. For example, we expect a diesel vehicle source contribution on average to drop on Saturday and Sunday relative to the rest of the week reflecting the reduced vehicle miles traveled on weekends (Chinkin et al., 2003; Dreher and Harley, 1998). We also expect a wood combustion source contribution to increase during the winter months from residential wood burning and possibly to reveal episodic spikes in the warmer months due to wildfire activity

and prescribed burns. Finally, simple wind trajectory analyses using meteorological data from a nearby monitoring station will be helpful in validating potential point sources identified by PMF.

The time series of $PM_{2.5}$ source concentrations at our residential monitoring site needs to be correlated with time series of population exposures to these emission sources. The ability of a single monitoring site to adequately reflect the time series of $PM_{2.5}$ components and sources for the Denver population might be questioned. Based on limited data on $PM_{2.5}$ mass concentrations from monitoring sites in Denver available at the planning stage of the study, it appeared that the $PM_{2.5}$ time series were reasonably homogeneous over space in Denver. Recent findings from studies in other cities in which PM components have been measured at multiple sites over time have indicated that this may not be true for $PM_{2.5}$ components (Marmur et al., 2006; Ito, Xue and Thurston, 2004; Kim et al., 2005). Also, examination of more recent concurrent $PM_{2.5}$ component time series data from the only other speciation site in Denver in which at least a subset of components is available indicates that there may be substantial spatial-temporal variability in $PM_{2.5}$ components.

It is possible that spatial variability in $PM_{2.5}$ component time series will bias the health effect estimates. A simple offset of concentration level from site to site, with differences in concentrations remaining constant over time and the shape of the time series preserved over space, does not result in bias in time series health effect estimates (Zeger et al., 2000). However, a difference in the shape of the time series can result in bias. In the final year of the study, we are planning a limited and focused $PM_{2.5}$ monitoring campaign in the Denver area to allow an assessment of spatial-temporal variability of short-term concentrations of the $PM_{2.5}$ components and the source concentrations. Simulation studies are also planned to provide insight into the degree of bias in the health effect estimates likely to be produced by such variability.

Identifying adverse health impacts of specific emission sources has potential advantages over identifying impacts associated with other measures of $PM_{2.5}$, such as mass or component concentrations. Emission sources are a main determinant of chemical composition and other features of $PM_{2.5}$. If these features in turn determine toxicity, then the toxicity of the $PM_{2.5}$ mix in a given setting is in large part determined by the relative contribution of individual emission sources. Policies aimed at reducing exposure to specific sources could then be considered as more focused and efficient alternatives to ameliorating the adverse public health impacts of $PM_{2.5}$ exposure than existing approaches that focus on other $PM_{2.5}$ metrics. Identifying especially toxic sources of $PM_{2.5}$ may also provide insight into features that confer toxicity. Caution should be exercised, however, given the potential for bias in the source estimates generated from the factor analysis. The synthetic data exercise carried out in this paper suggests that bias can be large if sources do not have well characterized marker species or are highly correlated with other sources.

Another advantage of taking a source-oriented approach to specifying exposure to air pollution is that specific source effects might in fact be generalizable. Many pollution sources present in Denver, such as diesel, gasoline and natural gas combustion, residential wood burning, and food cooking are common to many urban areas. While there is clearly heterogeneity in single pollutant effects from city to city when using the traditional air pollutant concentration approach to studying air pollution effects, as well as limited generalizability of single city findings, it is not known whether such heterogeneity will be present in estimated effects of pollution sources. Because a given urban area under study does not experience pollution from all potential sources does not seriously undermine this approach. Of course, if effects are due to synergistic interactions of source emissions, only limited generalizations across cities would be possible. For this reason, it could be valuable

to incorporate several cities into a unified sources-effects analysis. The lack of the needed detailed daily monitoring data, such as those being obtained in this study, currently limits the feasibility of a multi-city approach.

The initial array of health endpoints being examined in the DASH study should provide insight into the range of health effects associated with $PM_{2.5}$ source exposures. Many other possible endpoints could be coupled with the existing source exposure characterization to greatly enhance the value of the original study. The air monitoring, chemical speciation and source apportionment carried out in DASH can therefore be a valuable resource for further epidemiological investigation into health effects of $PM_{2.5}$ source emissions.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

Shown is a map of the Denver metropolitan area, including the five counties included in the DASH study and the locations of the residential monitoring site (star) and the STN site in Commerce City (circle).



Figure 2.

Average composition of $PM_{2.5}$ incorporating 4.5 years of speciated measurements at the DASH residential monitoring site. Organic carbon has been converted to organic carbon mass by multiplying by a factor of 1.41 required for mass closure.



Figure 3.

Correlation (r=Pearson correlation coefficient) between daily $PM_{2.5}$ mass and component concentrations at the northeast Denver STN site in Commerce City and the DASH residential site on Palmer Elementary School from 07/01/02 through 12/31/05.



Figure 4.

Daily 24-hour average $PM_{2.5}$ mass and component concentrations ($\mu g/m^3$) at the northeast Denver STN site in Commerce City (red circles) and the corresponding concentrations at the DASH residential site on Palmer Elementary School (black triangles). The smooth curves are loess smooths with windows widths of 0.1.

Table 1

Descriptive data on PM2.5 and PM2.5 component mass concentrations, July 1, 2002 – December 31, 2006.

					Ā	ercentil	e	
	Z	Mean	SD	Minimum	25 th	50^{th}	75 th	Maximum
$PM_{2.5} \ (\mu g/m^3)$	1628	7.81	4.93	-1.17	4.79	6.78	9.24	40.53
sulfate $(\mu g/m^3)$	1628	1.10	0.97	0.02	0.54	0.90	1.33	11.04
nitrate $(\mu g/m^3)$	1628	0.98	1.80	0.00	0.12	0.22	0.93	16.48
EC $(\mu g/m^3)^I$	1586	0.55	0.37	0.00	0.30	0.46	0.69	3.13
OC $(\mu g/m^3)^I$	1586	3.06	1.39	0.23	2.13	2.86	3.76	12.65
1		1						

EC = elemental carbon; OC = organic carbon

Assessment of the modeled factor contributions in predicting synthetic factors.

Factor	Source	$\underline{\mathbf{R}}^2$	<u>Normalized Mean Bias (%)</u>
1	ammonium nitrate	1.00	-54
2	ammonium sulfate	0.79	6
3	gasoline exhaust	0.52	76
4	diesel exhaust	0.82	-22
5	road dust	0.89	34
9	wood combustion	0.98	-143
7	meat cooking	0.14	-101
8	natural gas combustion	0.49	-57
6	vegetative debris	0.99	-78
* see text E	iquation 7		