Although the association between exposure to ambient fine particulate matter with aerodynamic diameter < 2.5 μm (PM$_{2.5}$) and human mortality is well established, the most responsible particle types/sources are not yet certain. In May 2003, the U.S. Environmental Protection Agency’s Particulate Matter Centers Program sponsored the Workshop on the Source Apportionment of PM Health Effects. The goal was to evaluate the consistency of the various source apportionment methods in assessing source contributions to daily PM$_{2.5}$ mass–mortality associations. Seven research institutions, using varying methods, participated in the estimation of source apportionments of PM$_{2.5}$ mass samples collected in Washington, DC, and Phoenix, Arizona, USA. Apportionments were evaluated for their respective associations with mortality using Poisson regressions, allowing a comparative assessment of the extent to which variations in the apportionments contributed to variability in the source-specific mortality results. The various research groups generally identified the same major source types, each with similar elemental makeups. Intergroup correlation analyses indicated that soil-, sulfate-, residual oil-, and salt-associated mass were most unambiguously identified by various methods, whereas vegetative burning and traffic were less consistent. Aggregate source-specific mortality relative risk (RR) estimate confidence intervals overlapped each other, but the sulfate-related PM$_{2.5}$ component was most consistently significant across analyses in these cities. Analyses indicated that source types were a significant predictor of RR, whereas apportionment group differences were not. Variations in the source apportionments added only some 15% to the mortality regression uncertainties. These results provide supportive evidence that existing PM$_{2.5}$ source apportionment methods can be used to derive reliable insights into the source components that contribute to PM$_{2.5}$ health effects.

Key words: fine particles, health effects, mortality, particulate matter, source apportionment, sulfate, timeseries, uncertainty. Environ Health Perspect 113:1768–1774 (2005). doi:10.1289/ehp.7989 available via http://dx.doi.org/ [Online 1 September 2005]
toxicologic evaluation of the extent to which the toxicity of ambient PM mass varies by particle type and source.

Because source composition and/or physical properties of particles vary between different source categories, the mass can be statistically apportioned into contributions from various source categories, opening the possibility of evaluating PM component effects using epidemiologic methods presently used on the PM mass. As discussed by Hopke et al. (in press), this area of research, called receptor modeling, has been active for over 3 decades. A number of accepted methods are being used to apportion the total mass into source categories, and these source apportionment methods can now be used as inputs to epidemiologic models of the human health effects of air pollution. However, to date only a small number of published efforts have related source-apportioned PM impacts to human health effects (e.g., Laden et al. 2000; Mar et al. 2000; Ozkaynak and Thurston 1987). The effect of the imputation of these apportionments on the ability of epidemiologic methods to evaluate the health effects associated with various PM components is uncertain. Because a number of methods are used to determine source contributions to PM mass impacts, and application of these methods varies among researchers, their application, although providing new insights, can also be expected to introduce added uncertainty into the derivation of estimates of PM toxicity [e.g., to the estimation of mortality relative risks (RRs) per amount of mass of fine particulate matter $\leq 2.5 \mu m$ in aerodynamic diameter ($PM_{2.5}$)]. The scientific and regulatory community is uncertain whether meaningful and reliable source apportionments of $PM_{2.5}$ health effects are possible with today’s data and methods. A workshop was therefore organized by a consortium of U.S. Environmental Protection Agency (EPA) PM centers to assess the extent to which variations in current source apportionment methods and their application may affect the ability of epidemiologic studies to discern PM health effects on a source-specific basis.

On 29–30 May 2003, the U.S. EPA PM centers sponsored the Workshop on the Source Apportionment of PM Health Effects, hosted by the New York University (NYU) PM Research Center. The specific goal of this workshop was to evaluate the variability of the various PM source apportionment approaches in assessing PM health contributions to ambient $PM_{2.5}$ concentrations in real-world data sets and to then assess the influence of this variability on the ability of statistical time-series analyses to discern which source categories contribute significantly to daily $PM_{2.5}$ mass–mortality associations. No new health or environmental data were generated by participants during this effort. Instead, the same pre-existing reference PM mass and constituent data sets from two cities (Washington, DC, and Phoenix, AZ) were sent to various leading source apportionment research groups in advance of the workshop (in December 2002), and each group individually analyzed the same data sets for daily source $PM_{2.5}$ contributions. These various daily $PM_{2.5}$ mass apportionments were then independently submitted before the workshop (in April 2003), and each was individually evaluated for their respective associations with daily mortality in each city in a consistent manner across the various apportionment research groups/methods. The PM–mortality health effects time-series modeling evaluations were conducted for the Washington and Phoenix data sets by researchers at the NYU and University of Washington U.S. EPA PM Research Centers, respectively. Washington and Phoenix were selected for this workshop analysis because the PM data available from these cities in past years were collected and analyzed for trace constituents in a manner similar to that used by the U.S. EPA in the nationwide Speciation Trend Network (STN), so as to come to workshop conclusions relevant to that developing STN data set. In addition, the consideration of these two very different cities with differing sources and weather provided a broader test of the consistency of these methods than would a single city or two cities from the same region of the country. Keeping the health effects model consistent across the various source apportionment researchers and methods allowed a separate discernment of the extent to which variability in the source apportionment step contributed to variability in the ultimate health effects analyses results.

The goals of the workshop were to bring together key researchers to assess the reliability of source apportionment–health effects methods by analyzing daily mortality with existing $PM_{2.5}$ data sets similar to those now being collected by the U.S. EPA Specialization Network.

Table 1. Summary of workshop goals and participating research institutions.

<table>
<thead>
<tr>
<th>Workshop goals</th>
<th>Participating research institutions</th>
</tr>
</thead>
<tbody>
<tr>
<td>To bring together key researchers to assess the reliability of source apportionment–health effects methods by analyzing daily mortality with existing $PM_{2.5}$ data sets similar to those now being collected by the U.S. EPA Specialization Network.</td>
<td>Brigham Young University (BYU)</td>
</tr>
<tr>
<td></td>
<td>Clarkson University (CU)</td>
</tr>
<tr>
<td></td>
<td>Harvard University (HU)</td>
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<td></td>
<td>New York University (NYU)</td>
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<td></td>
<td>University of Rochester and GSF (UR/GSF)</td>
</tr>
<tr>
<td></td>
<td>University of Southern California (USC)</td>
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<tr>
<td></td>
<td>University of Washington (UW)</td>
</tr>
</tbody>
</table>

GSF, German National Research Center for Environment and Health.

In Washington the $PM_{2.5}$ samples were collected on Wednesdays and Saturdays at the Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring site located in downtown Washington. A total of 718 samples were collected between 31 August 1988 and 31 December 1997. Integrated 24-hr $PM_{2.5}$ samples were collected on Teflon.
nylon, and quartz filters. The Teflon filters were used for mass concentrations and analyzed via particle-induced X-ray emission for the elements Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn; via X-ray fluorescence for elements Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rh, Sr, Y, Zr, Mo, Rh, Pd, Ag, Cd, Sn, Sb, Te, I, Cs, Ba, La, W, Au, Hg, and Pb; and via proton elastic scattering analysis for elemental hydro.

The nylon filter was analyzed by ion chromatography for sulfate, nitrate, and chloride. The quartz filters were analyzed by the IMPROVE method for temperature-resolved organic and EC fractions and the gaseous pollutants. Such FA and principal components analysis (PCA) models attempt to simplify the description of a system by determining a minimum set of basis vectors that span the data space to be interpreted. In other words, a new set of variables is found as linear combinations of the measured variables so that the observed variations in the system can be reproduced by a smaller number of these causal factors. This approach has been widely used in studies of airborne PM composition data (Gao et al. 1994; Hopke et al. 1976; Roscoe et al. 1982).

Traditional FA and PCA are useful for identifying source components contributing to the PM mass but do not directly provide an apportionment in the form presented above. However, the solutions can be manipulated to provide such a quantitative solution. One approach is specific rotation FA (Koutrakis and Spengler 1987), which uses a targeted Procrustes factor rotation. An alternative approach, absolute principal-component analysis (APCA) (Thurston and Spengler 1985), has also been used to produce quantitative apportionments. Two more-recent approaches are Unmix (Henry and Kim 1999; Kim and Henry 1999, 2000a, 2000b) and positive matrix factorization (PMF) (Paatero 1997; 1999; Paatero et al. 2002). These and similar multivariate techniques, described and documented in more detail in Hopke et al. (in press), have been applied by the different research groups to achieve source apportionments of the Washington and Phoenix PM$_{2.5}$ data sets (Table 2).

After all the estimated source-specific impact assessments were submitted by workshop participants, the agreement across source apportionment analyses was evaluated. This was first evaluated by an intercomparison of the various analyses’ respective mean
estimates of source-specific mass impacts in each city. In addition, as the various source apportionment results were to be employed as inputs into a daily time-series mortality analyses, the time-series intercorrelations of their respective daily estimates of source impacts were also evaluated and intercompared across source categories in each city.

**Health effects modeling.** After the source apportionments were submitted, all Washington and Phoenix daily source apportionments were provided to K. Ito of the NYU PM Center and T. Mar of the University of Washington PM Center, respectively, for inclusion in time-series mortality models to assess the resulting variations in their source-specific health effects estimates (RRs). The city-specific mortality models employed are described below.

The model-building steps of the Washington time-series mortality model development used in these analyses (Ito et al., unpublished data) were designed to be similar to those used in past studies of PM$_{2.5}$ mass as, follows:

- We first developed the base mortality model as a function of season and other temporal trends in Poisson generalized linear models (GLMs) (McCullagh and Nelder 1989). Using natural splines, we fit a smooth function of time to mortality to adjust the model for seasonal trends and unmeasured seasonal confounders, such as influenza epidemics. The inclusion of this term also reduces undesirable residual autocorrelation and overdispersion in the mortality regression, so the choice of the spline degrees of freedom (df) for smoothing of time (df = 38, or 4 per year) was based both on the fit to the mortality series and minimization of autocorrelation of the model residuals.
- Weather variables and a day-of-week variable were then also incorporated into the base model, consistent with past general practice in PM$_{2.5}$ modeling, including a) natural splines of the same-day temperature with 4 df to fit “hot” temperature effects; b) natural splines of the average of lags 1 through 3 of daily temperature (i.e., up to 3 days before the date of death) to fit “cold” temperature effects; and c) an indicator for “hot” (daily mean temperature > 80°F) and “humid” (daily relative humidity > 70%) days to fit the interaction. The end result of this step was a base model to which air pollutant variables could be added and evaluated.
- To the base model, each of the alternative source components was individually added (for each research group/method) to separately test the individual associations of each source category with mortality, after controlling for the variables considered in the base model. The RR associated with both an interquartile (25th to 75th percentile) and a 5th- to 95th-percentile increase in the source estimate was computed for lag days 0 to 5 for each of the source apportionment analyses. This approach provided directly comparable mortality effect estimates for each source category and for apportionment modeling results of participating groups.

The basic steps of time-series model development used in the Phoenix analyses (Mar et al., in press) were the same as for Washington. Similarly, associations between source contributions and cardiovascular and total nonaccidental mortality were analyzed using Poisson GLMs in S-PLUS 2000 (Insightful Inc., Seattle, WA, USA). The same Phoenix base mortality model was applied to source apportionment analyses of all groups to provide a consistent basis for comparison across source components and groups (i.e., to eliminate model specification variability from the analysis). The base model controlled for extreme temperatures using an indicator variable, mean temperature, relative humidity, day of week, and time trends. Natural spline smoothers were used for time trends, temperature, and relative humidity. We applied 12 df for the smoothing of time trend (i.e., 4 df per year). The degrees of freedom for the natural splines for time trends were selected to minimize autocorrelation in the residuals and the Akaake Information Criterion (AIC) (Akaike 1974). For the analysis of cardiovascular mortality, 5 spline df and 2 days lag for temperature were incorporated, based on past experience with models of PM$_{2.5}$ and mortality in this city. For the total mortality analysis, 5 spline df and 1 day lag for temperature were employed, and 2 df for the smoothing of relative humidity with 0 days lag for both the cardiovascular and total mortality analyses. The degrees of freedom and the lags were chosen to minimize the AIC. As in the case for the Washington analyses, the respective estimated source contributions of the various research groups were added to this base model, in turn, as the particle pollution variable. The RR associated with both an interquartile (25th to 75th percentile) and a 5th- to 95th-percentile increase in the source estimate was computed for lag days 0 to 5 for each of the source apportionment analyses. Again, this consistent mortality analysis approach across source apportionments allowed a direct comparison of the daily mortality effect estimates across the various source apportionment analyses in each city.

Finally, we evaluated the size and significance of the additional variability introduced to the PM–mortality, time-series analysis by variations in the source apportionment process across groups and methods, consistent with the primary goal of this workshop. To this end, the various source apportionments’ resulting mean mass contributions and estimated percent excess deaths per 5th- to 95th-percentile increment increase by source-apportioned PM$_{2.5}$ were intercompared and then analyzed (within each city) by analysis of variance (ANOVA) and a GLM. This allowed us to compare variations in model estimates that were due to “between-source” versus “within-source” (i.e., variation due to different analyses).

**Results**

**Source apportionment intercomparisons.** As described in Hopke et al. (in press), the various source apportionment analyses from each of the participating research groups were intercompared in two ways: a) by comparing the mass contributions attributed to each of the sources; and b) by calculating the correlation coefficients between the source contributions from PM$_{2.5}$ from the various groups within source groups. The solutions of the various groups were compared with each other on an equal basis, because an accepted “gold standard” method does not exist at this time for source apportionment. Table 2 notes the source apportionment analyses performed on these data sets. Figures 1 and 2 present the means and distributions of the resulting PM$_{2.5}$ mass source apportionment for each source category in Washington and Phoenix, respectively. Most groups were able to identify the same major sources in their source apportionment analyses of the trace constituent data. However, not all sources were identified by all researchers; some groups did not provide impacts for all possible source categories. Two researchers from BYU contributed separate analyses: Eatough (BYU1) and Christensen (BYU2). In this plot, when researchers broke out the source impacts differently from other researchers (e.g., when secondary sulfates were broken into sulfates 1 and sulfates 2, or traffic was subdivided into categories, such as diesel vs. gasoline fueled motor vehicles), the results have been grouped to provide more directly comparable totals. The mass apportionment uncertainties included in Figures 1 and 2 visually indicate an overall consistency in impacts by source category, as they provide confidence intervals (CIs) that overlap across analyses of the various groups, especially for the larger mass contributors. To be more quantitative, we conducted an ANOVA F-test of the within-source versus between-source variations for each of the major source categories in Figures 1 and 2. The results indicated significantly greater variability ($p < 0.001$) across source categories than across investigators/methods (i.e., investigator/method variations were small compared with source-to-source variations). Overall, these plots and statistical analyses indicate that, although the estimated mass impact results vary across analyses and not all sources were identified by all investigators (especially in the case of the smaller mass impact sources), there is both
qualitative and quantitative consistency in the major PM$_{2.5}$ contributing sources identified and their mass impacts across the independent analyses of these data by the various research groups and apportionment methods.

Because these apportionment results were to be applied in time-series analyses, another evaluation of the consistency of the source apportionments across research groups and apportionment methods was conducted. Variability was examined in the paired correlations of estimated daily source apportionment mass contributions in the various analyses over time and within each city. As shown in Figure 3A for Washington and Figure 3B for Phoenix, the sulfate-containing, crustal, and nitrate components exhibited among the highest mean intercorrelations across the various research groups in these cities. Among the chief PM$_{2.5}$ mass contributors (Figures 1 and 2), the weakest cross-analyses correlations in Figure 3 were usually found for the sources with the greatest uncertainty in their composition (i.e., lacking unique constituents for unique identification), notably, traffic and wood burning in Washington, and wood burning and metals in Phoenix.

**Time-series mortality effect estimate intercomparisons.** The source apportionment results for each group were combined with the mortality data in Washington and Phoenix, and time-series mortality regressions were then run (Ito et al., in press; Mar et al., in press). Figure 4 displays the resulting mean RR estimates and 95% CIs of cardiovascular (CV) and total daily mortality for each major source category identified in Washington and Phoenix for the overall workshop estimate, with source apportionment interanalysis variation excluded and interanalysis variation included. Results were derived using the lag of maximum association in each analysis. It is clear from the comparisons that the variability introduced by the uncertainty of the across-source apportionments and analyses is small, relative to the overall uncertainty of these estimates. In quantitative terms, the percent increase in the uncertainty (i.e., in the CI) for the mortality RR of each displayed source category in Washington added by the interanalysis variability was as follows: soil (23% for CV, 18% for total); traffic (12% for CV, 16% for total); and sulfate (25% for CV, 26% for total). In the Phoenix mortality analyses, the percent increase in the uncertainty (i.e., in the CI) for the mortality RR of each displayed source category added by the interanalysis variability was as follows: soil (4% for CV, 7% for total); traffic (6% for CV, 33% for total); and sulfate (7% for CV, 5% for total). Thus, while the uncertainty added by the differences in source apportionments varies from source to source in these cases, the overall average increase is about 15%, which suggests that the error added by variability in source apportionment approach is quite small relative to the baseline uncertainty inherently associated with making these time-series pollution RR estimates.

The between-source variation in these daily mortality RRs was also compared with within-source variations (variation due to different analyses). As shown in Table 3, significantly larger variation was found between

![Figure 1](image1.png)

**Figure 1.** Mean, interquartile range (box), and range (maximum–minimum) of mass impacts predicted by each research group’s source apportionment analysis of the Washington PM$_{2.5}$ data set. MR, multiple regression. (A) Soil; (B) nitrates; (C) traffic; (D) wood burning; (E) secondary SO$_x$; (F) residual oil; (G) sea salt; (H) incinerator.

![Figure 2](image2.png)

**Figure 2.** Mean, interquartile range (box), and range (maximum–minimum) of mass impacts predicted by each research group’s source apportionment analysis of the Phoenix PM$_{2.5}$ data. (A) Soil; (B) secondary SO$_x$; (C) traffic; (D) metals/industry/smelter; (E) vegetation/wood burning; (F) sea salt.
Overall, these results indicate that
in both cities and death categories (was a statistically significant predictor of RR
ences), whereas the between-source variation
from 0.38 to 0.65 for between-group differ-
categories in both cities (with was a nonsignificant predictor for both death
press). In the GLM, between-group variation
(in a GLM) of the individual investigator esti-


Figure 3. Box and whisker plots of the distributions of temporal correlation coefficients (r) between all possible pairs of similar source contributions resolved for (A) Washington and (B) Phoenix.

Figure 4. Mean RR estimates and 95% CIs for each major source category in Washington (A) cardiovascular and (C) total nonaccidental mortality, and Phoenix (B) cardiovascular and (D) total nonaccidental mortality for the overall workshop estimate, with source apportionment interanalysis variation excluded and with the interanalysis variation included.

Table 3. ANOVA analysis of source-specific mortality RR estimates.

<table>
<thead>
<tr>
<th>Mortality category</th>
<th>ANOVA p-value</th>
<th>Source category variance (%)</th>
<th>Research group variance (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Washington CV</td>
<td>&lt; 0.001</td>
<td>47.5</td>
<td>9.5</td>
</tr>
<tr>
<td>Washington total</td>
<td>&lt; 0.001</td>
<td>80.0</td>
<td>2.6</td>
</tr>
<tr>
<td>Phoenix CV</td>
<td>&lt; 0.001</td>
<td>76.3</td>
<td>4.5</td>
</tr>
<tr>
<td>Phoenix total</td>
<td>&lt; 0.001</td>
<td>64.8</td>
<td>8.3</td>
</tr>
</tbody>
</table>

Discussion and Conclusions
With regard to the PM$_{2.5}$ mass apportionments, the findings of this intercomparison among results from some of the leading source apportionment research groups indicate that the same major source types (those that contribute most of the PM$_{2.5}$ mass at each site), with similar elemental makeups (i.e., key tracers), are consistently identified by different groups in each city. Methods generally yielded the most consistent results (i.e., the highest correlations across groups over time) for sources with the most definable (unique) tracers or combinations of tracers in each city.

In Washington, soil, secondary sulfate and nitrate, oil burning, and incineration were most unambiguously identified by various methods; wood burning, salt, and traffic were less well correlated across analyses. In Phoenix, soil, traffic, secondary sulfate, and sea spray were most highly correlated across analyses; wood and vegetative burning, metals industry particles, and coal fly ash were less well correlated. Based on the relative sizes of these intergroup intercorrelations for each of the source types in these two cities, the soil-, sulfate-, residual oil-, and salt-associated mass components were generally seen to be most unambiguously identified by the various source apportionment methods, while vegetative burning and traffic were less well correlated across groups. However, the source mass

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impacts predicted for the various source categories were generally not significantly different from one another across the research groups, indicating consistency in the source apportionment results. The addition of further tracers/analyses may be required to improve the consistency of the less well-discriminated sources. For example, the measurement of low-volatility organic compounds has been suggested as one way to better discern traffic-related PM components (Schauer et al. 1996; Schauer and Cass 2000). Overall, however, although there are no gold standard correct answers for the source identification and apportionments in the real-world data sets considered in this workshop, the apportionment consistency in the largest PM$_{2.5}$ source contributors across researchers in these cities, often using differing statistical methods, indicates reliability in the source apportionment approach.

With regard to the health effects apportionments to the different source components of PM$_{2.5}$, the between-source variation in daily mortality RR was significantly larger than the between-research group variation in reported RRs. Thus, analysis-to-analysis variability in the source apportionments was small compared with the overall uncertainty in the mortality RR estimates. In addition, between-group variation in RR estimates was nonsignificant, whereas the between-source type variation was statistically significant. This result indicates that variations in choice of research group or source apportionment method have only a small effect on variations in the RR estimates, relative to the variations in RR caused by different source components. Indeed, in mortality categories where significant PM$_{2.5}$ mass–daily mortality associations were detected in these cities (e.g., for cardiovascular deaths in both cities), most source categories were non-significant contributors. However, the most strongly associated source categories showed statistically significant contributions. Across these two cities, the most consistently associated PM$_{2.5}$ source category was sulfate-associated mass. The source RR estimates generally had overlapping confidence bands, indicating that larger numbers of observations will be required in each of these cities to have enough power to significantly differentiate the impacts of the various source impacts. The overall source-specific RR estimates derived in this work appeared reasonable when compared with the limited source-specific mortality analyses published in the past, but many more source apportionment–mortality analyses of this type must be done before broad-based comparisons with the source-specific RR results from this workshop are possible.

Overall, the results of this intercomparison of the health effects apportionments found that variations in PM source apportionment research group or method introduced relatively little uncertainty into the evaluation of differences in PM toxicity on a source-specific basis, adding an average of only approximately 15% to the overall source-specific mortality uncertainties. Variations in these apportionment modeling choices do not prevent the consistent discernment of variations in the relative strengths of source-specific PM$_{2.5}$ mortality associations. However, the uncertainty added by the source apportionment estimation suggests that longer data records may be required for significant effects to be detectable in source-specific analyses than for PM$_{2.5}$. The conduct of daily speciation sampling (rather than every third day) in major U.S. cities would be one way to rapidly improve the power of future source apportioned PM timeseries health effects analyses. Daily sampling would also better clarify the potentially differing distributed-lag natures of the various source-specific impacts identified in this workshop. Although further research and the possible addition of more key tracers to the speciation of PM$_{2.5}$, are needed to better characterize ambient tracer profiles for sources with less well-defined compositional characteristics (e.g., for vegetation burning and traffic), the results of this workshop indicate that present-day PM$_{2.5}$ source apportionment methods can provide valuable insights into the source components that contribute most to PM$_{2.5}$-health effects associations.

**References**


