The Workshop on the Source Apportionment of PM Health Effects:

Inter-Comparison of Results and Implications

Final Report 06-01
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New York State
Energy Research and
Development Authority

NYSERDA

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The Workshop on the Source Apportionment of PM Health Effects: Inter-Comparison of Results and Implications

Final Report

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PREFACE

The New York State Energy Research and Development Authority is pleased to publish "Workshop on the Source Apportionment of particulate matter (PM) Health Effects: Inter-comparison of Results and Implications." The report was prepared by the principal investigator, Philip Hopke of Clarkson University, and George Thurston and Kazuhiko Ito of New York University.

In this study, two relatively large PM datasets from Phoenix, AZ and Washington, D.C., were analyzed using different source-apportionment approaches by seven research groups. The resulting apportionments were then evaluated for their associations with daily mortality. Overall, the results indicated that variations in choice of source apportionment method have only a small effect on variations in daily mortality relative risk, compared to the variations in relative risk caused by different source components. Additional research will be needed in identifying sources with less well-defined characteristics, such as vegetative burning and mobile sources. However, these results indicate PM source apportionment methods can be used to provide valuable insights into sources that contribute most to $PM_{2.5}$–health effects associations.

The work was funded by the **New York Energy Smart**[SM] Environmental Monitoring, Evaluation, and Protection (EMEP) Program. This study is one of a broader portfolio of research projects characterizing PM, performing source apportionment on PM datasets, and addressing policy-relevant questions for PM control strategies in New York State.

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INTRODUCTION

Current National Ambient Air Quality Standards (NAAQS) for airborne particulate matter (PM) use airborne particle mass as the indicator for making air quality determinations. However, it seems highly likely that some types of particles are more toxic than others. Thus, the focus on all particles that contribute mass may lead to inefficient and ineffective control strategies relative to being able to focus directly on those particles that cause the adverse human health effects. However, there are an enormous number of possible chemical species associated with PM. Thus, a potentially more effective approach would be to consider the airborne particulate matter as a mixture of mixtures. Various source types (spark-ignition vehicles, diesel powered vehicles, coal-, gas-, or oil-fired power plants, incinerators, etc) have characteristic chemical and/or physical patterns (mixtures). It may be useful to examine the relationship of specific source emissions to adverse health effects, since if a limited number of sources contributed significantly to the effects, more targeted control strategies could be devised to focus on those sources.

By identifying source composition or physical properties profiles, the contributions of different source types to the airborne PM mass may be apportioned. This area of research, called Receptor Modeling, has been an area of active research for over 30 years. Recently, new tools such as advanced factor analysis models have been developed that permit more effective source apportionments to be performed. At the same time, the U.S. Environmental Protection Agency (EPA) has deployed a network of particulate matter samplers that are collecting samples at urban sites across the United States and thereby producing data that can be used for source apportionment and possible time series health effects studies.

In May 2003, the EPA's PM Centers Program, in conjunction with the New York State Energy Research and Development Authority, sponsored the "Workshop on the Source Apportionment of PM Health Effects" to explore the current state of the art in receptor modeling and the potential for the incorporation of source apportionment in health effects modeling.

OBJECTIVES

The objectives of the receptor modeling and health effects modeling exercise were:

1. To ascertain if quantitative relationships can be observed between apportioned source contributions and human mortality using particle composition and mortality data from two different cities, and
2. To ascertain if different receptor modeling methods significantly affect the contribution/mortality relationships.

STUDY DESIGN

Seven research groups participated in the estimation of source apportionments of $PM_{2.5}$ mass samples from Washington, DC and Phoenix, AZ that were collected and chemically analyzed in a manner similar to that used in the EPA's Speciation Trend Network. These two data sets were chosen because they had been previously analyzed for source apportionment, and mortality data for both cities were readily available. The apportionment method applied to the $PM_{2.5}$ mass and trace constituent data varied among research groups, ranging from simple mass regressions on selected single tracer elements for each source class (e.g., vanadium for oil), to more intricate factor analysis based methods including Positive Matrix Factorization (PMF) and UNMIX. The resulting apportionments were then evaluated for their associations with daily mortality in a consistent manner in each city. This comparison allowed an assessment of the extent to which variability in the source apportionment results contributed to variability in the PM component mortality analyses results.

RESULTS

Similar source profiles were extracted from these data sets by the investigators using different factor analysis methods. There was good agreement among the major source types resolved. Analysis of Variance (ANOVA) analyses were made to intercompare the apportioned source contributions from the multiple analyses for each city. Figures S1 and S2 present the results of the ANOVA analyses.

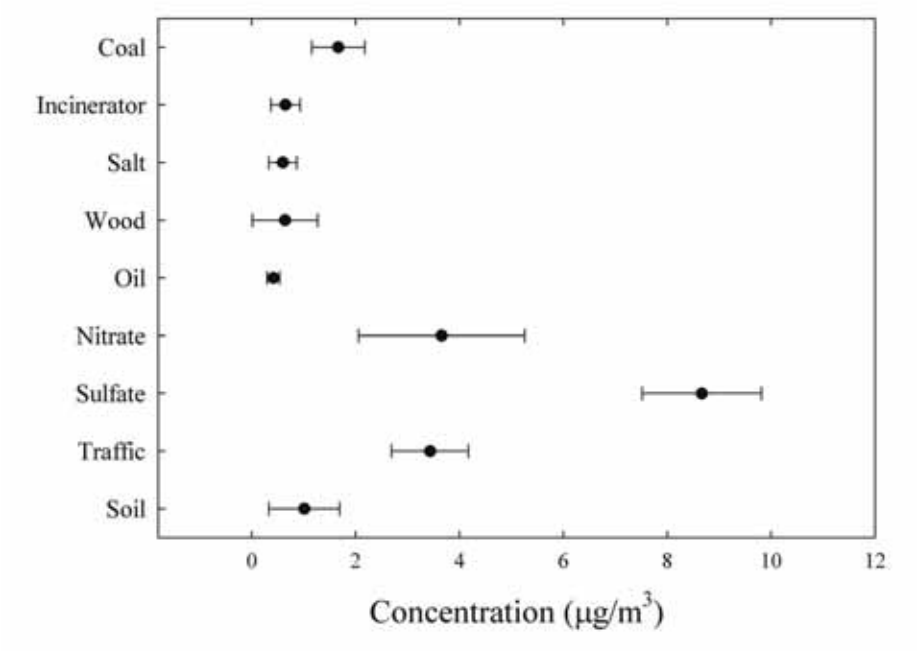

Figure S1. Plot of the ANOVA results for the Washington, DC data.

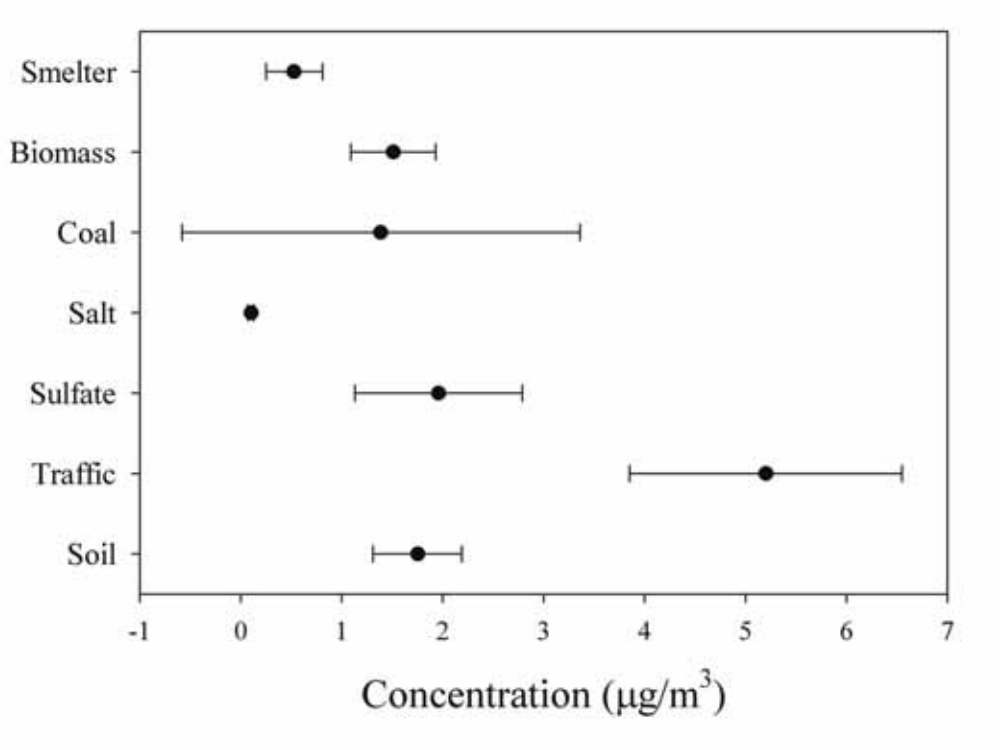

Figure S2. Plot of the ANOVA results for the Phoenix, AZ data.

Crustal (soil), sulfate, oil, and salt were the source types that were most unambiguously identified (having generally the highest correlation across the sites). Traffic and vegetative burning showed considerable variability among the results with variability in the ability of the methods to partition the motor vehicle contributions between gasoline and diesel vehicles. However, if the total motor vehicle contributions are estimated, good correspondence was obtained among the results. The source impacts were especially similar across various analyses for the larger mass contributors (e.g., in Washington, secondary sulfate standard error (SE) =7%, and 11% for traffic; in Phoenix, secondary sulfate SE=17%, and 7% for traffic). Especially important for time-series health effects assessment, the source-specific impacts were found to be highly correlated across analysis methods/researchers for the major components (e.g., mean analysis to analysis correlation, $r > 0.9$ for traffic and secondary sulfates in Phoenix and for traffic and secondary nitrates in Washington. The sulfate mean r value is > 0.75 in Washington.). Overall, although these intercomparisons suggest areas where further research is needed (e.g., better division of traffic emissions between diesel and gasoline vehicles), they provide support for the contention that $PM_{2.5}$ mass source apportionment results are consistent across users and methods, and that today's source apportionment methods are robust enough for application to $PM_{2.5}$ health effects assessments.

To assess the health effects of the apportioned source classes, a Poisson Generalized Linear Model (GLM) was used to estimate source-specific relative risks at lags 0-4 days for total non-accidental, cardiovascular, and cardio-respiratory mortality adjusting for weather, seasonal/temporal trends, and day-of-week. The plot of estimated total mortality relative risk per 5th-to-95th percentile increment in source-apportioned $PM_{2.5}$ in Washington, DC by source type and investigators/methods are shown in Figure S3. Source-related effect estimates and their lagged association patterns were similar across investigators/methods. The varying lag structure of associations across source types, combined with the Wednesday/Saturday sampling frequency made it difficult to compare the source-specific effect sizes in a simple manner. The largest (and most significant) percent of excess deaths per 5^{th}-to-95^{th} percentile increment of apportioned $PM_{2.5}$ for total mortality was for secondary sulfate (variance-weighted mean percent excess mortality = 6.7% (95% confidence interval: 1.7, 11.7)), but with an unusual lag structure (lag 3 day). Primary coal-related $PM_{2.5}$ (only three teams) was also significantly associated with total mortality with a 3-day lag, as for sulfate. Risk estimates for traffic-related $PM_{2.5}$, while significant in some cases, were more variable. Soil-related PM showed smaller effect size estimates, but they were more consistently positive at multiple lags. The cardiovascular and cardio-respiratory mortality associations were generally similar to those for total mortality. Alternative weather models generally gave similar patterns, but sometimes affected the lag structure (e.g., for sulfate). Overall, the variations in relative risks across investigators/methods were found to be much smaller than those across estimated source types or across lag days for these data.

The associations between the participant's estimated source contributions of $PM_{2.5}$ for Phoenix, AZ for the period from 1995-1997 and cardiovascular and total non-accidental mortality were also analyzed using Poisson GLM. The total mortality results are provided in Figure S4. The base model controlled for extreme temperatures, relative humidity, day of week, and time trends using natural spline smoothers. The same mortality model was applied to all of the apportionment results to provide a consistent comparison across source components and investigators/methods. Of the apportioned anthropogenic $PM_{2.5}$ source categories, secondary sulfate, traffic, and copper smelter-derived particles were most consistently associated with cardiovascular mortality. The source types with the largest cardiovascular mortality effect size were secondary sulfate (median estimate = 16.0% per 5^{th}-to-95^{th} percentile increment at lag 0 day among eight investigators/methods) and traffic (median estimate = 13.2% per 5^{th}-to-95^{th} percentile increment at lag 1 day among nine investigators/methods). For total mortality, the associations were weaker. Sea salt was also found to be associated with both total and cardiovascular mortality, but at 5 day lag. Fine particle soil and biomass burning factors were not associated with increased risks. Variations in the maximum effect lag varied by source category, suggesting that past analyses considering only single lags of $PM_{2.5}$ may have underestimated health impact contributions at different lags. Further research is needed on the possibility that different $PM_{2.5}$ source components may have different effects on lag structure. There was considerable consistency in the health effects results across source apportionments in their effect estimates and their lag structures. Variations in results across investigators/methods were small compared

to the variations across source categories. These results indicate reproducibility of source apportionment results across investigative groups and support the applicability of these methods to effects studies. The consistency among the results suggests the robustness of the source apportionment in health effects analyses, but remaining issues, including accuracy of source apportionment and source-specific sensitivity to weather models, need to be investigated.

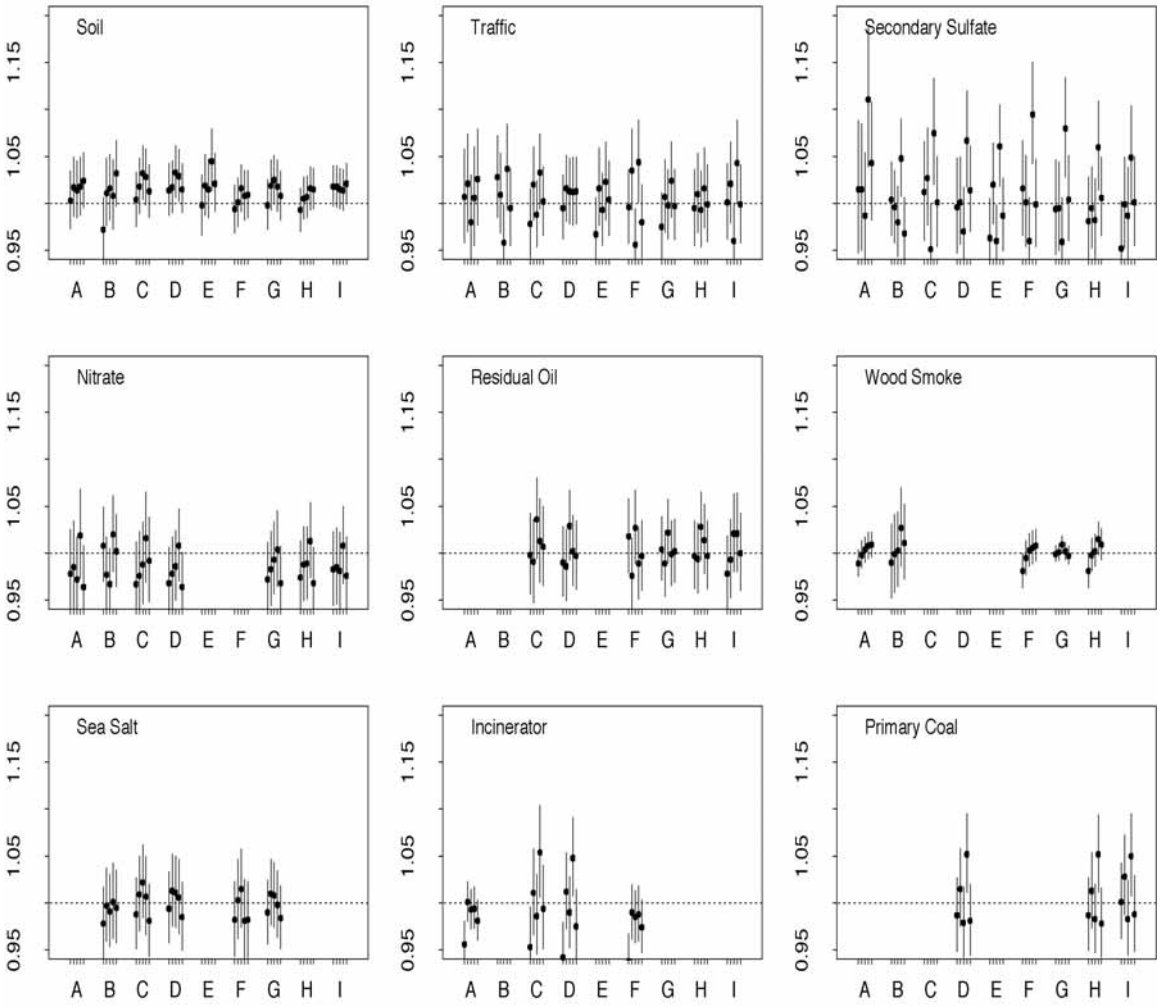

Figure S3. Estimated total non-accidental mortality relative risk per 5th-to-95th percentile increment in source-apportioned $PM_{2.5}$ in Washington, DC by source type and investigators/methods. The letters denote the various source apportionments that were provided for the health effects modeling. The five consecutive estimates are for lags 0 to 4 days.

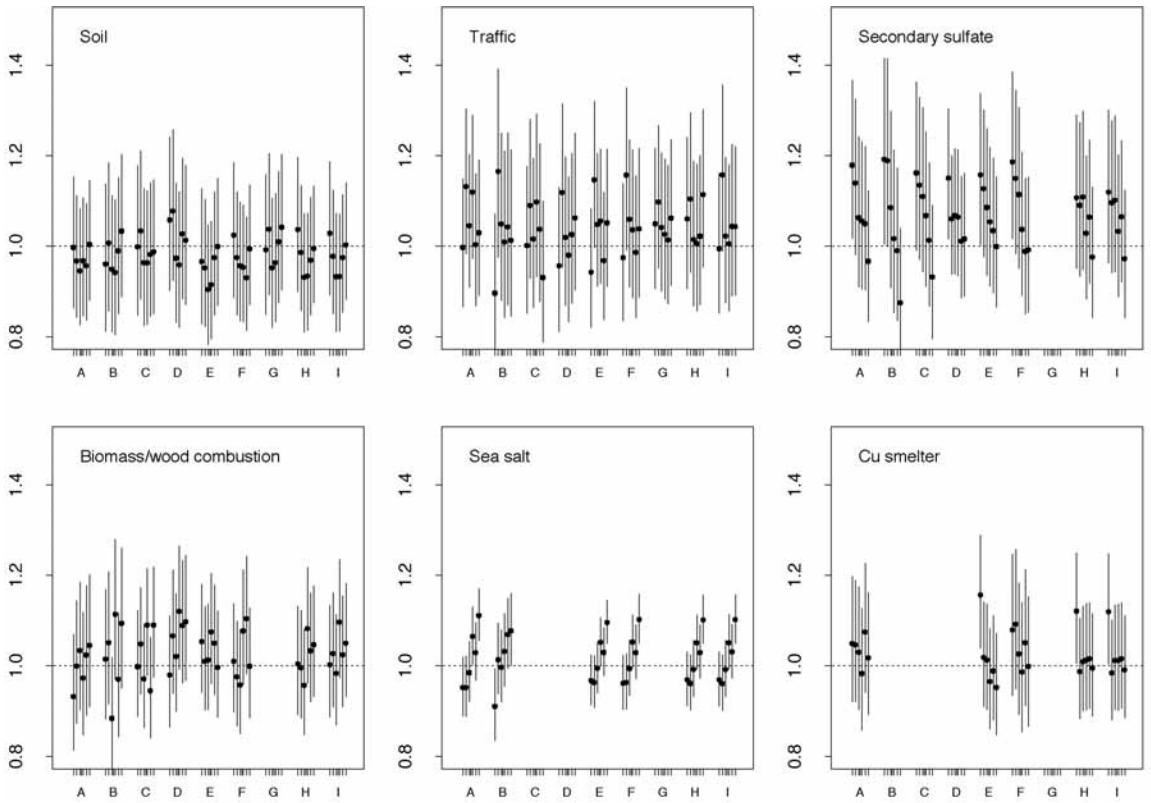

Figure S4. Estimated total non-accidental mortality relative risk per 5th-to-95th percentile increment in source-apportioned $PM_{2.5}$ in Phoenix, AZ by source type and investigators/methods. The letters denote the various source apportionments that were provided for the health effects modeling. The five consecutive estimates are for lags 0 to 4 days.

CONCLUSIONS

Many similar source types were identified even though various investigators were using different analysis methods. Crustal (soil), sulfate, oil, and salt were most unambiguously identified. Because of the differences in the resolution of motor vehicle sources, the individual traffic sources are not as well correlated among the results. If the total motor vehicle contributions are estimated, better correspondence among the results is obtained. While these analyses indicate that it may be possible to separate diesel from gasoline vehicle impacts, further research is needed to ascertain the degree to which the various components of motor vehicle emissions (diesel and spark-ignition) can be separately identified and quantified. However, the overall consistency by source category provided impacts that were very similar for the larger mass contributors across various analyses (e.g., in Washington, secondary sulfate SE =7%, and =11% for traffic impacts; in Phoenix, secondary sulfate SE=17%, and 7% for traffic).

Especially important for time-series health effects assessment, the source-specific impacts across analyses were also found to be highly correlated across analyses methods/researchers for the major components (e.g., mean analysis to analysis correlation, r greater than 0.9 for traffic and secondary sulfates in Phoenix and for traffic and secondary nitrates in Washington. The sulfate has mean correlation coefficients, r greater than 0.75 in Washington.). Overall, although these intercomparisons suggest areas where further research is needed (e.g., distinguishing traffic emissions between diesel and gasoline), they also provide considerable support to the contention that $PM_{2.5}$ mass source apportionment results are consistent across users and methods, and that today's mass apportionment methods are robust enough for reliable application to $PM_{2.5}$ health effects assessments.

Regarding the health effects apportionments to the different source components of $PM_{2.5}$, the between-source variation in daily mortality relative risk (RR) was much larger than the between-research group variation in reported RR's. Between group variation was found to be non-significant, while the between source type variation was statistically significant. This 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 cardio-vascular deaths in both cities), most source categories were found to be non-significant contributors, but 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.

Overall, the results of this inter-comparison 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 relative risk uncertainties. Thus, variations in these choices do not prevent the consistent discernment of variations in the strength of source-specific $PM_{2.5}$ mortality associations. However, the uncertainty that is 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 $PM_{2.5}$. Although further research is needed in defining tracer profiles for sources with less well-defined compositional characteristics (e.g., for vegetative burning and traffic) in order to allow more exact quantitative source-specific mass toxicity evaluations, these results provide substantiation that present-day $PM_{2.5}$ source apportionment methods are reliable, and can be used to provide valuable insights into the source components that contribute most to $PM_{2.5}$-health effects associations.

Section 1
INTRODUCTION

Airborne particulate matter (PM) air pollution is presently regulated by the National Ambient Air Quality Standards (NAAQS) using gravimetric mass as the particle metric to assess air quality. However, there are an enormous number of different chemical species associated with the various types of ambient particles, depending upon their source origins (e.g., see Cooper and Watson 1980). For example, primary particles emitted from coal combustion are characteristically highly enriched with arsenic and selenium, while residual oil combustion particles are more enriched in nickel and vanadium, and soil particles are especially enriched in the crustal elements (silicon, aluminum, etc.). In addition, secondary components of particles (such as sulfates, nitrates, and organic compounds) are formed in the atmosphere from gaseous pollutant emissions. These secondary components can either condense on primary particles or form secondary particles that can then collide and coagulate with primary particles. Thus, individual particles in an urban airshed can contain both primary and secondary components. The composition of ambient aerosols have therefore been found to reflect source PM emission characteristics differences over space (e.g., between cities) and time (e.g., across seasons) (e.g., see Spengler and Thurston 1983). Since the composition of particle types varies so much, it is probable that some types of particles are more toxic than others. Thus, treating all particles that contribute to the mass concentration equally in the regulatory process may lead to inefficient protection of public health, relative to focusing PM regulations and controls more directly on those particle classes that are most associated with adverse human health effects. Therefore, a potentially more effective regulatory approach would be to address the individual types of particles independently, focusing control efforts on the most toxic categories. However, because toxicities of individual source components are not yet certain, and because virtually all published PM-health effects studies to date have used PM mass (in various size categories) as the particle pollution index, the current NAAQS for airborne PM use airborne particle mass as the indicator for making air quality compliance determinations. This equal treatment of all particles that contribute to mass, irrespective of composition, may presently be leading to less optimal control strategies to avoid the adverse human health effects of PM, potentially causing the present PM ambient standard to be less protective of health in some areas of the nation than others. This indicates that there is a need for epidemiological and toxicological evaluations into the extent to which the toxicity of ambient PM mass varies by particle type and source.

The fact that the source composition and/or physical properties of particles vary between different source categories allows the mass to be statistically apportioned into contributions from the various source categories, opening the possibility of evaluating PM component effects using epidemiological methods presently used on the PM mass. As discussed in more detail in Hopke et al. (2006), this area of research, called Receptor Modeling, has been an area of active research for over three decades. There are now a number of accepted methods that are being used to apportion the total mass into source categories, and

these source apportionment methods can now be used as inputs to epidemiological models of the human health effects of air pollution. However, to date, there have only been a small number of published efforts which relate source-apportioned PM impacts to human health effects (e.g., Laden et al. 2000; Mar et al. 2000; Ozkaynak and Thurston 1987; Tsai et al., 2000), and experience is limited as to the effect the imputation of these apportionments may have on the ability of epidemiological methods to evaluate the health effects that may be associated with the various PM components. There are presently a number of methods that have been applied to determine source contributions to PM mass impacts, and there is also variability in how these methods are applied from researcher to researcher. Therefore, their application, while 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 per amount of $PM_{2.5}$). Thus, there remains uncertainty in the scientific and regulatory community 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. EPA PM Centers with the overall aim of assessing the extent to which variations in present-day source apportionment methods and their application may affect the ability of epidemiologic studies to discern PM health effects on a source-specific basis.

On May 29-30, 2003, the EPA's PM Centers, in conjunction with the New York State Energy Research and Development Authority, sponsored the "Workshop on the Source Apportionment of PM Health Effects". The workshop was 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 source contributions to ambient fine PM ($PM_{2.5}$) 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 approach taken was to provide pre-existing reference PM mass and constituent data sets from two cities (Washington, DC and Phoenix, AZ) to various leading source apportionment research groups in advance of the workshop (in December, 2002), and have each group individually analyze the same data sets for daily source $PM_{2.5}$ contributions. These various daily $PM_{2.5}$ mass source apportionments were then independently submitted prior to 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, DC and Phoenix, AZ data sets by researchers at the NYU and University of Washington EPA PM Research Centers, respectively. Washington, DC, and Phoenix, AZ were selected for this workshop analysis because PM composition data were available from these cities based on particle collection and chemical analysis in a manner similar to the methods used by the EPA in the nationwide Speciation Trend Network (STN). Thus, the conclusions from this study would be relevant to the developing data set from that network.

In addition, the consideration of these two very different cities with differing sources and weather provides a broader test of the consistency of these methods than would a single city, or would two cities from the same region of the country. In addition, both data sets had been previously examined for source apportionment. Song et al. (2001) had analyzed the Washington, DC data, and both Ramadan et al. (2000) and Mar et al. (2000) had studied the Phoenix data. 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: 1) 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 EPA STN; and, 2) to identify key future research needs for source apportionment health effects evaluation. As noted in Table 1, research groups from seven institutions (with most groups being affiliated with one of the five EPA PM Centers), using various source apportionment approaches, participated in this workshop. Thurston et al. (2005) has provided an overall summary of the workshop. The detailed comparisons of the source apportionments has been presented by Hopke et al. (2006). The health effects modeling for Washington, DC are summarized by Ito et al. (2006) while the parallel results for Phoenix, AZ are described by Mar et al. (2006).

Table 1. Participating research institutions.

PARTICIPATING RESEARCH INSTITUTIONS
1. Brigham Young University (BYU)
2. Clarkson University
3. Harvard University (HU)
4. New York University (NYU)
5. University of Rochester and GSF (UR/GSF)
6. University of Southern California (USC)
7. University of Washington (UW)

Section 2
METHODS

As summarized in Table 2, the methods applied to the mass and trace constituent data varied from research group to research group, including methods ranging from simple mass regressions on selected single tracer elements for each source class (e.g., vanadium for oil), to more elegant factor analysis based methods including Absolute Principal Component Analysis (APCA), Positive Matrix Factorization (PMF), and UNMIX. These various apportionments were then separately evaluated for their respective associations with daily mortality in a consistent manner across apportionments by researchers at the NYU (Washington, DC data set) and University of Washington (Phoenix, AZ data set) PM Centers. By keeping the health effects models consistent across researchers, this allowed an assessment of the extent to which variability in the source apportionment step contributed to variability in the subsequent analysis of health effects. The details of these analyses are presented in more detailed companion papers (Hopke et al. 2006; Ito et al. 2006; Mar et al. 2006), and are combined and summarized in this overview paper.

Table 2. Summary of the source apportionment analyses performed by each participating group.

Research Institution(s)	Phoenix, AZ	Washington, DC
BYU	UNMIX	UNMIX Iterated, Confirmatory FA
Clarkson	PMF2 and Expanded Model (ME)	PMF2
HU	Target Rotated PCA	Target Rotated PCA
NYU	PMF, APCA	PMF, APCA, Single Element mult. regr.
UR/GSF	APCA	
USC	UNMIX	UNMIX
UW	PMF	

DATA SETS ANALYZED

The two $PM_{2.5}$ mass and composition data sets employed in the source apportionments were selected based upon their ready availability for analysis, the similar availability of a compatible daily mortality record for health effects analysis, and the fact that their $PM_{2.5}$ composition analyses were similar in many ways to those characteristics that are available to researchers from the new EPA $PM_{2.5}$ STN. In this way, analyses of existing data sets could quickly be accomplished to provide information relevant to analyses that might be conducted in the future with the rapidly expanding EPA STN database. Brief descriptions of these databases are provided below, and more detailed descriptions are provided in the companion workshop papers (Hopke et al. 2006; Ito et al. 2006; Mar et al. 2006).

Particulate Matter Data Sets

In Phoenix, AZ, daily, integrated 24-hour samples were collected on 37 millimeter (mm) diameter Teflon and quartz filter media for fine particle mass and species measurements using a dual fine particle sequential sampler (DFPSS). Some 981 samples were collected during the time period from March 1995 through June 1998. Each sample was characterized by the measured concentrations of the following 46 chemical elements: Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Mo, Rh, Pd, Ag, Cd, Sn, Sb, Te, I, Cs, Ba, La, W, Au, Hg, Pb, organic carbon (OC), and elemental carbon (EC). The analytical uncertainty estimates associated with each measured concentration and the detection limits for both instruments were also included.

In Washington, DC, the $PM_{2.5}$ samples were collected on Wednesdays and Saturdays at the IMPROVE monitoring site located in downtown Washington, DC. Some 718 samples were collected between August 31, 1988 and December 31,1997. Integrated 24-hour $PM_{2.5}$ 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 (PIXE) for the elements Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, via X-ray fluorescence (XRF) for elements Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Mo, Rh, Pd, Ag, Cd, Sn, Sb, Te, I, Cs, Ba, La, W, Au, Hg, Pb, and via proton elastic scattering analysis (PESA) for elemental hydrogen concentration. The Nylon filter was analyzed via ion chromatography (IC) for sulfate, nitrate, and chloride. The quartz filters were analyzed via IMPROVE/TOR protocol for temperature resolved organic and elemental carbon fractions.

Daily Mortality Data Sets

Washington, DC death records were extracted from the National Center for Health Statistics database for the period 8/31/88-12/31/97, and daily counts were aggregated for the District of Columbia and the surrounding six counties: Montgomery Co., MD; Prince George's Co., MD; Fairfax Co., VA; Alexandria city, VA; Fairfax city, VA; and, Falls Church city, VA. Three categories of deaths were analyzed: (1) total non-accidental; (2) cardiovascular; and, (3) cardiovascular plus respiratory.

Phoenix, AZ mortality data from 1995 to 1997 were obtained from the Arizona Center for Health Statistics. In this analysis, we included only mortality counts for residents over 65 from zip code regions thought to be most represented by the EPA monitoring platform (see Mar et al. 2003). We evaluated total non-accidental mortality (ICD-9 codes <800.00) and cardiovascular mortality (ICD9 codes 390.00-448.99) from 2/9/95 to 12/31/97. From 1995 to 1997 there were a total of 9081 cases of total non-accidental deaths and 4109 cases of cardiovascular deaths.

DATA ANALYSIS METHODS

Source Apportionment Modeling

The above-described $PM_{2.5}$ mass and composition data sets were provided to each participating research group in December 2002, so that they could independently analyze them using their preferred source apportionment technique(s). To allow a consistent inter-comparison of results across research groups, participants were requested to submit results in a standardized format and with a list of items to describe the details of source apportionment analysis (e.g., type and extent of rotation, treatment of outliers, criteria used to include species in the analysis, etc.). Of the eleven potential participants to whom the data were sent, eight participants/teams from seven institutions submitted source apportionment results by the required deadline (April, 2003).

As described in more detail in the companion paper by Hopke et al. (2006), the fundamental principle of source apportionment, or receptor, modeling is that mass conservation can be assumed, and a mass balance analysis can be used to identify and apportion sources of airborne particulate matter in the atmosphere. If the number and nature of the sources affecting the air monitoring station are known, then the only unknown is the mass contribution of each source to each sample, S_{jk}. These values can be estimated using regression. This approach was first independently suggested by Winchester and Nifong (1971) and by Miler et al. (1972), and is now called the Chemical Mass Balance (CMB) model (Chow and Watson 2002, Cooper and Watson 1980; Cooper et al. 1984). In general, CMB models assume that the recorded aerosol mass (M_k) in $\mu g/m^3$ is due to the sum of impacts by individual sources (S_{jk}):

$$M_k = \sum_{j=1}^{p} S_{jk} \qquad [1]$$

where: k = 1,2, ...m days; j = 1,2, ...p sources; and, the total concentration of aerosol property C_{ik} (i.e., element i's ambient concentration on day k at a site) is:

$$C_{ik} = \sum_{j=1}^{p} S_{jk} f_{ij} \qquad [2]$$

where: f_{jj} = the mass-fraction of property i in emissions from source j. Thus, if the source profiles (f_{ij}) are known, the source contributions (S_{jk}) can be determined from the linear regression of the C_{ik} on the f_{ij}.

However, if (as is more usually the case), the source emission "signatures" are not known exactly, but only qualitatively (e.g., that vanadium is enriched in residual oil combustion particles, but the exact percentage is not known), then factor analyses methods are applied to identify and quantify the sources and their impacts.

The factor analysis approach to source apportionment assumes that the total concentration of each "observable" (element) is made up of the sum of contributions from each of p pollution source components:

$$Z_{ik} = \sum_{j=1}^{p} W_{ij} P_{jk} \qquad [3]$$

where: $Z_{ik} = \dfrac{C_{ik} - \overline{C}_i}{S_i}$ (the standardized z-score of element i's kth observation), and; [4]

P_{jk} = the jth factor component's value on the kth day;
W_{ik} = the scoring coefficient matrix of the components; and
s_i = the standard deviation of element i

With respect to CMB models, the P_{jk} are equivalent to the S_{jk} source impacts; and, the W_{ij} are equivalent to the F_{ij} source profiles. However, the P_{jk} and W_{ij} are derived by the factor analysis from the correlation matrix, and are outputs of the factor analysis (instead of inputs, as is the case for CMB). Such factor analysis approaches generally have the major advantage that they can identify and quantify non-traditional aerosols, such as secondary aerosols (formed in the atmosphere), and can incorporate non-PM tracers, such as the gaseous pollutants. Such Factor Analysis (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. It has been widely used in studies of airborne particulate matter composition data (e.g., 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 factor analysis (Koutrakis and Spengler 1987) that uses a targeted Procrustes factor rotation. An alternative approach called Absolute Principal Components 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; Kim and Henry 2000a&b) 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. (2006), have been applied by the different research groups in order to achieve source apportionments of the Washington, DC and Phoenix, AZ $PM_{2.5}$ data, as documented in Table 2.

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

Health Effects Modeling Analyses

Once the source apportionments were submitted, all the Washington, DC and Phoenix, AZ daily source apportionments were provided to Dr. Kazuhiko Ito of the NYU PM Center and Dr. Therese Mar of the University of Washington PM Center, respectively, for inclusion in time-series mortality models to assess the resulting variations in their resulting source-specific health effects estimates (i.e., Relative Risks). The city-specific mortality models employed are described below.

The model building steps of the Washington, DC time-series mortality model development used in these analyses (Ito et al., 2006) 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 GLM (McCullagh and Nelder, 1989). Using natural splines, we fit a smooth function of time to mortality in order to adjust the model for seasonal trends and unmeasured seasonal confounders, such as influenza epidemics. The inclusion of this term also reduces undesirable residual auto-correlation and over-dispersion 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 auto-correlation 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: (1) natural splines of the same-day temperature with four degrees of freedom to fit "hot" temperature effects; (2) 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, (3) an indicator for "hot" (daily mean temperature above 80 degrees) and "humid" (daily relative humidity above 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) in order to separately test the individual associations of each source category with mortality, after controlling for the variables considered in the base model. The relative risk associated with both an inter-quartile (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 participating groups' apportionment modeling results.

The basic steps of time-series model development used in the Phoenix, AZ analyses (Mar et al. 2005) were the same as for Washington, DC. Similarly, associations between source contributions and cardiovascular and total non-accidental mortality were analyzed using Poisson GLM in SPLUS 2000 (Insightful Inc., Seattle, WA). The same Phoenix base mortality model was applied to all group's source apportionment analyses in order 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 degrees of freedom for the smoothing of time trend (i.e., 4 df per year). The df for the natural splines for time trends were selected to minimize autocorrelation in the residuals and the Akaike Information Criterion (AIC). 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, DC analyses, the various research groups' respective estimated source contributions were added to this base model, in turn, as the particle pollution variable. The relative risk associated with both an inter-quartile (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, as 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 5^{th}-to-95^{th} percentile increment increase by source-apportioned $PM_{2.5}$ were inter-compared and then analyzed (within each city) by analysis of variance (ANOVA) and a general linear model in order to compare variations in model estimates between-source vs. within-source (i.e., due to different analyses).

Section 3
RESULTS

SOURCE APPORTIONMENT INTER-COMPARISONS

As described in more detail in Hopke et al (2006), the various source apportionment analyses from each of the participating research groups were inter-compared in two ways: 1) by comparing the mass contributions attributed to each source; and, 2) by calculating the correlation coefficients between the source contributions from $PM_{2.5}$ from the various groups within source groups. The various groups' solutions were compared with each other on an equal basis, as it was not possible to compare the various solutions to an accepted "gold standard" method, as one does not exist at this time for source apportionment. Table 2 notes the source apportionment analyses that were performed on these data sets, while Figures 1 and 2 present the mean and standard errors of the resulting $PM_{2.5}$ mass source apportionment for each source category in Washington, DC and Phoenix, AZ, respectively. While most groups were able to identify the same major sources in their source apportionment analyses of the trace constituent data, not all sources were identified by all researchers, as reflected by the fact that not all groups provided impacts for all possible source categories. In this plot, when differing researchers broke out the source impacts differently than 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 that overlap across the various groups' analyses, especially for the larger mass contributors. To be more quantitative, we conducted an Analysis of Variance (ANOVA) F-test of the within-source vs. between-source variations for each of the major source categories in Figures 1 and 2, and the results indicated there was significantly greater variability ($p<.001$) across source categories than across investigators/methods (i.e., that investigator/method variations were small compared to source-to-source variations). Overall, it can be seen from these plots and statistical analyses that, while there is some variability in the estimated mass impact results 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.

Since 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 an examination of the variability in the paired correlations of the various analyses' estimated daily source apportionment mass contributions over time, within each city. As shown in Figure 3a for Washington, DC and Figure 3b for Phoenix, AZ, the sulfate-containing, crustal, and nitrate components exhibited among the highest mean inter-correlations across the various research groups in these cities. Among the chief $PM_{2.5}$

mass contributors (as shown in 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 constituent(s) for unique identification), notably traffic and wood burning in Washington, DC, and wood burning and metals in Phoenix, AZ.

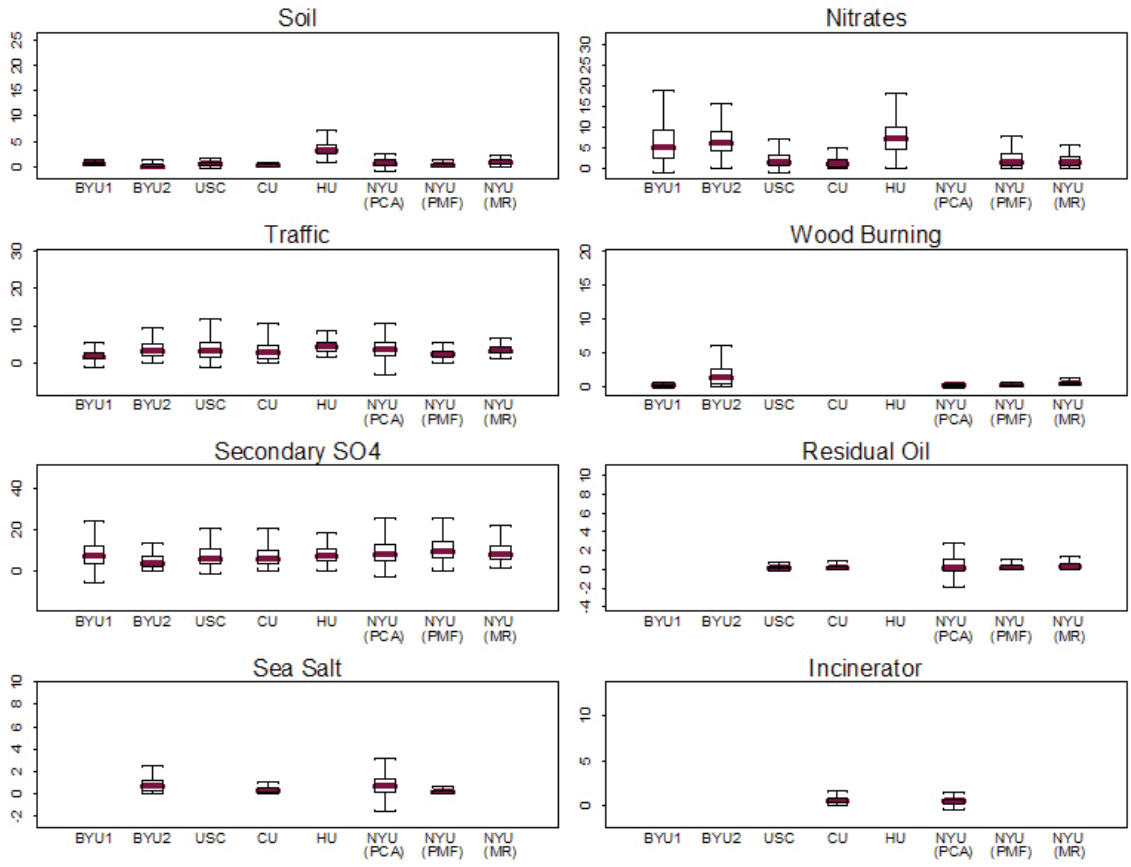

Figure 1. Mean and Range of Mass Impacts Predicted by Each Research Group's Source Apportionment Analysis of the Washington, DC $PM_{2.5}$ data set (µg/m^3).

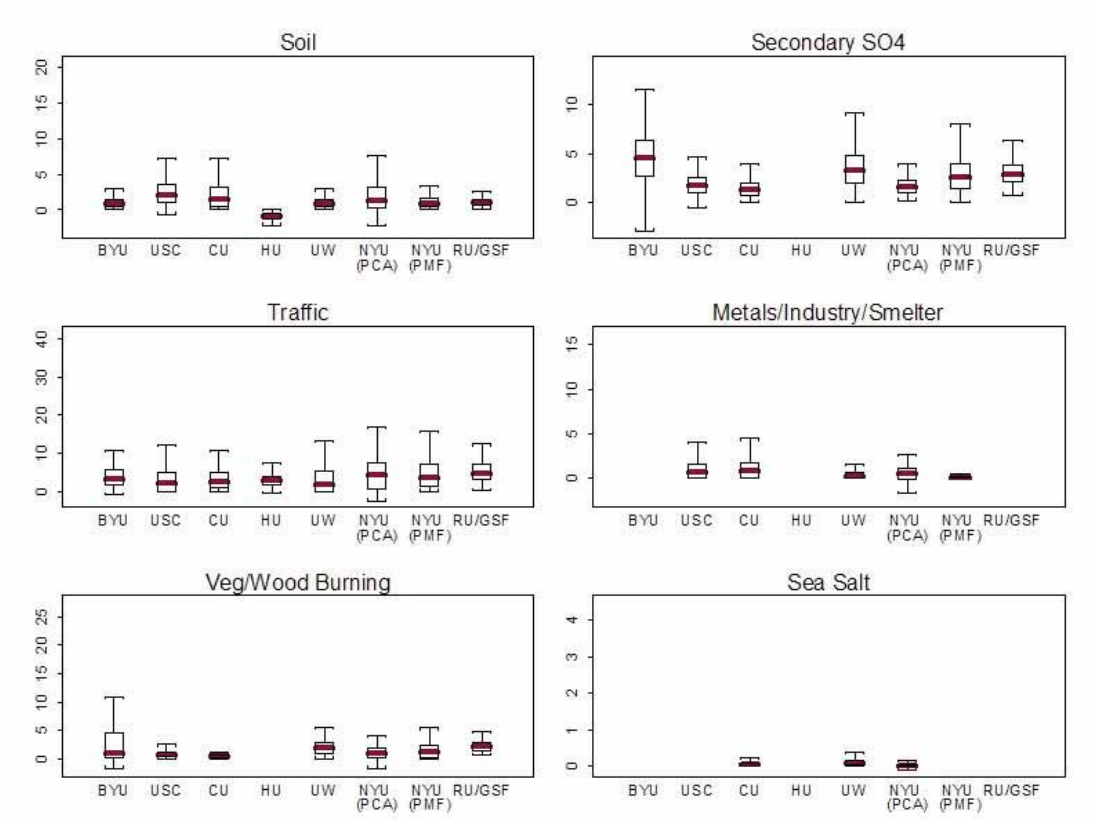

Figure 2. Mean and Range of Mass Impacts Predicted by Each Research Group's Source Apportionment Analysis of the Phoenix, AZ $PM_{2.5}$ Data (µg/m^3).

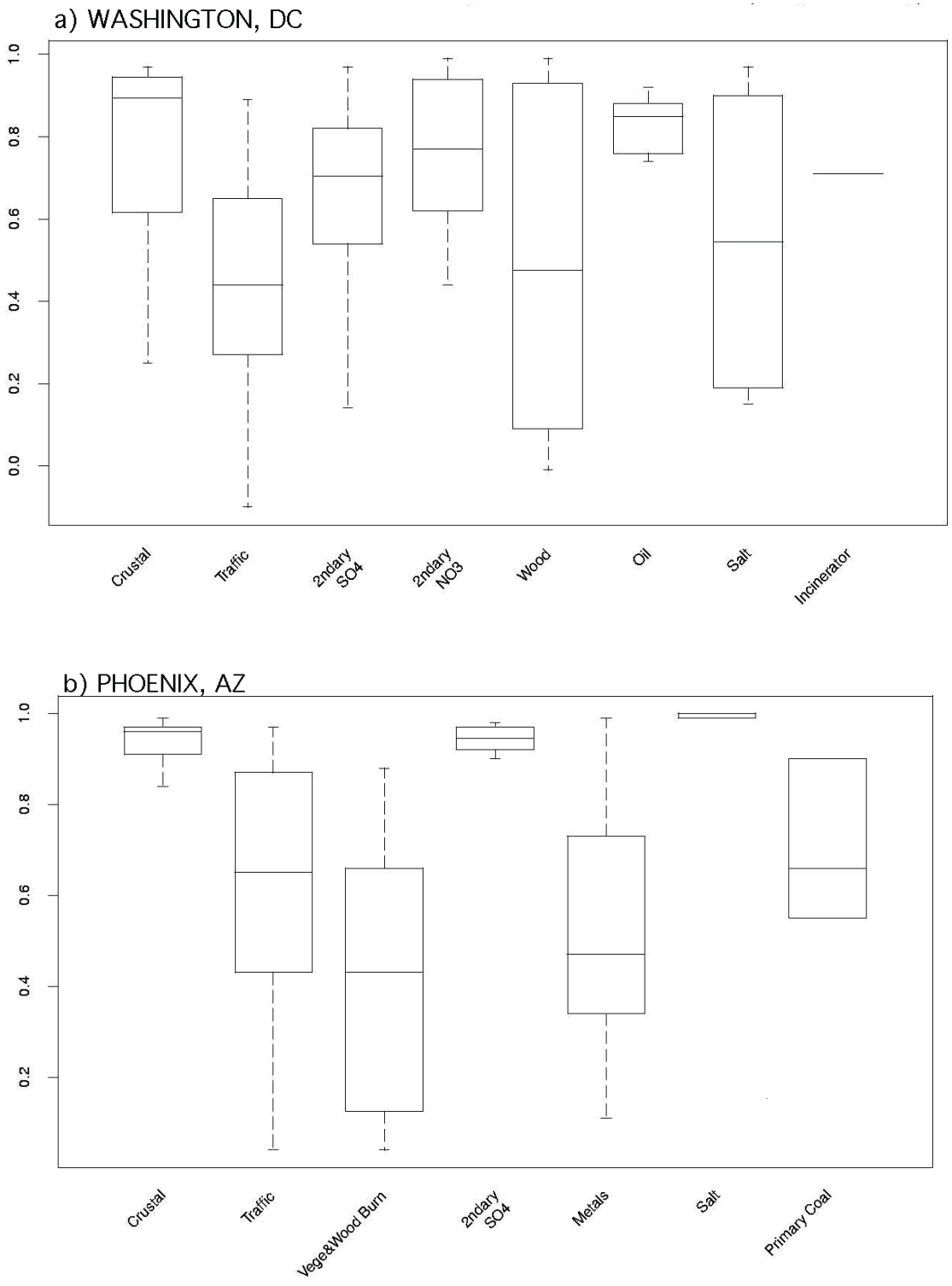

Figure 3. Box and whisker plots of the distributions of temporal correlation coefficients between all possible pairs of similar source contributions resolved for: a) Washington, DC; and, b) Phoenix, AZ.

TIME-SERIES MORTALITY EFFECT ESTIMATE INTERCOMPARISONS

The source apportionment results for each group were combined with the mortality data in Washington, DC and Phoenix, AZ, and time-series mortality regressions were then run, as described more fully in Ito et al. (2006) and Mar et al. (2006), respectively. Figure 4 displays the resulting mean relative risk estimates (and 95% confidence intervals(CI)) of cardio-vascular (CV) and total daily mortality, for each major source category identified in Washington, DC and Phoenix, AZ, for the overall workshop estimate with source apportionment inter-analysis variation excluded (—)and with the inter-analysis 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 across-source apportionment groups and analyses is small, relative to the overall uncertainty of these estimates. In quantitative terms, the % increase in the uncertainty (i.e., in the CI) for each displayed source category's mortality RR in Washington, DC added by the inter-analysis variability was: 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 % increase in the uncertainty (i.e., in the CI) for each displayed source category's mortality RR that was added by the inter-analysis variability was: 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.

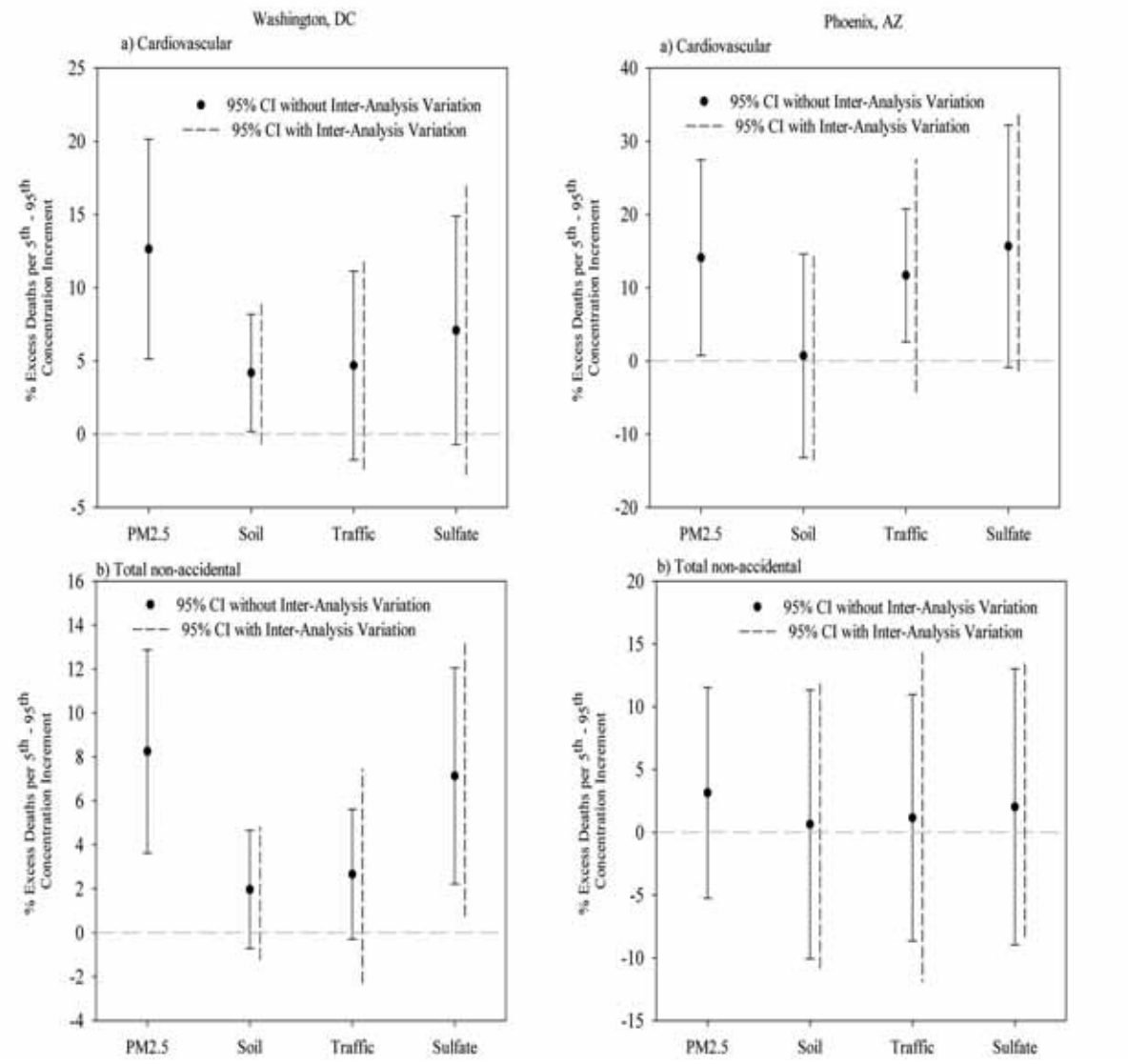

Figure 4. Mean Relative Risks (RR) Estimates and 95% Confidence Intervals of Cardio-vascular and Total Daily Mortality, for each Major Source Category in Washington, DC and Phoenix, AZ, for the Overall Workshop Estimate with Source Apportionment interanalysis variation excluded (—), and with the interanalysis variation included (----).

The between-source variation in these daily mortality relative risks (RR's) was also compared to within-source variations (due to different analyses). As shown in Table 3, significantly larger variation was found between-sources than between-research groups in reported RR's ($p<.001$) using an ANOVA (in a general linear model) of the individual investigator estimates and variances (for each death category in each city) that are presented in Ito et al. (2006) and Mar et al. (2006). In the general linear model, between-group variation was found to be a non-significant predictor for both death categories in both cities (with p-values ranging from 0.38 to 0.65 for between-group differences), while the between-source type variation was a

statistically significant predictor of RR in both cities and death categories ($p<.001$). Overall, these various results indicate that variations in choice of research group or source apportionment method have only a small effect on variations in the RR estimates for identified sources, relative to the variations in RR caused by different source components and the mortality regression process, and that researcher variations in source apportionment applications should not be a barrier to comparing the source-specific $PM_{2.5}$ RR's.

Table 3. ANOVA Analyses of Source-Specific Mortality Relative Risk Estimates

Mortality Category	ANOVA p-value	% Source Category Variance	% Research Group Variance
Washington, DC Cardiovascular	<0.001	47.5%	9.5%
Washington, DC Total	<0.001	80.0%	2.6%
Phoenix, AZ Cardiovascular	<0.001	76.3%	4.5%
Phoenix, AZ Total	<0.001	64.8%	6.3%

The size of the source-specific RR estimates from these analyses can also be compared with other published source-category effect estimates, although very few are available to-date. The most consistently significant category was secondary sulfates, and these have been most widely examined before in the published literature. In this case, the total mortality RR estimates for the secondary sulfate component came to 5.2% change per 10 ug/m^3 in Phoenix and 3.8% per 10 ug/m^3 in Washington, DC. This is somewhat larger than the sulfate dominated "Coal" component reported by Laden et al. (2000), but much smaller than that derived from Ozkaynak and Thurston (1987) which indicated some 8% per 10 ug/m^3 for this component, but that study was of annual mortality associated with long-term exposures, rather than the daily mortality considered here. It is interesting, however, that the Washington, DC component estimate from this work (3.8% per 10 ug/m^3 for the sulfate component) is very close to the sulfate-related coal component value derived by Laden and colleagues for Boston, MA (2.8%). Motor vehicles was another component that approached significance in this work, yielding a 0.9% per 10 ug/m^3 RR in Phoenix, and 4.2% in Washington. These are similar to the 3.4% per 10 ug/m^3 found by Laden and colleagues (Laden et al. 2000), and the 2% per 10 ug/m^3 derived from the work of Ozkaynak and Thurston (1987). Thus, these source-specific estimates appear reasonable when compared to the limited source-specific mortality analyses done in the past, but much more work of this type needs to be done in the future before broad-based comparisons to the RR results from this workshop will be possible.

Section 4
DISCUSSION AND CONCLUSIONS

Regarding 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 (i.e., that contribute most of the $PM_{2.5}$ mass at each site) are consistently identified by the different groups in each city, with similar elemental make-ups (i.e., key tracers). Methods were generally found to yield the most consistent results (i.e., the highest correlations across groups over time) for sources with the most definable (i.e., most unique) tracers or combinations of tracers in each city. In Washington, DC, soil, secondary sulfate and nitrate, oil burning and incineration were most unambiguously identified by various methods, while wood burning, salt and traffic were less well correlated across analyses. In Phoenix, AZ, soil, traffic, secondary sulfate and sea spray were most highly correlated across analyses, while wood and vegetative burning, metals industry particles, and coal fly ash were less well correlated. Based on the relative sizes of these inter-group 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 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, while there are no "gold standard" correct answers for the source identification and apportionments in such "real world" data sets as considered in this workshop, the apportionment consistency found for the largest $PM_{2.5}$ source contributors across different researchers in these cities, often using differing statistical methods, indicates reliability of the source apportionment approach.

Regarding the health effects apportionments to the different source components of $PM_{2.5}$, the between-source variation in daily mortality relative risk (RR) was significantly larger than the between-research group variation in reported RR's. Thus, analysis-to-analysis variability in the source apportionments was small in comparison to the overall uncertainty in the mortality RR estimates. In addition, between-group variation in RR estimates was found to be non-significant, while the between-source type variation was statistically significant. This 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 cardio-vascular deaths in both cities), most source categories were found to be non-significant contributors, but 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. However, the source RR estimates generally had overlapping confidence bands, indicating that larger numbers of observations will be required in each of these cities in order to have enough power to significantly differentiate the impacts of the various source types. In addition, the overall source-specific RR estimates derived in this work were found to appear reasonable when compared to the limited source-specific mortality analyses published in the past, but many more source apportionment-mortality analyses of this type need to be done in the future before broad-based comparisons to the source-specific RR results from this workshop will be possible.

Overall, the results of this inter-comparison 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 relative risk uncertainties. Thus, 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 that is 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 time-series health effects analyses, as well as also serving to better clarify the potentially differing distributed-lag natures of the various source-specific impacts identified in this workshop. Although further research, and possibly the addition of more key tracers to the speciation of $PM_{2.5}$, is needed to better characterize ambient tracer profiles for sources with less well defined compositional characteristics (e.g., for vegetative burning and traffic), the results of this workshop provide substantiation 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

Chow, J., and Watson, J. (2002) Review of $PM_{2.5}$ and PM_{10} Apportionment for Fossil Fuel Combustion and Other Sources by the Chemical Mass Balance Receptor Model, *Energy & Fuels* 16: 222–60.

Cooper, J.A., and Watson, J.G. Jr. (1980) Receptor Oriented Methods of Air Particulate Source Apportionment, *J. Air Pollut. Control Assoc.* 30(10):1116–25.

Cooper, J.A., Watson J.G., and Huntzicker, J.J. (1984) The Effective Variance Weighting for Least Squares Calculations Applied to the Mass Balance Receptor Model, *Atmospheric Environ.* 18: 1347–55.

Gao, N., Cheng, M.D., Hopke, P.K. (1994) Receptor Modeling for Airborne Ionic Species Collected in SCAQS, 1987, *Atmospheric Environ.* 28: 1447–70.

Henry, R.C., and Kim, B.M. (1999) Extension of Self-Modeling Curve Resolution to Mixtures of More Than Three Components. Part 1. Finding the Basic Feasible Region, *Chemom. Intell. Lab. Syst.* 8: 205–16.

Hopke, P.K., Gladney, E.S., Gordon, G.E., Zoller, W.H., and Jones, A.G. (1976) The Use of Multivariate Analysis to Identify Sources of Selected Elements in the Boston Urban Aerosol, *Atmospheric Environ.* 10: 1015–25.

Hopke, P.K., Ito, K., Mar, T., Christensen, W., Eatough, D.J., Henry, R.C., Kim, E., Laden, F., Lall, R., Larsen, T., Liu, H., Neas, L., Pinto, J., Stölzel, M., Suh, H., Paatero, P., and Thurston, G. (2005) PM Source Apportionment and Health Effects: Intercomparison of Source Apportionment Results. Submitted to *J. Expo. Anal. Environ. Epidemiol.*

Ito, K., Christensen, W., Eatough, D.J., Henry, R.C., Kim, E., Laden, F., Lall, R., Larson, T.V., Neas, L., Hopke, P.K., and Thurston, G. (2005) An Investigation of Inter-Method Variability in Associations between Source-Apportioned Fine Particle Mass and Daily Mortality in Washington, DC. Submitted to *J. Expos. Anal. Environ. Epidemiol.*

Kim, B.M., and Henry, R.C. (1999) Extension of Self-Modeling Curve Resolution to Mixtures of More Than Three Components. Part 2. Finding the Complete Solution, *Chemom. Intell. Lab. Syst.* 49: 67–77.

———. (2000a) Extension of Self-Modeling Curve Resolution to Mixtures of More Than Three Components. Part 3. Atmospheric Aerosol Data Simulation Studies, *Chemom. Intell. Lab. Syst.* 52: 145–54.

———. (2000b) Application of SAFER Model to the Los Angeles PM_{10} Data, *Atmospheric Environ.* 34: 1747–59.

Koutrakis, P., and Spengler, J.D., 1987. Source Apportionment of Ambient Particles in Steubenville, OH Using Specific Rotation Factor Analysis, *Atmospheric Environ.* 21: 1511–19.

Laden, F., Neas, L.M., Dockery, D.W., and Schwartz, J. (2000) Association of Fine Particulate Matter from Different Sources with Daily Mortality in Six U.S. Cities; *Environmental Health Perspectives* 108: 941–47.

Mar, T.F., Norris, G.A., Koenig, J.Q., and Larson, T.V. (2000) Associations between Air Pollution and Mortality in Phoenix, 1995–1997, *Environmental Health Perspectives* 108: 347–53.

Mar, T.F., Koenig, J.Q., Larson, T.V., Christensen, W., Eatough, D.J., Henry, R.C., Ito, K., Kim, E., Laden, F., Lall, R., Neas, L., Hopke, P.K., and Thurston, G. (2005) An Investigation of Inter-Method Variability in Associations between Source-Apportioned Fine Particle Mass and Daily Mortality in Phoenix, AZ, submitted to *J. Expo. Anal. Environ. Epidemiol.*

McCullagh, P., and Nelder, J.A. (1989) *Generalized Linear Models*, London: Chapman and Hall.

Miller, M.S., Friedlander, S.K., and Hidy, G.M. (1972) A Chemical Element Balance for the Pasadena Aerosol, *J. Colloid and Interface Science* 1972, 39: 165–76.

Ozkaynak, H., and Thurston, G.D. (1987) Associations between 1980 U.S. Mortality Rates and Alternative Measures of Airborne Particle Concentration. *Risk Anal.* 7: 449–60.

Paatero, P. (1997) Least Squares Formulation of Robust, Non-Negative Factor Analysis, *Chemom. Intell. Lab. Syst.* 37: 23–35.

———. (1999)The Multilinear Engine: A Table-Driven Least Squares Program for Solving Multilinear Problems, Including the n-Way Parallel Factor Analysis Model, *J. Computational and Graphical Stat.* 8: 854–88.

Paatero, P., Hopke, P.K., Song, X.H., and Ramadan, Z. (2002) Understanding and Controlling Rotations in Factor Analytic Models, *Chemom. Intell. Lab. Syst.* 60: 253–64.

Ramadan, Z., Song, X.H., and Hopke, P.K. (2000) Identification of Sources of Phoenix Aerosol by Positive Matrix Factorization. *J. Air Waste Manage. Assoc.* 50: 1308-1320.

Roscoe, B.A., Hopke, P.K., Dattner, S.L., and Jenks, J.M. (1982) The Use of Principal Components Factor Analysis to Interpret Particulate Compositional Data Sets, *J. Air Pollut. Control Assoc.* 32: 637–42.

Schauer J.J., Rogge, W.F., Hildemann, L.M., Mazurek, M.S., Cass, G.R., and Simoneit, B.R.T. (1996) Source Apportionment of Airborne Particulate Matter Using Organic Compounds as Tracers, *Atmospheric Environ*. 30:22; 3837-3855.

Schauer, J.J., and Cass, G.R. (2000) Source Apportionment of Wintertime Gas-phase and Particle-phase Air Pollutants Using Organic Compounds as Tracers, *Environ. Sci. Technol.* 34: 1821-1832.

Spengler, J.D., and Thurston, G.D.(1983) Mass and Elemental Composition of Fine and Coarse Particles in Six U.S. Cities, *J. Air Pollut. Control Assoc.* 22(12): 1162–71.

Song, X.H., Polissar, A.V., and Hopke, P.K. (2001) Source of Fine Particle Composition in the Northeastern U.S. *Atmospheric Environ.* 35: 5277-5286.

Thurston, G.D., and Spengler, J.D. (1985) A Quantitative Assessment of Source Contributions to Inhalable Particulate Matter Pollution in Metropolitan Boston, *Atmospheric Environ.* 19: 9–26.

Thurston, G.D., Ito, K., Mar, T., Christensen, W.F., Eatough, D.J., Henry, R.C., Kim, E., Laden, F., Lall, R., Larson, T.V., Liu, H., Neas, L., Pinto, J., Stölzel, M., Suh, H., Hopke, P.K. (2005) The

Workshop on the Source Apportionment of PM Health Effects: Inter-Comparison of Results and Implications, submitted to *Environmental Health Perspectives.*

Tsai, F.C., Apte, M.G., and Daisey, J.M. (2000) an Exploratory Analysis of the Relationship Between Mortality and the Chemical Composition of Airborne Particulate Matter, *Inhalation Toxicology* 12 (Suppl. 2): 121-135.

Winchester, J.W., and Nifong, G.D. (1971) Water Pollution in Lake Michigan by Trace Elements from Pollution Aerosol Fallout, *Water, Air, Soil Pollut.* 1: 50–64.

Appendix A

List of Participants Attending the Workshop

Source Apportionment Workshop Attendees

Arden Conference Center

May 28-30, 2003

Overnight Attendees	Affiliation
Ellen Burkhard	NYSERDA
William Christensen	BYU
Delbert J. Eatough	BYU
Tom Gentile	NYSDEC
Ronald Henry	USC
Phil Hopke	Clarkson
Kazuhiko Ito	NYU-SOM
Eugene Kim	Clarkson
Francine Laden	Harvard School of Public Health
Tim Larsen	University of Washington
Hao Liu	University of Washington
Dan Luttinger	NY State Dept. of Health
Therese Mar	University of Washington
Lucas Neas	U.S. EPA
Joe Pinto	U.S. EPA
Matthias Stölzel	Institute of Epidemiology, GSF, Germany
Helen Suh	Harvard of Public Health
Geoffrey Sunshine	Health Effects Institute
William Wilson	U.S. EPA

Day Guests	
Mitch Cohen	NYU-SOM
Jing-Shiang Hwang	NYU-SOM
Patrick Kinney	Columbia University
Ramona Lall	NYU-SOM
Shao-I Hsu	NYU-SOM
Polina Maciejczyk	NYU-SOM
Colette Prophete	NYU-SOM
George Thurston	NYU-SOM
Dritan Xhillari	NYU-SOM

Appendix B

Agenda for the Source Apportionment and Health Workshop

AGENDA

Workshop on PM Source Apportionment and Health Effects
An EPA-PM Center Workshop on PM Health Effects
Arden House Conference Center (http://www.ardenhouse.com/)
Harriman, NY

Wednesday, May 28
4-6PM Registration at Arden House Conference Center and Hotel
6-7PM Dinner at Hotel
7:30-9PM: Introduction to Workshop Goals and Approach
7:30 Welcome and Introduction to Workshop: George Thurston and Helen Suh
8:00 Description of Phoenix, AZ PM Data set : W.E. Wilson
8:30 Description of Washington, DC Data set: P.K. Hopke

Thursday, May 29
8:00 –9:00 AM: Breakfast
9:00-9:30AM: The Role of SA in PM Health Effects Analysis: G. Thurston
9:30-10 AM The PMF Method of Source Apportionment: Phil Hopke
10:00-11AM: The UNMIX Method of Source Apportionment: Ron Henry
11:30-Noon: Harvard Analyses of Phoenix and Washington, DC Data sets: F. Laden
Noon-1PM: Lunch
1:-1:30: BYU Analyses of Phoenix and Washington, DC Data sets: D. Eatough and W. Christensen
1:30-2:00: Washington State Analyses of Phoenix and Washington, DC Data sets: Tim Larsen and T. Mar
2:00-2:30: NYU Analyses of Phoenix and Washington, DC Data sets: K. Ito
2;30-3:00: Clarkson Analyses of Phoenix and Washington, DC Data sets: P. Hopke
3_3:30 Break
3:30-4:00: USC Analyses of Phoenix and Washington, DC Data sets: R. Henry
4:00-4:30: Intercomparison of Various Groups SA Results: G. Thurston
4:30-5:00: Group Discussion of Variations in Results and Implications to Health Assessment: Led by H. Suh and G. Thurston
6:00 Dinner
7:30-9PM Informal Get-Together at Arden Conference Center

Friday, May 30
8:00 –9:00 AM: Breakfast
9:00-9:30 AM: Description of Health Effects Modeling and Intercomparison of Results for Washington, DC: K. Ito
9:30-10:00: Description of Health Effects Modeling and Intercomparison of Results for Phoenix, AZ: T. Mar
10-10:30: Summarizing the Overall Precision of Health effects Estimates from Two Cities: G. Thurston
10:30-11:30: Group Discussion of Variations in Results and Implications to Health Effects Assessment of PM by Source Led by H. Suh and G. Thurston
11:30-12:00: Development of Conclusion and Recommendations
12:00: Lunch

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Inter-Comparison of Results and Implications

Final Report 06-01

State of New York
George E. Pataki, Governor

New York State Energy Research and Development Authority
Vincent A. DeIorio, Esq., Chairman
Peter R. Smith, President and Chief Executive Officer

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