Final Technical Report

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Objective(s) of the Research Project: The main objective of this project was to quantitatively characterize spatio-temporal measurement error of ambient air particulate matter (PM) and gaseous co-pollutants measured at routine regulatory-based air monitors as a function of site characteristics using the entire U.S. air-monitoring network. The rationale was that, while PM_{10} was often reported to be associated with health outcomes most significantly among the criteria air pollutants, this may have been in part due to differential exposure characterization error—PM_{10} may have less measurement error than other pollutants. Furthermore, we expected that PM_{10} might have varying exposure characterization error across U.S. due to varying source types, which may result in heterogeneity of estimated PM_{10} risk estimates across cities. The prevailing hypothesis was that the PM_{10} and gaseous co-pollutants data from a single air monitoring station could adequately reflect the population exposure for the entire city, and that resulting risk estimates and their significance are not biased. Also, during the course of this project, the new $PM_{2.5}$ chemical speciation network's data became available (from ~ 2001). Therefore, we also examined exposure characterization error across components of $PM_{2.5}$.

Summary of Findings:

Technical Aspects

Monitor-to-monitor correlation was computed for PM_{10} and gaseous criteria pollutants in seven North-Central States: Illinois, Indiana, Michigan, Ohio, Pennsylvania, Wisconsin, and West Virginia (Ito, et al., 2001). These states cover 312,968 square miles, and contain over 56 million people. The study area also included major cities, such as Chicago, Cincinnati, Cleveland, Columbus, Detroit, Indianapolis, Milwaukee, Philadelphia, and Pittsburgh. Air pollution data for PM_{10}, SO_2, O_3, NO_2, and CO were retrieved from U.S. EPA's Aerometric Information Retrieval System (AIRS, now called Air Quality System, or AQS) for study period 1988-1990 for these states. The AIRS working file format AMP355 was used. The number of monitoring stations for these states was 287, 295, 241, 80, and 108 for PM_{10}, SO_2, O_3, NO_2, and CO, respectively. Since most of the PM_{10} data were collected on an every-6th-day sampling schedule (gaseous pollutants and weather variables were collected every day) at most sites, the data analyses were

to be conducted for the PM_{10} sampling days only (total of 183 possible days during the 3 year period), in order to use comparable sample sizes. After removing seasonal trends, the monitor-to-monitor temporal correlation among the air pollution/weather variables within 100 mile separation distance in these areas could generally be ranked into three groups: 1) temperature, dew point, relative humidity ($r>0.9$); 2) O_3, PM_{10}, NO_2 (r: 0.8-0.6); and 3) CO, SO_2 ($r<0.5$). Using the subsets for separation distance less than 100 miles, regression analyses of these monitor-to-monitor correlation coefficients were also conducted with explanatory variables including separation distance, qualitative (land use, location setting, and monitoring objectives), and quantitative (large and small variance) site characteristics, and region indicators for Air Quality Control Regions (AQCR). The AQCRs are EPA-designated regional boundaries that were established based on jurisdictional boundaries, urban-industrial concentrations, and other factors such as air sheds, for the purpose of providing adequate implementation of National Ambient Air Quality Standards (NAAQS). The separation distance was a significant predictor of monitor-to-monitor correlation decline especially for PM_{10} and NO_2 (~0.2 drop over 30 miles). Site characteristics were, in some cases, significant predictors of monitor-to-monitor correlation, but the magnitudes of their impacts were not substantial. Regional differences, as examined with AQCR indicators were, in some cases (e.g., Metropolitan Philadelphia) substantial, to the extent that the pollutants that have generally poor monitor-to-monitor correlation in the overall 7 states data (i.e., SO_2 and CO) showed monitor-to-monitor correlation comparable with PM_{10} and O_3.

We extended the analysis to the nationwide data for years 1988-1997 (Ito, et al., 2005). We retrieved data from the EPA's AIRS database. The data analyses were to be conducted for the PM_{10} sampling days only for all the air pollutants, in order to use comparable sample sizes. The total number of monitors analyzed were 1892, 1133, 969, 352, and 632, for PM_{10}, O_3, SO_2, NO_2, and CO, respectively. Monitor-to-monitor correlations were computed for all the monitors within each of the AQCRs, limiting the scale of separation distance within the region. The medians of the within-AQCR median separation distance for PM_{10}, SO_2, O_3, NO_2, and CO were 26, 19, 26, 18, and 11 miles, respectively. The AIRS database contains monitor characteristic data elements associated with each air pollution monitor. These include: *Land Use* (Residential, Commercial, Industrial, Agricultural, Forest, Desert, and Mobile); *Location Setting* (Urban, Suburban, and Rural); and *Monitoring Objective* (Maximum Concentration, Population Exposure, Background, Source, and Objective Changed). The overall rankings in monitor-to-monitor correlation on the average were, in descending order, O_3, NO_2, and PM_{10}, ($r \sim 0.6$ to 0.8) > CO ($r < 0.6$) > SO_2 ($r < 0.5$). The resulting median monitor-to-monitor correlation for each monitor was modeled as a function of qualitative site characteristics (i.e., land-use, location-setting, and monitoring-objective), and quantitative information (median separation distance, longitude/latitude or regional indicators) for each pollutant using Generalized Additive Models (GAM). To assure convergence of the GAM's iterative estimation procedure, the convergence criteria parameters, as suggested by Dominici, et al. (2002), were as follows: epsilon = 10^{-14}; back-fitting epsilon = 10^{-14}; maximum iteration = 1000; and back-fitting maximum iteration = 1000. A smoothing function of median separation distance was included in the regression with locally estimated smoothing. The correlation was assumed to decline uniformly as a function of separation distance. Therefore, a relatively wide span of 0.4 was chosen for all the air pollutants. Based on our previous analysis using a smaller geographic coverage of North-central states (Ito, et al., 2001), having a very large or small variance of temporal fluctuations was a significant predictor of a low correlation. Therefore, indicator variables for the monitors with the largest

and smallest variance in the five percentiles were also included in the regression model. Regional variation, or heterogeneity, of the monitor-to-monitor correlation was modeled using two alternative approaches: 1) using the seven regional categories; and 2) applying a smooth function of longitude/latitude. As with the qualitative monitor characteristic variables, the seven regional categories were arbitrarily numbered and modeled as a group, using the smoothing spline function with six degrees of freedom. Since the regional differences in the distribution of median separation distance may influence the estimate of regional variation, even with the simultaneous inclusion of median distance in the model, the analysis was also repeated using the data stratified by the separation distance. The data were split in half at the median, and also in fourths at quartile values. In the model using smoothing function of longitude/latitude ('loess' in S-plus), a series of spans (0.4, 0.3, 0.2, 0.10, 0.05, 0.02, and 0.01) were used because we did not have strong assumptions regarding the pattern and smoothness of the regional variation pattern. The span for the final model was chosen for each air pollutant based on: 1) a visual inspection of the spatial pattern of the predicted values; 2) distribution of the predicted values; and 3) the generalized cross-validation values computed for each of the spans. Both the separation distance and regional variation were important predictors of the correlation. For PM_{10}, for example, the correlation for the monitors along the East Coast was higher by ~0.2 than for western regions. The qualitative monitor characteristics were often significant predictors of the variation in correlation, but their impacts were not substantial in magnitude for most categories.

These results suggest that the apparent regional heterogeneity in PM_{10} effect estimates, as well as the differences in the significance of health outcome associations across pollutants may, in part, be explained by the differences in monitor-to-monitor correlations by region and across pollutants. To examine this issue, we also conducted a regression analysis to see if the heterogeneity of PM_{10} risk estimates across the National Morbidity, Mortality, and Air Pollution Study (NMMAPS) 90 cities (publicly available) could in part be explained by the difference in the extent of PM_{10} monitor-to-monitor correlation. Of the 90 cities, 83 cities could be matched with the median PM_{10} monitor-to-monitor correlation discussed above. The inverse-variance weighted regression of PM_{10} mortality risk estimates on the median PM_{10} correlation showed a positive prediction of PM_{10} mortality risk estimates with a slope of 0.14 (95% CI: [-0.02, 0.30]) per 0.1 increment of PM_{10} correlation.

Possible exposure characterization errors across $PM_{2.5}$ components were examined using $PM_{2.5}$ chemical speciation data collected at three locations in New York City during 2001–2002 (Ito, et al., 2004). The species that are associated with secondary aerosols (e.g., SO_4, NH_4, NO_3, organic carbon [OC], etc.) tended to show high monitor-to-monitor correlations, whereas the species that are likely associated with more local sources (e.g., elemental carbon [EC] as a traffic source marker) showed lower monitor-to-monitor correlations. Source-apportionment using these data was also conducted for each monitor's data. The estimated source-apportioned $PM_{2.5}$ mass generally showed the highest monitor-to-monitor correlation for the secondary aerosol factor (r range: 0.72–0.93). The correlation for the more localized traffic-related factor was more variable (r range: 0.26–0.95). The estimated mean $PM_{2.5}$ mass contributions by source/pollution type across the monitors varied least for the secondary aerosol factor. We also extended the analysis to 28 metropolitan statistical areas (MSA's) where multiple monitors generated $PM_{2.5}$ chemical speciation data for the years 2001-2003. We analyzed a set of key $PM_{2.5}$ components that were of interest in terms of toxicological effects, source signature, and generally large

signal-to-noise ratios: i.e., Ni, V, Pb, Cr, Mn, Fe, Si, As, Se, SO_4, NH_4, NO_3, EC, and OC. Again, the species associated with secondary aerosols (e.g., SO_4, NO_3) showed high monitor-to-monitor correlation. However, the monitor-to-monitor correlation for other species varied widely across the MSA's, likely reflecting the variation in the levels and major source types across the MSAs.

The monitor-to-monitor correlations discussed above are pertinent to the interpretation of results from short-term effects (i.e., time-series and longitudinal) studies. However, we also examined potential exposure characterization errors that are pertinent to long-term effects (i.e., cohort and cross-sectional) studies, using the same 28 MSA's. We found that, for the key $PM_{2.5}$ components, the coefficient of variation (CV) for across-MSA variation was generally far larger than those for within-MSA variations, with only a few exceptions. This result suggests that the quality of spatial resolution of the key $PM_{2.5}$ components is sufficient and adequate for the analysis of cross-sectional cohort data.

The results from the source apportionment workshop (Thurston, et al., 2005; Hopke, et al., 2006; Ito, et al., 2006; Mar, et al., 2006) also provided information regarding the exposure characterization error associated with source-apportioned $PM_{2.5}$. The comparison of source-apportioned $PM_{2.5}$ across investigators using the $PM_{2.5}$ chemical speciation data sets from Phoenix, AZ and Washington, DC found that soil-, secondary sulfate-, residual oil combustion-, and salt-associated mass were most unambiguously identified by various methods, whereas vegetative burning and traffic were less consistently identified. Combined with the result suggestive of varying exposure characterization error across $PM_{2.5}$ species, and across U.S. regions mentioned above, a systematic examination of multi-city time-series health effects analysis will be needed. We compared the mean levels of key $PM_{2.5}$ chemical species and the published PM_{10} mortality risk estimates in the 60 MSA's in the NMMAPS study for which speciation data were available, and found that the city-to-city variation of PM_{10} risk estimates could be better explained by some $PM_{2.5}$ chemical species (Ni and V) than others (Lippmann et al., 2006). Thus, the city-to-city variation in PM health risk estimates may be modified by components of $PM_{2.5}$. Our future research will directly examine this issue.

Conclusions

There are differential exposure characterization errors across PM and gaseous pollutants at ambient levels. PM_{10}, $PM_{2.5}$, O_3, and NO_2 tend to have moderate to high temporal correlations across monitors within cities, compared to CO and SO_2. These errors also vary by region and site-specific characteristics. Some of the differences in the observed health effects across pollutants in the past health effects studies may be explained by our findings. However, the estimated ecologic level exposure characterization errors did not explain the city-to-city variation in the PM_{10} mortality risk estimates substantively. Components of PM may play roles in the city-to-city variation of PM health risks. A more comprehensive assessment of the overall exposure characterization error will need to consider personal level exposure error, but such information is currently available from only a few cities.

The project was technically feasible to conduct.

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Supplemental Keywords: NA

Relevant Web Sites: http://www.med.nyu.edu/environmental/
http://es.epa.gov/ncer/science/pm/centers.html