APPENDIX AVAILABLE ON THE HEI WEB SITE

Research Report 178

National Particle Component Toxicity (NPACT) Initiative Report on Cardiovascular Effects

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Section 1: NPACT Epidemiologic Study of Components of Fine Particulate Matter and Cardiovascular Disease in the MESA and WHI-OS Cohorts

Appendix J. Source Apportionment Literature Review for the Six MESA Cities

Note: Appendices that are available only on the Web have been assigned letter identifiers that differ from the lettering in the original Investigators' Report. HEI has not changed the content of these documents, only their identifiers.

Appendix J was originally Appendix I

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Although this document was produced with partial funding by the United States Environmental Protection Agency under Assistance Award CR–83234701 to the Health Effects Institute, it has not been subjected to the Agency's peer and administrative review and therefore may not necessarily reflect the views of the Agency, and no official endorsement by it should be inferred. For the research funded under the National Particle Component Toxicity initiative, HEI received additional funds from the American Forest & Paper Association, American Iron and Steel Institute, American Petroleum Institute, ExxonMobil, and Public Service Electric and Gas. The contents of this document also have not been reviewed by private party institutions, including those that support the Health Effects Institute; therefore, it may not reflect the views or policies of these parties, and no endorsement by them should be inferred.

This document was reviewed by the HEI NPACT Review Panel but did not undergo the HEI scientific editing and production process.

APPENDIX I: Source apportionment literature review for the six MESA cities

A number of investigators have examined the relative influence of regional versus urban scale sources in areas relevant to the six MESA cities. An excellent review on this subject was done by Allen and Turner (2008), including a discussion results from studies in Baltimore (Ouglei, et al., 2006) and New York City (Dutkiewicz et al, 2004 & 2006; Qin et al, 2006; Lall and Thurston, 2006; Ito et al., 2004) . They conclude that transport of aerosol over distances of 100-1000 km has a significant impact on both rural and urban $PM_{2.5}$, sulfate and organic carbon levels, but local sources are also significant contributions of elemental carbon and trace metals. More recently, Fairlie and colleagues (Fairlie et al, 2009) reported that in Baltimore approximately 60% of the sulfate aerosol predicted by CMAQ was transported from sources outside the urban area, with higher percentages in late summer. They traced high concentration summertime episodes to upwind pathways passing through Ohio, Pennsylvania and Maryland. Using an enhanced CMAQ model, Burr and Zhang (2011) apportioned $PM_{2.5}$ to ten source categories over the eastern U.S. Their model predictions included sites in Chicago, New York City and Charlotte, NC. They predicted ~20% of the summertime $PM_{2.5}$ was from coal combustion in Chicago and NYC, and ~50% in Charlotte, with wintertime values of 5-15% across the three cities. They also attributed ~20% of the wintertime $PM_{2.5}$ to biomass combustion in Charlotte and NYC, but less than 10% in Chicago, with significantly less biomass impacts in the summer at all three sites. Diesel and gasoline emissions both contributed between 5 and 10% of the $PM_{2.5}$ at all three sites. Industrial emissions were a larger contributor in Chicago (~20%) than at either of the other two locations. Liu et al. (2005, 2006) found that CMAQ predictions of biomass contributions at three of four paired urban rural sites in Georgia

and Alabama were higher than those from measurement based receptor models. Bari et al. (2003) looked at regional versus local sources of sulfate, PM2.5, and SO2 along with other vapor phase components in New York City and deduced that 43%, 30% and 14% of the locally measured sulfate, PM2.5, and SO2 were transported from upwind sources outside the urban region. Their results are consistent with those of Schwab et al. (2004) who observed relatively uniform sulfate concentrations across NYC with little urban excess compared with a nearby non-urban site. Based on earlier work by Kleeman and Cass (1999, 2001), Ying and Kleeman (2006) used a 3-D source oriented Eulerian air quality model to apportion secondary sulfate, nitrate and ammonium ion in Southern California based on an episode in 1996. Basin wide, they attributed ~67% of the nitrate to transportation related sources, whereas the majority of the sulfate was attributed to upwind sources outside the region. Locally generated sulfate was attributed to diesel engines and fuel combustion processes using high sulfur fuel.

Other studies that have relied on the variability of measured species to apportion source contributions at one or more sites within one or more of the six MESA cities have also provided important insights. These will be briefly discussed for each city where published information is available.

In Winston Salem, the relative importance of biomass combustion inferred from the CMAQ models discussed earlier is consistent with the observations by Sheesley et al. (2007) in using chemical mass balance with organic source markers. They found that ~30% of the measured OC in Winston Salem was attributable to biomass combustion, primarily in the Winter and Spring, with similar magnitude contributions from mobile sources. They also reported about ~30% of PM2.5 from secondary sulfate and ~10% from secondary nitrate. Geron (2009) also

reported significant contributions from biomass burning at a forested site near Durham, N.C. using carbon isotope tracers.

In New York City, Qin et al. (2006) used PMF to analyze species measurements taken over a four year period at five sites, two of which were in or near the Bronx where MESA subjects reside. At these two sites, they reported average contributions to PM2.5 from a secondary sulfate feature (~6-7 ug/m^3), a secondary nitrate feature (~1- 2 ug/m^3), a crustal feature (~1 ug/m^3), a spark ignition vehicle feature (~1-2 ug/m^3), an oil combustion feature (~0.5 – 1.5 ug/m^3), a diesel exhaust feature (~0.4 ug/m^3), and an aged sea-salt feature (~0.5 ug/m^3). Ito et al. (2004) used absolute principal component analysis (APCA) and PMF to analyze measurements at these same locations. They found a subset of similar sources contribution to PM2.5 at these same two sites near the Bronx, including a crustal feature (~2-3 ug/m^3 from APCA; ~0.5-1 ug/m^3 from PMF), a secondary sulfate feature (~6-7 ug/m^3 from APCA; ~4-5 ug/m^3 from PMF), a traffic feature (~3-6 ug/m^3 from APCA, ~3 ug/m^3 from PMF)and a residual oil/incineration feature (~2-4 ug/m^3 from APCA, ~3 ug/m^3 from PMF). Similar sources were deduced by Li et al. (2004) at a site in Queens using short-term sampling methods. Peltier and Lippmann (2008; 2009; 2010; 2011) examined the spatial distribution of weekly average selected trace metals and $PM_{2.5}$ concentrations across New York city based on simultaneous monitoring at a set of 12 monitoring sites. They observed a relatively uniform distribution of $PM_{2.5}$ across the city, but systematic spatial variation in a number of trace elements.. They observed a separate spatial pattern of Ni that was uncorrelated with V and had a maximum centered in the Bronx and a separate pattern of Ni and V elsewhere. They also observed spatial correlations and relatively uniform spatial patterns of Se, As and S in the summer indicative of contributions from upwind regional coal burning sources, and a spatially diverse and correlated

set of patterns of Pb and Zn as well as of Si, Al, and Ca indicative of crustal sources and road dust. None of the investigators discussed here reported contributions from biomass combustion in New York City.

In Baltimore, Ogulei et al. (2006) used PMF on measured species combined with a partial least squares relationship between factors and size distributions in order to identify twelve factors at the Baltimore supersite (Ondov et al., 2006) based on hourly measurements taken over six days in July and August of 2002. The average contributions to $PM_{2.5}$ were consistent with the following source categories: oil fired power plants (~3%), local secondary nitrate (~5%), local gasoline traffic (~8%), coal-fired power plants (~10%), regional secondary nitrate (~13%), secondary sulfate (~15%), diesel emissions (~2%), biomass from forest fire (~30%), particle nucleation (~1%), incinerator (~8%), crustal/road dust (~3%), and steel manufacturing (~2%). The secondary sulfate was lower during this sampling period than that previously reported (Ogulei et al., 2005).

In Chicago, Rizzo and Scheff (2007) used PMF and CMB to determine source contributions at STN sites. Major sources of $PM_{2.5}$ determined by CMB were secondary sulfate (~30%), secondary nitrate (~20%), motor vehicles (~30%) and biomass combustion (~10%). Major contributions deduced from PMF were secondary sulfate (~40%), secondary nitrate (~20%), motor vehicles (~20%), biomass combustion (~5%), and crustal material and road salt (~10%). Minor contributions were attributed to industrial sources using either method. Zhang et al. (2009) examined the sources of organic aerosol in four midwestern sites, including Northbrook, Illinois just north of Chicago. They used PMF along with organic markers to identify both primary organic aerosol (POA) and secondary organic aerosol (SOA). The POA was attributed to three sources-urban primary sources, mobile sources and other combustion

sources and accounted for 57% of the total fine particle organic carbon with higher contributions at the industrial site in East St. Louis (67%) and lower concentrations at the rural site in Bondville (44%).

In Minneapolis-Saint Paul, Chen et al. (2010) applied CMB to apportion sources contributing to measured species concentrations at two STN sites. They reported annual average $PM_{2.5}$ contributions at each site over a seven year period between January 2000 and December 2006 from ten source categories . Major sources and their contributions (in ug/m3) were soil dust (~0.3-0.4), gasoline vehicle (~1.0-1.2), diesel vehicle (~3-4), biomass combustion (~0.8), various dusts and road salt (~0.4 in total), secondary sulfate(~2.5), secondary nitrate (~3), and secondary organics (~0.1).

In Los Angeles, following earlier work by Manchester-Neesvig et al. (2003) on the sources of primary organic carbon and PM_{10}, Minguillon et al. (2008) applied CMB with organic tracers at seven measurement locations, including one in downtown LA, to apportion both fine and ultra-fine particulate organic carbon. At the downtown LA site, they apportioned fine particle contributions to OC (summer/winter in ug/m^3) from light duty vehicles (2/0.3), heavy duty vehicles (0.6/1.1), ships (.04/.04), biomass burning (.006/.015) and road dust (0.7/0.3). Kim and Hopke (2007) applied PMF to measurements from STN sites in Simi Valley, downtown LA and Rubidoux. They apportioned $PM_{2.5}$ concentrations to major sources, including secondary nitrate (29-46%), secondary sulfate (12-23%), gasoline vehicles (12-22%), and diesel vehicles (7-10%). Contributions from crustal material, sea salt, aged sea salt, biomass combustion and incinerators were also identified. In a subsequent study, Kim et al. (2010) used PMF2 and EPA PMF (ME-2) to confirm these results. Ault et al. (2009) examined the impact of emissions from the LA port region during regional transport events. Although their focus was on downwind

locations in San Diego, they found that accumulation mode particles enriched in soot, nickel iron and vanadium are important tracers of secondary sulfate enriched particles in the region. During regional transport events, these particles contribute 2-4 times more mass than their primary emissions and are thus an important source of secondary sulfate downwind. Sardar and Sioutas (2005) measured coarse, fine and ultra-fine particle species at four sites in the LA Basin. They found that nitrate and organic carbon dominated the accumulation mode mass, whereas organic carbon dominated the ultra-fine mode mass. They also observed road dust and brake wear elements (Fe,V, Ti, Mn, Zn and Cu) to be correlated in the coarse mode, and also observed these same elements to a lesser extent in the fine mode. Docherty et al. (2008) examined the sources of primary and secondary organic carbon in Riverside, CA. They found that secondary organic aerosol was a major source (70-90%) of organic carbon during midday periods, with slightly less contribution during morning traffic periods (~45%). They used CMB with organic markers to apportion the primary organic carbon (POC) to fossil fuel emissions (64% from gasoline vehicles), with only a small contribution from biomass burning (5%). However, POC was only a small fraction of the total PM2.5, and concluded that the majority of the OC was from secondary formation processes the majority of the time, not just during severe smog episodes.

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