



Identification of ambient PM_{2.5} sources and analysis of pollution episodes in Detroit, Michigan using highly time-resolved measurements

Masako Morishita^{a,*}, Gerald J. Keeler^a, Ali S. Kamal^a, James G. Wagner^b, Jack R. Harkema^b, Annette C. Rohr^c

^a Air Quality Laboratory, School of Public Health, The University of Michigan, 1415 Washington Heights, Ann Arbor, MI 48109, USA

^b Pathobiology and Diagnostic Investigation, Michigan State University, East Lansing, MI, USA

^c Electric Power Research Institute, Palo Alto, CA, USA

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ABSTRACT

Recent studies have indicated that the chemical composition of ambient PM_{2.5}, or fine particulate matter (aerodynamic diameter <2.5 μm), may be closely associated with the health effects induced by PM. Thus, identification of the sources of these components of PM_{2.5} is critical to understanding source-specific health risks posed by PM_{2.5}. However, ambient PM measurements are typically 24-h average concentrations, masking the temporal variability of individual sources. These daily samples can limit our understanding of the associations between sources and observed health effect parameters. In the present study, highly time-resolved sampling methods were utilized to characterize trace element concentrations and to obtain information on potential emission sources impacting a site in urban Detroit where a complex mixture of local point and mobile sources, as well as regional sources, is observed.

Positive matrix factorization (PMF) was applied to 117 8-h PM_{2.5} filter samples from which six major factors were extracted including secondary aerosol, gasoline-powered vehicles, iron and steel manufacturing, diesel-powered vehicles, refining, and sludge incineration. In addition, PMF was applied to 268 30-min PM_{2.5} samples where six major source factors were identified including secondary aerosol, gasoline- and diesel-powered vehicles, iron and steel manufacturing, refining, sludge incineration, and cement/lime production.

Contributions of various emission source factors and peak concentrations for each factor were characterized using detailed meteorology and, factor directionality analysis. Although some variations were observed between the 8-h integrated filter samples and the 30-min SEAS samples, the results from the two datasets are generally consistent, suggesting that 46% and 43% of ambient PM, respectively, is derived from local sources. These findings have advanced our understanding of temporal PM source-receptor relationships in a complex urban air shed.

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1. Introduction

Recent studies have hypothesized that differences in the composition of particulate matter (PM) may be responsible for the variable health effects induced by PM (Peng et al., 2009; Hong et al., 2007; Lippmann et al., 2006; Seagrave et al., 2006). It is generally recognized that the composition and size of PM is largely determined by its source, making identification of its source, and its size-segregated components, critical to defining source-specific health risks. Most regulatory air quality monitoring, as well as measurements performed for epidemiological studies, derives data on the chemical composition of atmospheric PM from 24-h average fine

fraction, referred to as PM_{2.5}, samples. However, daily-averaged concentrations of PM_{2.5}, as well as meteorological parameters, tend to significantly reduce data variability and can therefore limit our ability to detect the associations between emission sources/components and health effect parameters. This is particularly apparent in studies performed in large urban areas, such as Detroit, where the impacts of emissions from local point and mobile sources are rapidly changing (less than hourly timescales), and are added to the complex mixture of regional source emissions transported into southeast Michigan that vary on longer time scales (~days). Daily and weekly measurements hinder the statistical decoupling of PM emission sources and their contributions over space and time.

Semi-continuous samplers and methodologies have enabled researchers to track pollutants and to characterize possible emission sources in the ambient air in almost real-time. Among these time-resolved approaches, the semi-continuous aerosol sampler (SEAS)

* Corresponding author. Tel.: +1 734 615 6635; fax: +1 734 936 7283.

E-mail address: tomoko@umich.edu (M. Morishita).

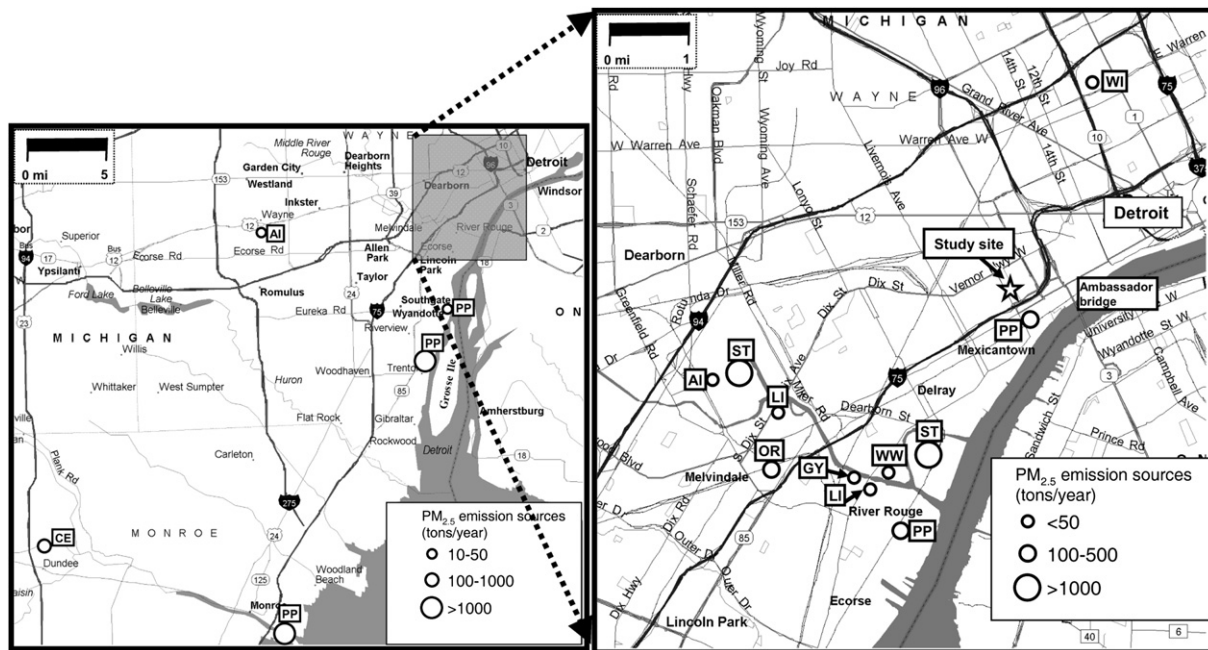


Fig. 1. Maps of southwest Detroit showing the location of the sampling site and major industrial sources for PM_{2.5} in Wayne and Monroe Counties, Michigan (USEPA, 2002) (AI: auto industries, CE: cement industries, GY: gypsum industries, LI: lime industries, OR: oil refineries, PP: coal-fired power plants, ST: iron/steel industries, WI: waste incinerator, and WW: waste water treatment and sludge incinerator).

has demonstrated the efficacy of high temporal-resolution ambient PM_{2.5} trace element determination for source characterization (Pancras et al., 2006; Ogulei et al., 2006; Kidwell and Ondov, 2001). Trace elements are particularly useful constituents of ambient PM as they can serve as tracers for specific emission sources and some do not transform significantly in the atmosphere (Henry, 1997). Moreover, numerous recent studies have reported that metal-enriched particles from combustion emission sources may be most closely associated with the adverse health impacts of ambient PM_{2.5} (Cavallari et al., 2008; Hirshon et al., 2008; Gavett et al., 2003; Samet et al., 2000) making semi-continuous monitoring of trace elements and identification of the elemental sources important to our understanding of source-specific morbidity and mortality.

In the present study, semi-continuous fine PM samples were collected as part of the Bi-City Concentrated Ambient Particle Study being conducted in Detroit and Steubenville, Ohio to investigate associations between a number of real-time cardiopulmonary endpoints, including heart rate variability, with the chemical and elemental composition of ambient PM_{2.5}; and ultimately the sources of the PM. Positive matrix factorization (PMF) modeling is used along with detailed meteorological analysis and particle morphological analysis to estimate the contribution of local emission sources to the measured PM. The aims of this paper are to: (1) present detailed source apportionment results for PM_{2.5} measured in Detroit, (2) compare the PMF results between 8-h integrated filter samples and semi-continuous elemental samples; and (3) evaluate the PMF modeling results using observed meteorological data and gaseous pollutant concentrations.

2. Materials and methods

2.1. Site description

AirCARE1, a 53-ft mobile air research laboratory, was located next to Maybury Elementary School in a residential neighborhood in southwest Detroit, where previous research studies have been

conducted to assess the impact of ambient PM and its components on human populations (Morishita et al., 2009; Baxter et al., 2008; Lewis et al., 2005). Fig. 1 is a map of the Detroit area and provides the location of AirCARE1 during the inhalation exposure studies. The map also indicates some of the major PM_{2.5} point sources in Wayne and Monroe Counties (USEPA, 2002). SW Detroit

Table 1 Summary of average composition of ambient PM_{2.5} measured by filter samples during the summers of 2004 and 2005.

	Means ± SD	
	2004 (N = 66)	2005 (N = 51)
Mass, μg m ⁻³	19 ± 10	23 ± 14
OC1, ng m ⁻³	733 ± 859	625 ± 275
OC2, ng m ⁻³	281 ± 208	279 ± 129
OC3, ng m ⁻³	400 ± 216	376 ± 173
OC4, ng m ⁻³	756 ± 349	737 ± 281
PC, ng m ⁻³	500 ± 366	762 ± 618
EC, ng m ⁻³	844 ± 422	943 ± 422
Mg, ng m ⁻³	28 ± 17	35 ± 22
Al, ng m ⁻³	82 ± 76	120 ± 55
S, ng m ⁻³	1884 ± 1401	2290 ± 1975
K, ng m ⁻³	70 ± 43	66 ± 84
Ca, ng/m ⁻³	173 ± 260	145 ± 109
Ti, ng m ⁻³	1.9 ± 1.5	1.6 ± 0.8
Mn, ng m ⁻³	6.2 ± 4.2	6.9 ± 6.1
Fe, ng m ⁻³	234 ± 185	281 ± 209
Ni, ng m ⁻³	4.2 ± 8.4	4.5 ± 6.6
Cu, ng m ⁻³	6.0 ± 11.7	4.1 ± 2.8
Zn, ng m ⁻³	62 ± 57	65 ± 95
As, ng m ⁻³	2.6 ± 2.1	1.4 ± 0.9
Se, ng m ⁻³	4.3 ± 3.8	1.7 ± 1.6
Rb, ng m ⁻³	0.2 ± 0.2	0.2 ± 0.1
Sr, ng m ⁻³	1.2 ± 1.0	0.9 ± 1.0
Cd, ng m ⁻³	0.5 ± 0.5	0.2 ± 0.4
Sb, ng m ⁻³	1.7 ± 1.7	1.5 ± 1.6
Ba, ng m ⁻³	6.7 ± 3.6	4.9 ± 2.5
La, ng m ⁻³	0.7 ± 1.8	0.3 ± 0.6
Ce, ng m ⁻³	0.5 ± 1.0	0.2 ± 0.4
Pb, ng m ⁻³	8.2 ± 5.8	7.1 ± 11.1

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