Atmospheric Environment 84 (2014) 65–77

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Constrained source apportionment of coarse particulate matter and selected trace elements in three cities from the multi-ethnic study of atherosclerosis

Timothy M. Sturtz [a], Sara D. Adar [b,c], Timothy Gould [a], Timothy V. Larson [a,c,*]

[a] Department of Civil and Environmental Engineering, University of Washington, Box 352700, Seattle, WA 98195-2700, USA
[b] Department of Epidemiology, School of Public Health University of Michigan, 1415 Washington Heights, Ann Arbor, MI 48109-2029, USA
[c] Department of Environmental and Occupational Health Sciences, University of Washington, Box 357234, Seattle, WA 98195-7234, USA

HIGHLIGHTS

- Receptor-oriented source apportionment using the multilinear engine.
- Modeled coarse particles sampled spatially across three urban regions.
- Applied source profile constraints using prior information.
- Separated ubiquitous and city-specific sources using a combined-cities model.

ARTICLE INFO

Article history:
Received 20 August 2013
Received in revised form
10 November 2013
Accepted 13 November 2013

Keywords:
Source apportionment
Coarse particulate matter
Air pollution
Positive matrix factorization
Brake wear
Tire wear
Soil
Agriculture
Trace elements

ABSTRACT

$PM_{10-2.5}$ mass and trace element concentrations were measured in Winston-Salem, Chicago, and St. Paul at up to 60 sites per city during two different seasons in 2010. Positive Matrix Factorization (PMF) was used to explore the underlying sources of variability. Information on previously reported $PM_{10-2.5}$ tire and brake wear profiles was used to constrain these features in PMF by prior specification of selected species ratios. We also modified PMF to allow for combining the measurements from all three cities into a single model while preserving city-specific soil features. Relatively minor differences were observed between model predictions with and without the prior ratio constraints, increasing confidence in our ability to identify separate brake wear and tire wear features. Brake wear, tire wear, fertilized soil, and resuspended soil were found to be important sources of copper, zinc, phosphorus, and silicon, respectively, across all three urban areas.

1. Introduction

There is ample evidence that long-term exposure to fine airborne particles ($PM_{2.5}$) is detrimental to human health (Pope and Dockery, 2006; U.S. EPA, 2009). In contrast, our understanding of the long-term effects of the coarse particle fraction ($PM_{10-2.5}$) is more limited (Dockery et al., 1993; Pope et al., 2002; Brunekreef and Forsberg, 2005; Weuve et al., 2012; Puett et al., 2009; Lippmann and Chen, 2009). One major challenge for chronic epidemiological studies is in accurately describing the long-term spatial gradients in coarse mode mass and species concentrations within urban areas. Recent work has focused on characterizing $PM_{10-2.5}$ spatial concentration gradients (Hwang et al., 2008; Godoy et al., 2009; Thornburg et al., 2009; Moore et al., 2010; Cheung et al., 2011; Eeftens et al., 2012a,b; Clements et al., 2012; Strak et al., 2011) and developing models to allow spatial interpolation (Yanosky et al., 2009; Peltier et al., 2011; Eeftens et al., 2012a). Another challenge is to characterize the sources that influence these gradients as well as the species that are associated with these sources.

Prior source apportionment studies of $PM_{10-2.5}$ have relied on either fully constrained models such as chemical mass balance

* Corresponding author. Department of Civil and Environmental Engineering, University of Washington, Box 352700, Seattle, WA 98195-2700, USA. Tel.: +1 206 543 6815; fax: +1 206 685 3836.
E-mail addresses: tlarson@u.washington.edu, tlarson@uw.edu (T.V. Larson).

1352-2310/$ – see front matter © 2013 Published by Elsevier Ltd.
http://dx.doi.org/10.1016/j.atmosenv.2013.11.031

(CMB), principal component analysis (PCA) and mass closure, (Paode et al., 1999; Manoli et al., 2002; Almeida et al., 2006; Stone et al., 2010; Daher et al., 2012; Oliveira et al., 2010; Waheed et al., 2012; Pakbin et al., 2011; Cheung et al., 2011), partially constrained models such as the constrained physical receptor model (COPREM) (Wahlin et al., 2006; Schauer et al., 2006a,b), or relatively unconstrained models such as factor analysis or positive matrix factorization (PMF) (Wang and Shooter, 2005; Gietl and Klemm, 2009; Begum et al., 2011; Begum, 2010; Kertész et al., 2010; J.S. Han et al., 2006; Oh, 2011; Tecer et al., 2012; Mazzei et al., 2007; Chan et al., 2008; Hwang et al., 2008; Godoy et al., 2009). Several of these studies have employed multiple sites within a city to capture spatial as well as temporal variability in the source contributions (Stone et al., 2010; Mazzei et al., 2007; Cheung et al., 2011; Chan et al., 2008; Hwang et al., 2008; Godoy et al., 2009; Pakbin et al., 2011).

While there has been a number of near-roadway studies examining the sources and components of non-exhaust $PM_{10-2.5}$ (Thorpe and Harrison, 2008; Harrison et al., 2012; Apeagyei et al., 2011; Schauer et al., 2006a,b; Han et al., 2011; Kennedy and Gadd, 2000; Garg et al., 2000; Iijima et al., 2008; Gietl et al., 2010; Grieshop et al., 2006; Bukowiecki et al., 2009; Von Uexküll et al., 2005; Sternbeck, 2002; Adachi and Tainosho, 2004; Johansson et al., 2009; Amato et al., 2011; Wahlström et al., 2009; Bukowiecki et al., 2010), only a few of the urban-scale source apportionment studies cited earlier have attempted to separate "road dust" into its separate components, including brake wear and tire wear (Wahlström et al., 2009; Amato et al., 2011; Schauer et al., 2006a,b; Bukowiecki et al., 2010; Harrison et al., 2012). The studies which did not separate road dust into its components commonly identified the dominant source of $PM_{10-2.5}$ as resuspended road dust for sites near roadways and as crustal material for non-roadway sites.

Here we use a partially constrained version of PMF (Amato et al., 2009; Reche et al., 2012; Brown et al., 2012) in order to examine the sources of $PM_{10-2.5}$ collected simultaneously at multiple sites in three urban areas during two-week periods in two different seasons. We use PMF with constraints imposed by prior knowledge of several important, ubiquitous source profiles, namely brake and tire wear. We furthermore impose additional constraints on the source contributions in order to combine all measurements into a single model. To our knowledge, this is the first application of a combined-cities PMF modeling approach with profile constraints to identify contributions of brake and tire wear in $PM_{10-2.5}$ across multiple urban areas. This work is part of a larger effort to examine the chronic health effects of $PM_{10-2.5}$ and selected species in these same cities under the auspices of the Multi-Ethnic Study of Atherosclerosis and Coarse Particulate Matter (MESA Coarse), an ancillary study of the Multi-Ethnic Study of Atherosclerosis and Air Pollution (MESA Air).

2. Materials and methods

2.1. Filter sampling and analysis

The MESA Air study leveraged the National Heart, Lung, and Blood Institute's Multi-Ethnic Study of Atherosclerosis (MESA) cohort to provide data for assessing the relationship between long-term exposures to fine ambient particulates and related health effects. The MESA cohort (Kaufman et al., 2012) was comprised of 6814 white, black, Hispanic, and Chinese participants located in six U.S. cities. As an ancillary study to MESA Air, MESA Coarse assesses the health implications associated with coarse mode particulate exposure in three of the MESA cohort cities, namely Chicago, Illinois, St. Paul, Minnesota, and Winston-Salem, North Carolina.

Paired, two-week average PM_{10} and $PM_{2.5}$ Teflon filter samples were simultaneously collected over two different two-week periods, in the winter and summer of 2009, in Chicago, IL, St. Paul, MN, and Winston-Salem, NC. The monitoring sites in each city (see Fig. 1) were residential locations of the existing MESA cohort selected to maximize variability in geographic features expected to influence coarse particles including land use, roadways, and vegetation as well as representative community monitoring sites. $PM_{10-2.5}$ mass concentrations were computed by the difference in collocated PM_{10} and $PM_{2.5}$ measurements. This "difference method" has been shown to be a reliable approach in estimating $PM_{10-2.5}$ in urban areas by the U.S. Environmental Protection Agency (Chen et al., 2011). At affiliated field centers in each sampled city, the Teflon filters were loaded into Harvard personal environmental monitors (HPEMs, Harvard School of Public Health, Boston, MA). These monitors were connected to a Medo VP0125 (MEDO USA, Inc., Roselle, IL) vacuum pump drawing 1.8 L min^{-1} air sample and equipped with a timer valve system that obtained a 50% duty cycle sample, where the flow alternated between the PM_{10} and $PM_{2.5}$ filter every 5 min to avoid filter overload.

PM_{10} and $PM_{2.5}$ mass concentrations were gravimetrically determined from weighing of Teflon filters at the University of Washington in a temperature and humidity controlled environment (Allen et al., 2001), and from the total volumetric flow of air sampled through the HPEMs. A Mettler-Toledo UMT-2 balance was used to determine sample mass following standard filter weighing procedures. Overall, the precision of duplicate PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ samples as measured by the average Relative Percent Difference was 2%, 10% and 18%, respectively. The filter samples were analyzed for a suite of 48 elements by X-Ray Fluorescence (XRF) at Cooper Environmental Services (Portland, OR). Method sensitivity was defined by a set of acceptable detection levels for a subset of 21 key elements from the Method IO-3.3 analyte list. The quality assurance and quality control data are provided in Tables A7 and A8.

2.2. PMF model inputs

Measurement uncertainty for coarse mode species j, σ_j, was calculated by combining the uncertainties of the PM_{10} and $PM_{2.5}$ measurements using standard error propagation as follows.

$$\left(\sigma_j^2\right)_{PM_{10-2.5}} = \left(\sigma_j^2\right)_{PM_{10}} + \left(\sigma_j^2\right)_{PM_{2.5}} \quad (1)$$

The measured coarse mode species concentrations were pre-processed to remove frequently below detection species and species with a signal to noise (Norris and Vedantham, 2008), S/N, <10. In addition, pre-processing included removal of sulfur samples identified as outliers (exceeding 2 standard deviations from the mean). Four samples were removed based on this criterion. The S/N cutoff choice was motivated by the consistently high signal to noise ratios of a subset of species and relatively low and variable ratios for some species depending upon city. Enrichment of the coarse mode for certain elements is not unexpected and has been documented in other literature (Amato et al., 2011b; Tecer et al., 2012; Carvalho and Freitas, 2011). The S/N criteria eliminated the following species: Ag, As, Au, Cd, Ce, Co, Cs, Eu, Ga, Hf, Hg, In, Ir, La, Mo, Nb, Rb, S, Sc, Se, Sm, Sn, Ta, Tb, V, W, and Y (see Table A1 in Appendix A). Although Sb had S/N < 10, we chose to include it in the models because of its value as a brake wear constraint variable described in the next section. We retained $PM_{10-2.5}$ mass but increased its uncertainty by a factor of 30 to avoid redundancy with all other measured species. The retention of coarse mass allows for the production of feature profiles in a gram per gram $PM_{10-2.5}$ basis. There were no missing