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Source apportionment of time and size resolved ambient particulate matter measured with a rotating DRUM impactor

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Abstract

Ambient particulate chemical composition data acquired from samples collected using a three-stage Davis Rotating-drum Universal-size-cut Monitoring (DRUM) impactor in Detroit, MI, between February and April 2002 were analyzed through the application of a three-way factor analysis model. $PM_{2.5}$ (particulate matter $\leqslant 2.5\,\mu m$ in aerodynamic diameter) was collected by a DRUM impactor with 3-h time resolution and three size modes ($2.5\,\mu m > D_p > 1.15\,\mu m$, $1.15\,\mu m > D_p > 0.34\,\mu m$ and $0.34\,\mu m > D_p > 0.1\,\mu m$). A novel three-way factor analysis model was applied to these data where the source profiles are a three-way array of size, composition and source while the contributions are a matrix of sample by source. Nine factors were identified: road salt, industrial (Fe + Zn), cloud processed sulfate, two types of metal works, road dust, local sulfate source, sulfur with dust, and homogeneously formed sulfate. Road salt had high concentrations of Na and Cl. Mixed industrial emissions are characterized by Fe and Zn. The cloud processed sulfate had a high concentration of S in the intermediate size mode. The first metal works represented by Fe in all three size modes and by Zn, Ti, Cu, and Mn. The second included a high concentration of small size particle sulfur with intermediate size Fe, Zn, Al, Si, and Ca. Road dust contained Na, Al, Si, S, K, and Fe in the large size mode. The local and homogeneous sulfate factors show high concentrations of S in the smallest size mode, but different time series behavior in their contributions. Sulfur with dust is characterized by S and a mix of Na, Mg, Al, Si, K, Ca, Ti, and Fe from the medium and large size modes. This study shows that the utilization of time and size resolved DRUM data can assist in the identification of sources and atmospheric processes leading to the observed ambient concentrations.

Keywords: High time resolution; Size distribution; Aerosol; Multilinear engine (ME); Rotating drum impactor (DRUM); Detroit; SXRF

1. Introduction

Multivariate receptor models have been used extensively for source apportionment of airborne particles (e.g., Henry, 1999, 2002; Hopke, 2003). The measured chemical composition data from the collected samples form a matrix. This matrix can be

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decomposed into two matrices representing sources contributions and sources profiles. Recently, analyses of particle size distribution data have also been performed to identify air pollution sources. Ruuskanen et al. (2001) analyzed ultrafine and accumulation mode particle data from three European cities to describe the differences in concentration levels and variations in particle number and mass concentrations between those cities. Wåhlin et al. (2001) quantified the emissions of ultrafine particles from diesel and petrol vehicles separately with respect to number and size distribution. Kim et al. (2004) applied positive matrix factorization to particle size distribution data measured in Seattle, WA and deduced four main sources contributing ambient $PM_{2.5}$ (particulate matter $\leqslant 2.5\,\mu m$ in aerodynamic diameter). Zhou et al. (2004, 2005) separated five sources from particle size distribution data measured in Pittsburgh, PA. Dillner et al. (2005) examined the application of cluster analysis to size distribution data to identify likely common sources of the observed species.

At the same time, methods that provide higher time resolution instruments ($\sim$1 h) have been developed for the measurement of airborne particulate species (Wilson et al., 2002). Several investigators (Khlystov et al., 1995; Slanina et al., 2001; Kidwell and Ondov, 2001; Orsini et al., 2003) have developed steam injection/impaction samplers that permit the collection of sufficient sample size to allow analyses for multiple species with time resolution as low as 30 min.

Several continuous sampling impactor systems (Tanaka et al., 1980; Cahill et al., 1981; Raabe et al., 1988; Shutthanandan et al., 2002) collect airborne particulate matter onto a substrate that permits subsequent time resolved elemental analysis. Initially, these analyses were performed using particle-induced X-ray excitation (PIXE), but there has been only limited use of these methods. One of these samplers, the Davis Rotating-drum Uniform-size-cut Monitoring (DRUM) impactor has been gaining popularity in recent years because recent work using synchrotron X-ray fluorescence (SXRF) on continuous or semi-continuous particle samples has suggested that short time (<1 h) resolution data can be obtained (Bench et al., 2002). However, data that contains both size and compositional information require advanced data analysis tools. Prior results (Dodd et al., 1991) suggest that source compositions are particle size dependent so that each size bin in the distribution would have a different composition for material coming from a given source. Thus, the data model must take such size–composition variation into account to properly resolve the ambient data. In this work, the combination of particle size and high time-resolution data were analyzed using a new model to permit the extraction of maximal information from the data. This model accounts for the variation in the composition of the source emissions in the different size ranges.

2. Data description

Samples were collected at the Allen Park site near Detroit, MI. Detroit is a major industrial center located in southeastern Michigan. The Allen Park site was located southwest of the Detroit industrial center. The site location is shown in Fig. 1.

Particles were collected and analyzed as a function of size, time, and chemical composition from 25 February to 10 April 2002 using a three-stage rotating DRUM impactor sampler. Particles were collected in three size modes, $2.5\,\mu m > D_p > 1.15\,\mu m$, $1.15\,\mu m > D_p > 0.34\,\mu m$, and $0.34\,\mu m > D_p > 0.1\,\mu m$ in diameter on lightly greased Mylar substrates with close to 100% of all samples successfully collected. The sample strips were analyzed in 3 h increments for elements for sodium and higher atomic number elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Zr, Mo, and Pb) using SXRF (Perry et al., 2004). These data are described in detail by Cahill et al. (2002).

Optical absorption data (Miller, 2002) at four wavelengths (350, 450, 550, and 650 nm) and the mass concentrations by β-gauge were also measured. A bare fiber directed light from a deuterium/tungsten–halogen fiber-optic light source onto the sample; a quartz collimating lens, attached to

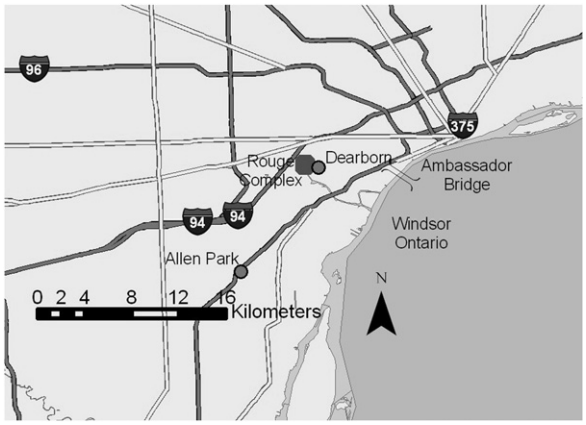

Fig. 1. Location of the Detroit area sampling site.