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Atmospheric aerosol over two urban–rural pairs in the southeastern United States: Chemical composition and possible sources

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Abstract

Positive matrix factorization (PMF) was used to infer the sources of $PM_{2.5}$ observed at four sites in Georgia and Alabama. One pair of urban and rural sites in each state is used to examine the regional and urban influence on $PM_{2.5}$ concentrations in the Southeast. Eight factors were resolved for the two urban sites and seven factors were resolved for the two rural sites. Spatial correlations of factors were investigated using the square of correlation coefficient (R^2) calculated from the resolved G factors. Fourier transform was used to define the temporal characteristics of $PM_{2.5}$ factors at these sites. Factors were normalized by using aerosol fine mass concentration data through multiple linear regression to obtain the quantitative factor contributions for each resolved factor. Common factors include: (1) secondary sulfate dominated by high concentrations of sulfate and ammonium with a strong seasonal variation peaking in summer; (2) nitrate and the associated ammonium with a seasonal maximum in winter; (3) "coal combustion/other" factor with presence of sulfate, EC, OC, and Se; (4) soil represented by Al, Ca, Fe, K, Si and Ti; and (5) wood smoke with the high concentrations of EC, OC and K. The motor vehicle factor with high concentrations of EC and OC and the presence of some soil dust components is found at the urban sites, but cannot be resolved for the two rural sites. Among the other factors, two similar industry factors are found at the two sites in each state. For the wood smoke factor, different seasonal trends are found between urban and rural sites, suggesting different wood burning patterns between urban and rural regions. For the industry factors, different seasonal variations are also found between urban and rural sites, suggesting that this factor may come from different sources or a common source may impact the two sites differently. Generally, sulfate, soil, and nitrate factors at the four sites showed similar chemical composition profiles and seasonal variation patterns reflecting the regional characteristics of these factors. These regional factors have predominantly low frequency variations while local factors such as coal combustion, motor vehicle, wood smoke, and industry factors have high frequency variations in addition to low frequency variations.

Keywords: Receptor modeling; $PM_{2.5}$; PMF; Urban; Rural; SEARCH; Factor contribution; Factor profiles; Time series analysis; Correlation coefficient

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1. Introduction

High concentrations of particulate matter (PM) are an important air pollution issue in the United States (Vedal, 1997; Rudell et al., 1999). Particles in the air may arise from a wide variety of natural or anthropogenic sources. Health effects of airborne particles have been studied extensively, and significant and positive associations between human mortality/morbidity and PM concentrations or some PM components have been observed in some studies but not in others (Dockery et al., 1993; Schwartz et al., 1993; Lipfert and Wyzga, 1995). More recent studies indicate that fine particles ($PM_{2.5}$, $<2.5\,\mu m$ in diameter) are more likely associated with adverse health effects (Gilliland et al., 2001; Peters et al., 2001; Pope et al., 2002) than other PM fractions.

In an effort to better characterize and understand the factors controlling near-surface PM concentrations in the Southeast, a multi-year study, the Southeastern Aerosol Research and Characterization project (SEARCH), was initiated in August 1998 and is scheduled to operate through 2005 (e.g. Hansen et al., 2003). SEARCH consists of 8 monitoring stations in 4 urban-rural pairs in 4 states: Alabama (North Birmingham [BHM] and Centreville [CTR]), Georgia (Atlanta [JST] and Yorkville [YRK]), Mississippi (Gulfport [GFP] and Oak Grove [OAK]), and Florida (Pensacola [PNS] and suburban Pensacola [OLF]). Measurements at each site include a wide range of gases (O_3, NO, NO_2, NO_y, HNO_3, SO_2, CO), PM mass ($PM_{2.5}$, $PM_{10-2.5}$), PM composition (elemental carbon (EC), organic carbon (OC), sulfate (SO_4), nitrate (NO_3), ammonium (NH_4), and trace metals), and meteorological parameters (wind speed, wind direction, temperature, relative humidity, barometric pressure, solar radiation, and rainfall).

Two objectives of the SEARCH study are to: (1) estimate the source contributions and (2) better understand the chemical composition of each source (Hansen et al., 2003; Zheng et al., 2002). Source apportionment studies using factor analysis have been previously conducted for the observations at the JST site from August 1998 to August 2000 (Kim et al., 2003a, b, 2004). These investigations provided important insight in the source apportionment at this site and showed some innovative use of speciated carbon fraction measurements.

Our main interests in the current study are to understand the urban-rural difference, the regional-local contrast, and the seasonal variations of the source-related factors. We apply positive matrix factorization (PMF) analysis (Chueinta et al., 2000; Lee et al., 1999; Paterson et al., 1999; Paatero and Tapper, 1993, 1994; Paatero, 1997; Polissar et al., 1998, 1999, 2001) to SEARCH $PM_{2.5}$ observations at four sites (two urban–rural pairs) in Alabama (urban BHM and rural CTR) and Georgia (urban Atlanta and rural Yorkville) from January 2000 to December 2002. By analyzing the urban-rural pairs, we examine their correlations influenced by regional sources and the factors contributing to urban–rural concentration gradients. To further examine the seasonal variations of the $PM_{2.5}$ factors, we make use of Fourier transforms to define the frequency variation of the factors. The power spectra provide a simple way to analyze the periodicity of time series data (Hies et al., 2000; Sebald et al., 2000). We expect the regional factors to have higher correlations among the sites and variations in lower frequencies than the local factors.

2. Sample collection and chemical analysis

$PM_{2.5}$ composition data analyzed in this study consist of the measurements taken at four sites of the SEARCH network (Fig. 1) (Hansen et al., 2003). In Georgia, the urban (JST) monitoring site is located 4 km northwest of downtown Atlanta; the rural (YRK) monitoring site is located 60 km northwest of the center of Atlanta. In Alabama, the urban (BHM) site is located 4 km north of downtown Birmingham; the rural (CTR) site is located about 70 km southwest of central Birmingham.

Daily integrated $PM_{2.5}$ samples were collected at the JST site. $PM_{2.5}$ samples were collected every third day at the BHM, CTR and YRK sites. Samples were collected by using particulate composition monitors (Atmospheric Research and Analysis, Inc., Durham, NC) that have three sampling lines (air flow rate $16.7\,l\,min^{-1}$) with inlets at 5 m above ground. More detailed descriptions can be found elsewhere (e.g., Kim et al., 2003a).

A total of 932 samples for the JST site, 336 samples for the BHM site, 347 samples for the YRK site and 338 samples for the CTR site were obtained and analyzed, covering the time period from January 2000 through December 2002. For each sample, concentrations of the following 19 chemical species were usually available:

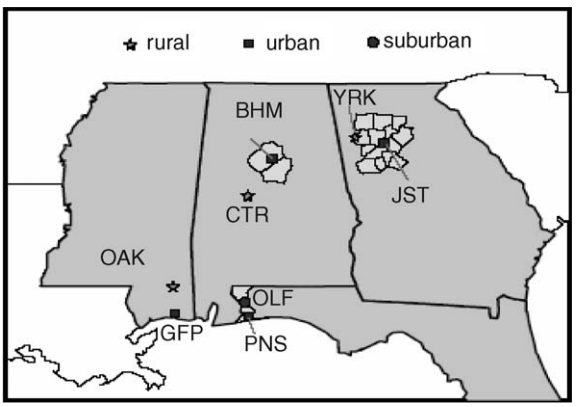

Fig. 1. Locations of SEARCH monitoring sites.