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Evaluation of the potential source contribution function using the 2002 Quebec forest fire episode

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Abstract

Particle data consisting of $PM_{2.5}$ (particulate matter $<2.5\,\mu m$ aerodynamic diameter), light scattering coefficient (b_{scat}), organic carbon (OC), elemental carbon (EC), and sulfate were measured for 2-h intervals from 1 July to 3 August 2002 in Philadelphia using semicontinuous monitors. The potential source contribution function (PSCF) combines the particle data with air parcel back trajectories to identify potential source areas and the preferred pathways that give rise to the observed high particle concentrations in Philadelphia. Five day back trajectories arriving 12 times a day at 500 m above ground level were calculated using NOAA HYSPLIT with the vertical mixing model. PSCF analyses were performed based on the average values as criterion values for the measured particle parameters. During this period, there was a major forest fire in central Quebec that penetrated the eastern United States to well south of Philadelphia, Pennsylvania. This event permits the testing of the ability of the PSCF analysis to identify the known source location of this fire, which is observed clearly in the maps for $PM_{2.5}$, OC and EC. The other maps can also be interpreted in terms of the likely sources of the observed concentrations.

Keywords: Receptor models; Potential source contribution function; $PM_{2.5}$; Long-range transport; Forest fire; Biomass burning

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1. Introduction

Atmospheric aerosols are implicated in a variety of environmental problems including acid rain, reduced visibility, modification of the atmospheric radiation balance, changing cloud properties, and causing adverse human health effects (EPA, 2004). Many epidemiological studies have now documented that elevated airborne particulate matter (APM) is associated with adverse health effects (Dockery et al., 1993; HEI, 2003). In order to reduce pollutant concentrations in the atmosphere, pollution sources must be identified, emission estimates made, contributions of various sources to the ambient concentrations estimated, and effective management strategies developed. A variety of receptor models have been developed to locate pollutant sources that combine meteorology with the measured chemical compositions so that the probable locations of emission sources can be found (Hopke, 2003). These methods combine estimates of the motion of the air backward in time with concentrations measured at a sampling site. These

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models include residence time analysis (RTA) (Poirot and Wishinshi, 1986; Poirot et al., 2001), quantitative transport bias analysis (QTBA) (Keeler and Samson, 1989), potential source contribution function analysis (PSCF) (Ashbaugh et al., 1985), concentration-weighted field (CWT) (Seibert et al., 1994), and residence time weighted concentration (RTWC) (Stohl, 1996).

In this study, the PSCF has been used to identify pollutant sources. It has been successfully applied in a number of studies (Ashbaugh et al., 1985; Zeng and Hopke, 1989; Cheng et al., 1993a,b, 1996; Gao et al., 1993, 1996; Hopke et al., 1995; Plaisance et al., 1996, 1997; Poissant, 1999; Cheng and Lin, 2001; Polissar et al., 2001a,b; Hafner and Hites, 2003; Hsu et al., 2003a,b; Biegalski and Hopke, 2004; Hoh and Hites, 2004; Wang et al., 2004). Thus, it is the most widely used trajectory ensemble model. The likely source areas observed in these studies have had good agreement with many known emission sources. However, there have been only limited opportunities to test the PSCF approach where there was a well-defined source that could be used to demonstrate the effectiveness of the method. Cheng and Lin (2001) have shown that PSCF can successfully locate emission source locations by examining data related to the 1998 Central American smoke events that had impacts on sampling sites in the Southern Great Plains. Given this one validation study, it is useful to examine other opportunities to test the utility of the PSCF method.

During early July 2002, there was a major forest fire in central Quebec whose plume penetrated down the coast of the eastern United States to well south of Philadelphia, where semi-continuous measurements were being made of a variety of airborne particle properties (Jeong et al., 2004a,b). This event permitted testing the ability of PSCF analysis to identify the known source location of this fire. The main objective of this study was to identify possible source locations of atmospheric aerosol by combining air parcel back trajectories with the semi-continuous particle data and ascertain if the PSCF analysis clearly defines the fire location.

2. Sample collection and chemical analysis

Particulate matter mass and composition were measured in Philadelphia from 1 July to 3 August 2002. Mass concentrations of $PM_{2.5}$, organic carbon, elemental carbon, black carbon, and sulfate were measured at a site northeast of Philadelphia (latitude 40.04°N, longitude 75.00°W). The monitoring site was located 400 m away from a light traffic road and 1 km away from a heavy traffic highway. It is approximately 15 km from downtown Philadelphia. Continuous $PM_{2.5}$ concentrations every thirty minutes were obtained by using a 30 °C Tapered Element Oscillating Microbalance (TEOM) (Rupprecht and Patashnick, Albany, NY) with a sample equilibration system (SES) dryer. Light scattering (b_{scat}) was measured using a Radiance Research Model 908 integrating nephelometer (lower detection limit and accuracy are about $10^{-6}\,\mathrm{m}^{-1}$ and 20%, respectively) with a Nafion drier on the inlet to reduce the influence of particle-bound water. Carbonaceous material in $PM_{2.5}$ was measured by a semi-continuous OC/EC field instrument (Sunset Laboratory, Forest Grove, Oregon, USA).

Sulfate concentrations were measured with high time resolution using a continuous sulfate analyzer originally designed by the Harvard school of public health (HSPH) (Allen, 2001) and built at Clarkson University. The continuous sulfate analyzer included three main parts, sample pretreatment system to segregate $PM_{2.5}$ using a sharp cut cyclone (SCC), and removal of positive and negative interferences such as SO_2 and NO_x using a sodium carbonate coated denuder and a carbon monolith. The heated converter (Thermo Environmental Model 350, Thermo Environmental Instruments Inc, Franklin, Massachusetts, USA) was operated at around 700 °C to reduce particulate sulfate to SO_2 and the SO_2 gas was introduced into an SO_2 analyzer (Thermo Environmental 43C-TLS). To estimate the background concentration, a zero particle purge system periodically supplied filtered air to the furnace that served as the blank values. These blank values were then subtracted from the measured concentrations. There were problems with the sulfate analyzer at the beginning of the sampling period and no data were collected until 6 July. Sulfate values were not corrected for the converter efficiency in the semicontinuous analyzer since the PSCF analysis only requires good relative values. The sulfate, $PM_{2.5}$, b_{scat}, OC, and EC data are described in detail by Jeong et al. (2004a,b). A 2-h time interval was chosen for sampling, analysis, and instrumental cool-down period.

As a quality control measure, 24-h samples were collected on prefired quartz filters. These filters were punched and measured for their integrated OC/EC concentrations using a laboratory Sunset OC/EC analyzer (Jeong et al., 2004a). The remaining portion of the filters were used for the analysis of the daily average sulfate using ion chromatography (Dionex 500, Dionex, Sunnyvale, California, USA) as described by Hopke et al. (2003). There was good agreement between the integrated filter values and the comparable 24-h averages derived from the semicontinuous measurements (Jeong et al., 2004a).

Fig. 1 shows the times series of the values of these variables during the period of the forest fire. During the first days of July, there was a sulfate episode that was not recorded because of the malfunction of the instrument. The fire episode began on 7 July and extended to 9 July. Further details are provided by Jeong et al. (2004b).