Available online at www.sciencedirect.com
SCIENCE DIRECT®
Atmospheric Environment 40 (2006) 1526–1537
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Source apportionment of PM2.5 in Beijing by positive matrix factorization

Yu Song[a], Yuanhang Zhang[a,*], Shaodong Xie[a], Limin Zeng[b], Mei Zheng[c], Lynn G. Salmon[d], Min Shao[b], Sjaak Slanina[b]

[a]*Department of Environmental Sciences, Peking University, Beijing 100871, China*
[b]*State Key Lab of Air Pollution Control and Simulation, Peking University, Beijing 100871, China*
[c]*School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA 30332-0340, USA*
[d]*Environmental Engineering Science Department, California Institute of Technology, Pasadena, CA 91125, USA*

Received 8 July 2005; received in revised form 19 October 2005; accepted 19 October 2005

Abstract

Air pollution associated with atmospheric fine particulate matter (PM2.5, i.e., particles with an aerodynamic diameter of 2.5 μm or less) is a serious problem in Beijing, China. To provide a better understanding of the sources contributing to PM2.5, 24-h samples were collected at 6-day intervals in January, April, July, and October in 2000 at five locations in the Beijing metropolitan area. Both backward trajectory and elemental analyses identified two dust storm events; the distinctly low value of Ca:Si (<0.2) and high Al:Ca (>1.7) in Beijing PM2.5 appear indicative of contributions from dust storms. Positive matrix factorization (PMF) was used to apportion sources of PM2.5, and eight sources were identified: biomass burning (11%), secondary sulfates (17%), secondary nitrates (14%), coal combustion (19%), industry (6%), motor vehicles (6%), road dust (9%), and yellow dust. The lower organic carbon (OC), elemental carbon (EC), SO_4^{2-}, and Ca values of yellow dust enable it to be distinguished from road dust. The PMF method resolved 82% of PM2.5 mass concentrations and showed excellent agreement with a previous calculation using organic tracers in a chemical mass balance (CMB) model. The present study is the first reported comparison between a PMF source apportionment model and a molecular marker-based CMB in Beijing.

Keywords: China; Fine particle; Dust storm; Source profiles; Backward trajectories

1. Introduction

Beijing, the capital of China, suffers from air pollution that has risen dramatically since the onset of rapid urbanization in the 1980s. The concentration of PM10 (particles with aerodynamic diameters of less than 10 μm) monitored by the Beijing Environmental Protection Bureau (BJEPB) during 2000–2004 showed particulate matter to be the major problem in Beijing (BJEPB, annual reports for 2000–2004). In these years, the annual PM10 concentrations ranged between 140 and 165 $\mu g\,m^{-3}$, while the PM2.5 exhibited mass concentrations of

*Corresponding author. Tel.: +86 10 62756592; fax: +86 10 62751927.
E-mail addresses: songyu@pku.edu.cn (Y. Song), yhzhang@pku.edu.cn (Y. Zhang).

1352-2310/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2005.10.039

$>100\,\mu g\,m^{-3}$, or about 60% of the PM10 concentrations in Beijing (He et al., 2001). PM2.5 is considered to be the more harmful to human respiration, as well as being responsible for degrading visibility (Bergin et al., 2001), and any efforts to improve the air quality of Beijing will require a clear understanding of the potential PM2.5 sources.

Several studies have identified possible sources of particulate matter in Beijing. He et al. (2001) used the chemical composition of PM2.5 collected from sites at Tsinghua University and Chegongzhuang from July 1999 to September 2000, and suggested sources to be dust storms, motor vehicle emissions, and biomass burning. Duan et al. (2004) demonstrated that summer burning of biomass could contribute 46–70% of the organic carbon (OC) in aerosols at the Ming Tombs, a rural site, and 10–43% of the OC at the Temple of Heaven, an urban site. Dan et al. (2004) regarded biomass burning and traffic and/or industry emissions as providing the major sources of OC and elemental carbon (EC) during summer, with coal consumption being the dominant contributor in winter. More recently, Zheng et al. (2005) used the chemical mass balance receptor model (CMB) to apportion OC in fine particles and the total PM2.5 mass as well. They concluded that the major sources of PM2.5 in Beijing were dust, secondary sulfate, secondary nitrate, coal combustion, mobile sources, secondary ammonium, biomass aerosol, cigarette smoke, and vegetative detritus. However, some of the source profiles used for their CMB model were the ones derived in the United States and may not be directly applicable in Beijing.

Unfortunately, measurements of source profiles have a difficult methodology and are time-consuming. A different but highly effective tool exists in using factor analysis to apportion sources without the chemical profiles, and in China, this option offers a viable alternative given the absence of a local source profile library.

The novel analysis provided by positive matrix factorization (PMF) is a powerful technique for particle apportionment. Cities where it has been applied successfully include Hong Kong (China; Lee et al., 1999), Toronto (Canada; Lee et al., 2003), Atlanta (USA; Kim et al., 2004), and Pittsburg (USA; Zhou et al., 2004). An excellent review of PMF modeling is presented in Hopke (2003).

In this study, we used the PMF method to determine PM2.5 sources in Beijing using the raw data from Zheng et al. (2005). Interpretation of the results was aided by comparing them to those obtained from the CMB model.

2. Methods

2.1. Data description

In January, April, July, and October 2000, PM2.5 samples were collected in Beijing for 24 h at five sites simultaneously at 6-day intervals. The five sampling sites are shown in the Supplemental Materials as Fig. S1, including the Ming Tombs (OT), the airport (NB), Beijing University (BJ), Dong Si EPB (XY), and Yong Le Dian (CH). Details of the sites are given in Zheng et al. (2005). In total, 100 samples were taken over 4 months. Three parallel filters were collected during each 24-h sampling period. For each sample, the 24-h PM2.5 mass concentrations were obtained and the chemical composition was then analyzed for sulfate, nitrate, chloride, and ammonium ions by ion chromatography (IC) and for metals by X-ray fluorescence (XRF) spectroscopy. The OC and EC were determined by NIOSH thermal-optical procedures. The detailed organic speciation obtaining at monthly was ascertained by GC/MS. Details of the particulate sampling procedures and the analytical methods are given in Zheng et al. (2005).

Most of the data presented in this paper are the same as those used by Zheng et al. (2005), which contained monthly data, particularly of organic tracers, averaged over 5 samples from each month. In addition, this paper considers the 24-h data, which include the OC, EC, and inorganic tracers, but the monthly organic speciation is not included.

2.2. Model description

The PMF model was developed by Paatero and Tapper (1994) and Paatero (2004). A conventional factor analysis model can be written as,

$$X = GF + E, \tag{1}$$

where X is the $n \times m$ matrix of ambient element concentrations, G is the $n \times p$ matrix of source contributions, F is the $p \times m$ matrix of source profiles, and E is the matrix of residuals not fitted by the model and is defined as

$$e_{ij} = x_{ij} - \sum_{k=1}^{p} f_{ik} g_{kj}, \tag{2}$$