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Source apportionment of Beijing air pollution during a severe winter haze event and associated pro-inflammatory responses in lung epithelial cells

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HIGHLIGHTS

- Daily $PM_{2.5}$ samples were collected in Beijing during an episode of severe haze in January, 2013.
- The CMB model was used to identify PM sources and quantify emissions for $PM_{2.5}$.
- Secondary sources and coal combustion were the largest contributors to ambient PM during haze episodes.
- Secondary sources and coal combustion are vital contributors to the health burden of air pollution.

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ABSTRACT

Air pollution is a leading risk factor for the disease burden in China and globally. Few epidemiologic studies have characterized the particulate matter (PM) components and sources that are most responsible for adverse health outcomes, particularly in developing countries. In January 2013, a severe haze event occurred over 25 days in urban Beijing, China. Ambient fine particulate matter ($PM_{2.5}$) was collected at a central urban site in Beijing from January 16-31, 2013. We analyzed the samples for water soluble ions, metals, elemental carbon (EC), organic carbon (OC), and individual organic molecular markers including n-alkanes, hopanes, PAHs and sterols. Chemical components were used to quantify the source contributions to $PM_{2.5}$ using the chemical mass balance (CMB) model by the conversion of the OC estimates combined with inorganic secondary components (e.g. NH_4^+, SO_4^{2-}, NO_3^-). Water extracts of PM were exposed to lung epithelial cells, and supernatants recovered from cell cultures were assayed for the pro-inflammatory cytokines by a quantitative ELLSA method. Linear regression models were used to estimate the associations between PM sources and components with pro-inflammatory responses in lung epithelial cells following 24-hrs and 48-hrs of exposure. The largest contributors to $PM_{2.5}$ during the monitoring period were inorganic secondary ions (53.2% and 54.0% on haze and non-haze days, respectively). Other organic matter (OM) contributed to a larger proportion of $PM_{2.5}$ during haze days (16.9%) compared with non-haze days (12.9%), and coal combustion accounted for 10.9% and 8.7% on haze and non-haze days, respectively. We found $PM_{2.5}$ mass and specific sources (e.g. coal combustion, traffic emission, dust, other OM, and inorganic secondary ions) were highly associated with inflammatory responses of lung epithelial cells. Our results showed greater responses in the exposure to 48-hr $PM_{2.5}$ mass and its sources compared to 24-hr PM exposure, and that secondary and coal combustion sources play an important role in short-term inflammation and require cost-effective policy to control their contributions to air pollution.

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1. Introduction

Air pollution is the leading environmental health risk factor in China (Yang et al., 2013) and globally (Lim et al., 2013). A causal relationship between exposure to ambient air pollution, particularly particles <2.5 microns in diameter ($PM_{2.5}$), and increased cardiovascular morbidity and mortality is well established (Brook et al., 2010). Even small increases in daily PM exposure are associated with cardiovascular hospitalizations and all-cause mortality in higher income countries (Burnett et al., 1999; Crouse et al., 2012; Lim et al., 2013) and urban China (Chen et al., 2012). The exact mechanisms by which combustion-generated PM affects cardiopulmonary risk are not entirely understood, though inflammatory mediators that are released from lung cells after contact with PM are thought to be central (Brook et al., 2010). Experimental studies in animals and humans have demonstrated increased cellular and pro-inflammatory cytokine content (e.g., IL-6, TNF-α, IL-8) in bronchial fluid and in circulating blood after acute exposure to urban PM (Becher et al., 2007; Harder et al., 2001; Eeden et al., 2001).

Increasing evidence suggests that the chemical composition in PM from different sources may affect its impact on human health (Bell and HEI Health Review Committee, 2012; Baumgartner et al., 2014). Studies in urban Los Angeles found associations between the specific chemical components of PM with biomarkers of systemic inflammation in older adults (Delfino et al., 2010a, b). In particular, polycyclic aromatic hydrocarbons (PAHs) and hopanes were more strongly associated with blood inflammation cytokines (e.g. IL-6) than were transition metals (e.g., V, Cr, Mn, Fe, Ni, Cu, Zn) and other organic components (e.g., n-alkanes, organic acids). A small number of studies have investigated the health impacts of PM components and sources in developing countries (Baumgartner et al., 2011, 2014; Wu et al., 2014). While there are many shared PM components in developed and developing countries, the concentrations, sources, and relative contribution of different sources to the PM mixture differ considerably (Rohr and Wyzga, 2012). Air pollution in urban Beijing is comprised of relatively higher concentrations of sulfur dioxide (SO_2), nitrogen dioxide (NO_2), black carbon (BC) and primary organic carbon (POC) than higher income regions, likely a result of traffic and greater fossil fuel and coal combustion from power plants (Lin et al., 2014). The average contribution of the mobile sources to $PM_{2.5}$ OC in central Los Angles (30%) was considerably higher than that in Beijing (13–19%), while significantly higher contribution of coal combustion to $PM_{2.5}$ OC in Beijing (2%–9%) was observed compared to that in central Los Angles (0%) (Heo et al., 2013; Zheng et al., 2005; Wang et al., 2009). Consequently, ozone production in Beijing is largely due to the oxidation of anthropogenic carbonaceous aerosols, which is different from ozone in the Eastern US which is largely attributed to the oxidation of reactive hydrocarbons (Tie et al., 2006). It is therefore plausible that urban PM in Beijing may share common toxicities with PM in higher-income countries as well as bear unique biological consequences.

In January of 2013, an extended episode of severe haze occurred in northern China, which involved the largest number of haze days (25) observed in a single month since 1961 (Zhang et al., 2014a,b,c; Huang et al., 2014). Daily ambient $PM_{2.5}$ concentrations reached 755 μg m^{-3}, which is over 30 times higher than the WHO daily Air Quality Guideline (25 μg m^{-3}) (Zhang et al., 2014a,b,c; Huang et al., 2014). The atmospheric background field analysis indicated that the lower atmospheric pressure over mainland China and higher atmospheric pressure over the ocean to the east of China contributed to the abnormally weak East Asian winter monsoon over eastern China. Corresponding, anomalous southerly winds were dominated and under the control of the lower troposphere, which are conditions more favorable for transporting vapors into Beijing (Zhang et al., 2014a,b). As a result, an extended period of very low visibility (mean = 3.4 km ± 3.0) began on January 16 and continued through January 31 (Table S1), suggesting the meteorological conditions of haze weather (QX/T 113–2010). During this period, the worst visibility was recorded on January 29 (1.8 km), similar to average visibility (1.9 km) of serious fog days when relative humidity approaches 100%; saturated (Ding et al., 2014). Higher visibility days were observed on January 17 and January 24–26 (mean = 22.7 ± 16.8 km), primarily associated with elevated wind speed (mean: non-haze days versus haze days: 2.6 ± 1.8 m s^{-1} versus 1.5 ± 0.4 m s^{-1}). This is consistent with meteorological data indicating that a cold air front led to windy and cold air conditions throughout northeastern China during these four days (Supplementary information; Fig. S1).

We measured 24-hr ambient $PM_{2.5}$ concentrations during the haze episode at a central site in urban Beijing, and assessed its mass and chemical composition. The composition data were used to quantify the source contributions to the haze particles using the chemical mass balance (CMB) model. We also assessed whether the PM source contributors were associated with increased secretions of two pro-inflammatory cytokines, namely interleukin-6 (IL-6) and tumor necrosis factor alpha-α (TNF-α), in human lung epithelial cell lines. This study provides new information to better target PM composition and sources for emissions abatement, especially as parallel studies optimize the air quality regulations for countries like China with large populations that face considerable health risks associated with exposure to air pollution.

2. Materials and methods

2.1. Air pollution measurement

We collected outdoor 24-hr $PM_{2.5}$ samples from January 16–31, 2013 at a central site (N39°59′24.20″, E116°12′16.85″) in urban Beijing. The site is located next to one of the city's major highways. Air pollution monitors were placed on the roof of a building at a height of ~30 m. We collected integrated $PM_{2.5}$ mass on 90-mm quartz microfiber filters (prebaked, QMA, Whatman-GE Healthcare Biosciences Corp) using air impactor monitors (Qingdao Laoying Corp, Qingdao, China) that were operated at a flow rate of 100 L min^{-1}, as recommended by China's standard method for $PM_{2.5}$ collection (HJ93-2013). The quartz filters were baked for 4-hrs at 550 °C and conditioned in desiccators for 24-hrs prior to deployment for PM measurement. Filters were changed every morning at 10:00am during the measurement period. We collected an average of one field filter blank for every 6 samples. Blank filters were collected separately before and after the sampling for about 20 min by mounting blank filters onto the samplers without pumping into any air. The PM samples were individually sealed into an aluminum foil package and stored at −18 °C prior to gravimetric and chemical analysis. Daily atmospheric pressure, temperature, wind speed, and relative humidity were recorded using a Kestrel 3000 meteorological station (Kestrel Instrument, USA). Air mass back trajectories were calculated using National Oceanic and Atmospheric Administration (NOAA)'s ARL HYSPLIT 4.0 model (with meteorological data from the Global Data Assimilation System, GDAS) (Draxler and Rolph, 2013). We examined the impact of metrological factors on haze occurrence frequency during the measurement period, as recommended by standard methods in China for observing and forecasting haze weather (QX/T 113-2010) (Table S1) (Zhang et al., 2014a,b,c; Wang et al., 2014). Outdoor visibility was measured using a Model 6000 visibility sensor (Belfort Instrument, USA). Haze was defined as a visibility of <10 km, relative humidity of <95%, and a $PM_{2.5}$ mass