Science of the Total Environment 571 (2016) 902–908

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Response of aerosol composition to different emission scenarios in Beijing, China

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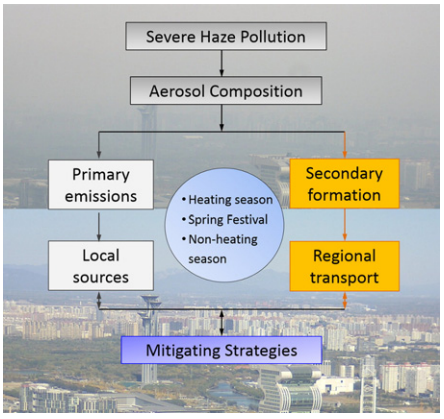
HIGHLIGHTS

- The response of aerosol composition to three different emission scenarios is elucidated
- Fossil organic aerosols from coal combustion and traffic emissions are significantly elevated in heating season
- Changes in anthropogenic activities during spring festival reduce cooking and traffic related aerosols substantially
- Local emission controls during haze episodes have small impacts on PM reductions due to enhanced secondary formation

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 8 June 2016
Received in revised form 10 July 2016
Accepted 11 July 2016
Available online 15 July 2016

Editor: D. Barcelo

Keywords:
Aerosol composition
Source emissions
Primary aerosol

ABSTRACT

Understanding the response of aerosol chemistry to different emission scenarios is of great importance for air pollution mitigating strategies in megacities. Here we investigate the variations in air pollutants under three different emission scenarios, i.e., heating season, spring festival holiday and non-heating season using aerosol composition and gaseous measurements from 2 February to 1 April 2015 along with source apportionment and FLEXPART analysis in Beijing. Our results showed substantially different aerosol composition among three emission scenarios that is primarily caused by different emission sources. All aerosol and gas species showed ubiquitously higher concentrations in heating season than non-heating season with the largest enhancement for fossil OA (FOA) and chloride. On average, the particulate matter (PM) level in winter heating season can be enhanced by 70% due to coal combustion emissions. In contrast, cooking aerosols and traffic related species showed significant reductions as a response of reduced anthropogenic activities during the spring festival holiday, sulfate and secondary organic aerosol (SOA) however even increased due to enhanced aqueous-phase production. Such

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http://dx.doi.org/10.1016/j.scitotenv.2016.07.073
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Secondary formation
Beijing

compensating effects resulted in small changes in PM levels for haze episodes during the holiday period despite reduced anthropogenic emissions. Our results have significant implications that local emission controls during winter severe pollution episodes can reduce primary aerosols substantially, but the mitigating effects can be significantly suppressed by enhanced secondary formation under stagnant meteorological conditions.

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1. Introduction

Air pollution resulted from high levels of atmospheric particulate matter (PM) has become a serious environmental issue in the developed regions of China. PM has great impacts on radiative forcing (Boucher et al., 2013), visibility deterioration (Zhang et al., 2010), air quality (Chan and Yao, 2008) and public health (Chen et al., 2013). Therefore mitigating PM levels is crucial to improve air quality in China. However this remains a great challenge as a result of (1) very high PM levels (e.g., 80.6 μg m^{-3} for the annual average $PM_{2.5}$ in 2015 in Beijing, http://www.bjepb.gov.cn/bjepb/413526/413663/413717/413719/index.html), (2) unclear sources, particularly secondary aerosols, (3) incomplete understanding of aerosol processes during stagnant meteorological conditions, and (4) nonlinear response of aerosol concentrations to source emission changes.

Air pollution in megacities strongly depends on primary source emissions and secondary formation. The emission sources can have significant changes during "special" events, e.g., 2008 Olympic Games, 2014 Asia-Pacific Economic Cooperation (APEC) conference, and 2015 Parade in Beijing. Strict emission controls in Beijing and surrounding regions were performed during these special events to ensure good air quality, which provided experimental opportunities to explore the impact of emission controls on haze pollution. For example, primary pollutants were observed to have significant reductions during the 2008 Olympic Games, secondary aerosols and ozone however showed increases after the full emission controls (Wang et al., 2010; Zhang et al., 2009), highlighting the different responses of primary aerosols and secondary species to emission changes. In contrast, secondary aerosols were observed to have the most reductions during the APEC period, while the changes in primary aerosols were much smaller (Sun et al., 2016; Xu et al., 2015). These results indicate the complex responses of aerosol chemistry to emission controls. Compared to these special events, the Chinese spring festival and heating season are more unique in studying the impacts of local emission controls and coal combustion emissions on PM pollution in winter. For example, Wang et al. (2015) observed an enhancement of PM_1 by 50% during heating period, and the increase was mainly contributed by organics, sulfate, and chloride from coal combustion emissions. Comparatively, the variations in anthropogenic sources during the holiday period exerted large impacts on the reductions in cooking-related aerosols, coal combustion aerosols and nitrogen monoxide, yet had small impacts on secondary species (Jiang et al., 2015). Although a few studies have investigated the responses of aerosol chemical properties under various emission scenarios (Huang et al., 2012; Jiang et al., 2015; Wang et al., 2015), our knowledge of the response of aerosol chemical properties to source emission changes is far from complete. Particularly, meteorological conditions and source areas could vary substantially in different years and different emission scenarios, which can have highly uncertain impacts on evaluating the impacts of emission controls.

In this work, we have characterized the variations in aerosol composition, sources, and process under three different emission scenarios, including heating season with significantly enhanced coal combustion emissions, spring festival holiday with largely reduced traffic and cooking emissions, and post-heating season, using highly time-resolved particle composition and gaseous measurements along with positive matrix factorization (PMF) and FLEXPART model analysis. In addition, the impacts of emission changes on primary and secondary aerosol species are also characterized by isolating meteorological effects and the influences of source areas. Our results have significant implications for policy makers to make control strategies in mitigating severe haze pollution in megacities.

2. Experimental method

2.1. Sampling site and measurements

The non-refractory submicron aerosol (NR-PM_1) species, gas precursors, and optical properties were measured at Institute of Atmospheric Physics, Chinese Academy of Sciences (39°58′28″N, 116°22′16″E, ASL: 49 m), a typical urban site located between the north 3rd and 4th ring roads, from 2 February to 1 April 2015. The sampling site is situated on the roof of a two-story building, approximately 40 m from the nearest traffic road to the north and ~400 m from Jingzang highway to the east. A more detailed description of the sampling site can be found in Sun et al. (2012b).

Organics (Org), nitrate (NO_3), sulfate (SO_4), ammonium (NH_4) and chloride (Chl) were measured in situ with an Aerodyne Aerosol Chemical Speciation Monitor (ACSM), and black carbon (BC) was measured with a "dual-spot" Aethalometer (model: AE33, Magee Scientific Corp.) (Drinovec et al., 2015). The light extinction and scattering coefficients of dry $PM_{2.5}$ at 630 nm were measured with a state-of-the-art Cavity Attenuated Phase Shift Single Scattering Albedo Monitor (CAPS PM_{ssa}) (Onasch et al., 2015). In addition, the gas precursors, e.g., O_3, NO, NO_x, and SO_2 were measured with multiple gas analyzers (Thermo Scientific), and the $PM_{2.5}$ was measured with a TEOM monitor. Meteorological variables including winds (wind direction and wind speed), temperature (T) and relative humidity (RH) were measured at 15 different heights on a meteorological tower, approximately 20 m to the sampling site. The meteorological parameters RH and T, and inorganic components determined by the ACSM were then input into a thermodynamic equilibrium model, ISORROPIA-II (Nenes et al., 1998) to predict liquid water content (LWC) that is related to inorganic components.

2.2. ACSM data analysis

The ACSM data was analyzed for the mass concentrations of NR-PM_1 species using the ACSM standard software (v2). A collection efficiency (CE) that varies as a function of the mass fraction of ammonium nitrate (Middlebrook et al., 2012) was used to compensate for the incomplete detection of aerosol particles which is primarily caused by particle bounce effects (Matthew et al., 2008). The impacts of RH and particle acidity on CE were small due to dry and overall neutralized submicron aerosol particles in this study (Middlebrook et al., 2012). As indicated in Fig. S1, PM_1 ($=NR - PM_1 + BC$) was correlated with $PM_{2.5}$ ($r^2 = 0.74$), and the regression slope (0.58) overall agrees with previous results in Beijing (Sun et al., 2013b; Wang et al., 2015). In addition, the extinction coefficient (b_{ext}) was tightly correlated with PM_1, and the mass extinction efficiency (5.1 m^2 g^{-1}) is also in agreement with the values in previous studies (Jiang et al., 2015). These results together indicate that the choose of CE of 0.5 is reasonable in this study.

PMF with the PMF2.exe algorithm (Paatero and Tapper, 1994) was applied on organic aerosols (OA) spectral matrices to identify potential OA factors. The PMF factors were evaluated using a PMF Evaluation Tool (PET, v 2.08D) (Ulbrich et al., 2009) and a three-factor solution was selected which includes a fossil OA (FOA) from traffic and coal combustion emissions, a cooking-related OA (COA), and an oxygenated OA (OOA).

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