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Characterization of coal burning-derived individual particles emitted from an experimental domestic stove

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ABSTRACT

Coal combustion in the domestic stoves, which is common in most parts of the Chinese countryside, can release harmful substances into the air and cause health issues. In this study, particles emitted from laboratory stove combustion of the raw powder coals were analyzed for morphologies and chemical compositions by using transmission electron microscopy (TEM) coupled with energy-dispersive X-ray spectrometry (EDX). The coal burning-derived individual particles were classified into two groups: carbonaceous particles (including soot aggregates and organic particles) and non-carbonaceous particles (including sulfate, mineral and metal particles). The non-carbonaceous particles, which constituted a majority of the coal burning-derived emissions, were subdivided into Si-rich, S-rich, K-rich, Ca-rich, and Fe-rich particles according to the elemental compositions. The Si-rich, S-rich and K-rich particles are commonly observed in the coal burning emission. The proportions for particles of different types exhibit obvious coal-issue dependence. Burning of coals with high ash yields could emit more non-carbonaceous particles, and burning of coals with high sulfur content can emit more S-rich particles. By comparing the S-rich particles from this coal burning experiment with those in the atmosphere, we draw a conclusion that some S-rich particles in the atmosphere in China could be mainly sourced from coal combustion.

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Introduction

The industrialization and urbanization in developing countries have led to severely deterioration of air quality (Li et al., 2007; Streets et al., 2008). Airborne particles are an important and complex constituent of the atmospheric system (Li and Shao, 2009), and they are often affected by meteorological conditions, such as temperature and relative humidity (Niu et al., 2016). Recently, in China, haze due to fine particles (*i.e.*, $PM_{2.5}$) pollution has occurred more frequently, resulting in serious influence on air quality, regional and global climate change, and human health, which has drawn great concern (Xu et al., 2003; Shao et al., 2006; Reiss et al., 2007; Zhang et al., 2012; Huang et al., 2014; Pokhrel et al., 2015). In the first quarter of 2013, China suffered from extremely severe and persistent haze pollution, influencing 1.3 million km^2 and 800 million people (Huang et al., 2014).

Although atmospheric particle pollution may be affected by meteorological factors in the air, the emission sources also

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play an important role. The fundamental reason for haze formation is the emission of particulate matter and gaseous species from fossil fuel combustion and biomass burning. A literature review demonstrated that the $PM_{2.5}$ from coal combustion and vehicle emissions is the dominant contributors to regional haze formation in China (Pui et al., 2014). Heavy haze episode occurred frequently in North China during spring and winter in recent years, which had a profound relationship with the coal combustion emission. The domestic combustion stoves were widely used for heating and cooking in the rural areas in China (Geng et al., 2012; Li et al., 2012; Geng et al., 2014). The vast household stoves fueled by coal have been the subject of interest recently due to the significant emission sources of various pollutants, including particulate matter, organic carbon, black carbon, greenhouse gases and toxic organic compounds (Finkelman et al., 1999; Streets et al., 2003; Zhang et al., 2009; Lei et al., 2011; Chen et al., 2015). Emissions from coal combustion represent an important source of gaseous and particulate pollutants in the atmosphere (Dockery et al., 1993; Andreae and Merlet, 2001; Kan et al., 2007; Jones et al., 2009). Some researchers showed that coal combustion emissions have a serious impact on visibility (Chen et al., 2015) and climate change (Kim et al., 2015). Coal burning emission has been regarded as one of the major sources of the atmospheric pollution in China (Sambandam et al., 2015; Lv et al., 2016; Yan et al., 2016).

China is the largest coal producer and consumer in the world. In 2014, China consumed approximately 2.81 billion tons of coal, which was equal to approximately 66% of the primary energy consumed in China (China Statistical Yearbook, 2015). Approximately 25% of the coal production in China is high sulfur coal, with a sulfur content exceeding 2 wt.%, and the burning of these high-sulfur coals discharges SO_2, together with NO_2 and particulate matter, into the atmosphere, resulting in atmospheric pollution (Chen et al., 2015; Saikia et al., 2015). In China, the consumption of raw coal was very large, especially in northern region, and burning raw coal can generate high levels of particles and thus have negative effect on atmospheric environment (Chai et al., 2016).

Although coal combustion is regarded as one of the major sources of air pollution, the estimated contribution of coal combustion emission to atmospheric particulate matter pollution vary greatly, from 18.2% (Chen et al., 2006) to 57% (Zhang et al., 2013). One of the main reasons for this issue is the uncertain emission inventory of coal combustion. A number of off-line chemical analyses have been carried out on the coal burning-derived particulate matters (Geng et al., 2012; Spears, 2013; Geng et al., 2014; H.F. Zhang et al., 2014; Chen et al., 2015; Saikia et al., 2015) and atmospheric particles (Gligorovski et al., 2008; Healy et al., 2013; Li et al., 2016; Gemayel et al., 2017). Some on-line techniques such as sing particle mass spectrometers were also used to study the sources of particles in the air (Bi et al., 2011; Li et al., 2017). These bulk methods can quantify chemical properties of aerosol species from coal combustion, but they cannot directly provide the characteristics of the individual particles. To our knowledge, morphologies and elemental compositions of individual particles from coal combustion are poorly characterized but are important in understanding their properties and their influences on atmospheric pollution. Therefore, this study plays an important role in clarifying the emission inventory for the source apportionment of ambient airborne particles.

Transmission electron microscopy coupled with energy-dispersive X-ray spectrometry (TEM-EDX) is a powerful tool for characterizing individual particles because it has advantages of high resolution and high magnification which provides some microstructural information of individual particles (Pósfai and Buseck, 2010). TEM has been widely used to measure individual particles in the atmosphere (Zhang et al., 2001; Li et al., 2003; Twohy et al., 2005; Okada et al., 2008; Li and Shao, 2009; Matsuki et al., 2010; Adachi and Buseck, 2011; Ueda et al., 2011; W.J. Li et al., 2013; Zhu et al., 2013; Duo et al., 2015). Morphologies and elemental compositions of coal burning-derived individual particles are critical for understanding the sources of atmospheric aerosol particles (Adachi et al., 2010; Freney et al., 2010; Niu et al., 2011).

The aims of this study are: (1) to classify the types of individual particles emitted from raw coal combustion in an experimental domestic stove based on the morphology and elemental composition revealed by TEM-EDX; (2) to study the source-specific properties of the individual particles from combustion of different coals.

UNCORRECTED PROOF

1. Sampling and experiment

1.1. Combustion system and sample collection

A laboratory-made combustion system was used to conduct the burning experiments, which were carried out at the Laboratory of the Chinese Academy of Environmental Sciences (Geng et al., 2012). The system was composed of a combustion stove with the smoke dilution tunnels and the smoke chambers. The coal-stove, which is widely used for cooking and heating in villages of China, was purchased from the grocery market. It has a metallic outer cover and thermal-insulated ceramic liner. The cylindrical inner volume is 0.01 m^3. The dilution tunnel consists of two main parts (an orthogonal pipe and a cylindrical tunnel both made of stainless steel) and an attached suction fan. The orthogonal pipe (length: 1.0 m, radius: 20 cm) was connected to the stove for flue gas introduction and first-step dilution with filtered air. At the end of the orthogonal pipe, a horizontal cylindrical tunnel (length: 4.0 m, radius: 40 cm) was connected for second-step dilution. At the end of the tunnel, there were several orifices for suction fans and sampling. During sampling process, all flue gases were introduced into dilution tunnel and mixed with filtered air. The flow rate of the suction fan was controlled by Venturic tube and fixed at 5800 L/min. To avoid particle losses, a dynamic dilutor was used to introduce the flue gas into the smog chambers. The principle of the dynamic dilutor is based on ejection dilution. Purified pressurized dilution air flows at high speed around an ejector nozzle and caused a pressure drop which draws a sample through the nozzle. The raw sample is instantaneously diluted as it mixes with the dilution airflow.

The residence time of flue gas in the dilution tunnel was 5.5 sec. After second-step dilution, the temperature of diluted flue gas was 30°C. During the experiments, the flow rate of the diluted flue gas into smog chamber was fixed at 100 L/min. The smoke chamber connected to the horizontal cylindrical tunnel

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