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Refined source apportionment of coal combustion sources by using single particle mass spectrometry

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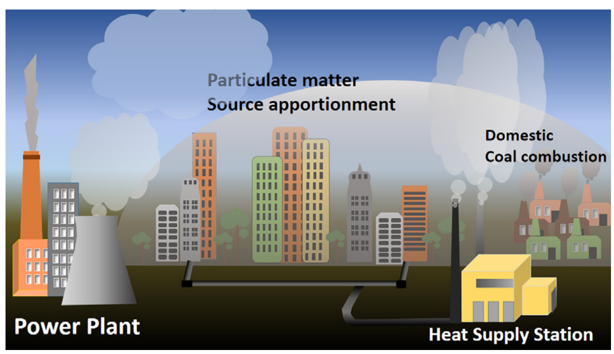
HIGHLIGHTS

- Size-resolved mass spectral features of three typical coal combustion source types were analyzed and compared.
- Signals of OC, nitrate, sulfate, Si, Al and Ca are tracers for these source types, and are effective in distinguishing them.
- Contributions of three source types were quantified in winter of North China with the mass spectral features obtained.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, samples of three typical coal combustion source types, including Domestic bulk coal combustion (DBCC), Heat supply station (HSS), and Power plant (PP) were sampled and large sets of their mass spectra were obtained and analyzed by SPAMS during winter in a megacity in China. A primary goal of this study involves determining representative size-resolved single particle mass spectral signatures of three source types that can be used in source apportionment activities. Chemical types describe the majority of the particles of each source type were extracted by ART-2a algorithm with distinct size characteristics, and the corresponding tracer signals were identified. Mass spectral signatures from three source types were different from each other, and the tracer signals were effective in distinguishing different source types. A high size-resolution source apportionment method were proposed in this study through matching sources' mass spectral signatures to particle spectra in a twelve days ambient sampling to source apportion the particles. Contributions of three source types got different size characteristics, as HSS source got higher contribution in smaller sizes, But PP source got higher contributions as size increased. Source contributions were also quantified during two typical haze episodes, and results indicated that HSS source (for central-heating) and DBCC source (for domestic heating and cooking) may contribute evidently to pollution formation.

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1. Introduction

High concentration of particulate matter is now the primary concern about air quality in China, which adversely affects climate and human health (Jimenez et al., 2009; Mimura et al., 2014; Keim et al., 2005). Coal accounts for 65.6% of all energy consumed (coal equivalent calculation) in China in 2014, and thermal power generation and heating supply consume the largest part of coal (Wang and Luo, 2017). Meanwhile, a large number of coal-fired industrial boilers are widely equipped. What's more, coal is widely used as a source of energy for cooking and heating in vast rural areas of China, and the highest levels of domestic coal consumption occur in northern China in winter for heating needs (Du et al., 2016; Bonjour et al., 2013). Recent years, a lot of studies were carried out to identify emission sources to particulate matter in China, coal combustion is considered as an important source of particulate matter (Sun et al., 2013). Most of the studies use receptor models (including PMF, CMB etc.), and only contributions from major sources can be evaluated. For example, they regard coal combustion in all sectors as a whole most of the times, very few studies can distinguish coal burning in one sector from another (Yuan et al., 2015; Li et al., 2015; Zhang et al., 2017; Peng et al., 2016). This is primarily due to the lack of comprehensive profiles of source types (Saarnio et al., 2014; Yan et al., 2016; Liu et al., 2016), as well as source-specific tracers. The coal types, combustion technologies and the end-of-pipe emission control policies are quite different in different sectors, which leads to different energy efficiency and subsequently different emissions (Ma et al., 2017; Du et al., 2016). Therefore, studying and establishing source profiles in different sectors is important for source apportionment study and policy making.

Haze episode has swept across China in recent years, the increase of haze has been observed in northern, eastern and southwestern China (Zhang et al., 2015; Gao et al., 2015; Yuan et al., 2015; Li et al., 2015). Particulate matter concentrations exhibit dramatic changes in a short term period in haze episodes (Xie et al., 2017; Lyu et al., 2016), which requires new technologies, which can characterize chemical and physical information of particulate matter quickly and make source apportionment in high time resolution (Zhang et al., 2017; Peng et al., 2016). The majority of source apportionment studies have traditionally used filter-based measurements (Lee et al., 2003; Waked et al., 2014; Lv et al., 2016), which usually requires a relatively long period of time (usually 24 h), and is not sensitive and fast-response enough to analysis the fast-changing episodes (Zhang et al., 2017). Single particle mass spectrometry, such as aerosol time-of-flight mass spectrometry (ATOFMS), single particle aerosol mass spectrometry (SPAMS), etc., can provide information on particle size and composition in real time. Till now, some studies have applied Single particle mass spectrometry to study the mass spectrum features of source emitted particles, including biomass burning, coal combustion, soil dust, and vehicle exhaust, etc. (Silva et al., 2000; Shields et al., 2007; Toner et al., 2006; Beddows et al., 2016; Dallosto et al., 2014; Silva et al., 1999; Xu et al., 2017). Besides, some studies use these features to apportion source contributions to ambient atmosphere particles (Toner et al., 2008; Dallosto et al., 2014; Healy et al., 2012; Dall'Osto et al., 2009; Zhang et al., 2009), which enrich source apportionment technical architecture, and provide more fast source apportionment results. However, studies on mass spectral fingerprints of different coal combustion source types were still scarce.

In this study, three typical coal combustion source types were sampled in a megacity during winter in China, including Domestic bulk coal combustion (DBCC), Heat supply station (HSS), and Power plant (PP). Large sets of single particle mass spectra of three source types were collected and analyzed by SPAMS, with vacuum aerodynamic diameter in the size range of 0.4–1.0 μm. Firstly, this study aims to explore the spectral features of these three coal combustion source types, and investigate their mass spectral feature size distribution patterns. Secondly, differences of mass spectra features between sources were analyzed, and the corresponding characteristic tracer signals were investigated. Thirdly, a new high-size resolution source apportionment method was proposed in this study by utilizing the source spectral features sufficiently, which can apportion ambient particles to these sources in different sizes. Finally, effects of coal combustion source emission on haze formation were assessed by investigating the source contributions during two haze episodes. To the best of our knowledge, this is the first study using SPAMS to study mass spectra features of three coal combustion source types, and apply them on source apportionment activity. Results of this study will be useful for exploring the pollution formation mechanism and policy control in China.

2. Method

Ambient particles were measured by SPAMS in an educational zone (38.988°N, 117.33°E) from 1st to 12th January of 2017 in Tianjin, China. The sampling site is located in a remote suburb area which is far from the city center. Only few vehicles ran on road near the monitor site. Lots of trees and many lakes are around the zone, and good afforest seldom lead to dust pollution. Besides, the educational zone situates near both countryside and residential districts (as shown in Fig. S1, Supplementary material), where DBCC and HSS was always used for heating or cooking in cold winter in north China. Therefore, all factors mentioned above make the zone a suitable site for studying coal combustion source's particle pollution effects.

2.1. Measurement site and instrumentation

This work reports the use of Single Particle Aerosol Mass Spectrometer (SPAMS) for the study of coal combustion particles. The SPAMS can give valuable insights into the size and composition of individual airborne particles (Spencer et al., 2007; Pratt and Prather, 2009; Ma, 2010; Toner et al., 2006), and been described in detail elsewhere (Li et al., 2011). The SPAMS developed by Guangzhou Hexin Analytical Company was applied to field measurement of singles particles, as well as source sampling activities. Briefly, particles are introduced into SPAMS through a vacuum pumped aerodynamic lens at a flow rate of 75 mL min^{-1}, then focused and accelerated to a terminal velocity and introduced to the sizing region. The velocity of each particle are determined by two continuous diode ND:YAG laser beams operated at 532 nm, and is used to calculate the single particle aerodynamic diameter. Finally, the sized particle reaches the laser desorption/ionization region, and the energy of desorption-ionization 266 nm laser was 0.6 mJ and the power density was kept at about 1.06×10^8W cm^{-2}. The laser generates positive and negative ions, those are analyzed by a bipolar time-of-flight mass spectrometer. The standard SPAMS commercial instrument can analyze particles ranging from ~200 to 2000 nm. Particles were sampled with a flow rate of about 80 mL/min. Polystyrene latex spheres (Duke Scientific Corp., Palo Alto) particles with aerodynamic diameters of 0.2, 0.3, 0.5, 0.72, 1.0, 1.3 and 2.0 μm were used for size calibration.

In this study, for sampling coal combustion particles samples from PP and HSS, vehicle loaded SPAMS were placed under the chimney in the factories. During the sampling, a stainless steel tube was inserted in the sampling hole on the chimney enough, to make sure the tube could reach inside of the chimney. Airflow together with particle samples were pump out (25 L·min^{-1}) through a closed silicone tube from chimney breast, and were then imported into the SPAMS. The sampling pump was used here to shorten the residence time of particle samples in the sampling tube. For sampling the real Domestic Bulk Coal Combustion particles in rural areas, particle samples were collected by a sampling bags (with the volume of 2 L), which were made by Teflon. These airbags were transferred to the laboratory timely within 4 h. SPAMS collects bipolar mass spectra of individual particle, as well as the size information. After the sampling experiment, lots of mass spectra of three source samples with vacuum aerodynamic diameter were