



Mass spectra features of biomass burning boiler and coal burning boiler emitted particles by single particle aerosol mass spectrometer



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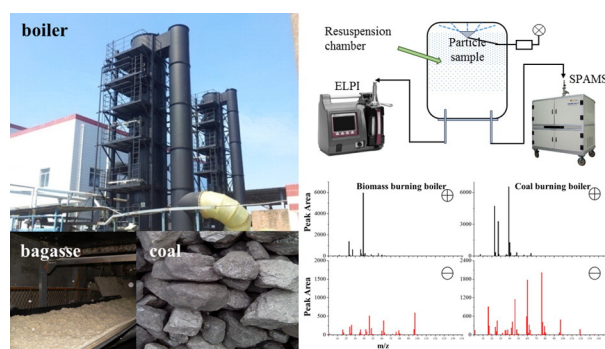
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HIGHLIGHTS

- Samples from Coal burning and Biomass burning boilers were sampled from a power plant and a sugar industry.
- Size-resolved single particle mass spectra signatures were analyzed by ART-2a.
- Part of the single particle mass spectra of samples were similar in the same size range, and can't be distinguished by ART-2a method.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 25 February 2017

Received in revised form 18 April 2017

Accepted 18 April 2017

Available online xxxx

Editor: D. Barcelo

Keywords:

Biomass burning boiler

Coal burning boiler

Single particle

Chemical composition

Size distribution

SPAMS

ABSTRACT

In this study, single particle mass spectra signatures of both coal burning boiler and biomass burning boiler emitted particles were studied. Particle samples were suspended in clean Resuspension Chamber, and analyzed by ELPI and SPAMS simultaneously. The size distribution of BBB (biomass burning boiler sample) and CBB (coal burning boiler sample) are different, as BBB peaks at smaller size, and CBB peaks at larger size. Mass spectra signatures of two samples were studied by analyzing the average mass spectrum of each particle cluster extracted by ART-2a in different size ranges. In conclusion, BBB sample mostly consists of OC and EC containing particles, and a small fraction of K-rich particles in the size range of 0.2–0.5 μm . In 0.5–1.0 μm , BBB sample consists of EC, OC, K-rich and Al_Silicate containing particles; CBB sample consists of EC, ECOC containing particles, while Al_Silicate (including Al_Ca_Ti_Silicate, Al_Ti_Silicate, Al_Silicate) containing particles got higher fractions as size increase. The similarity of single particle mass spectrum signatures between two samples were studied by analyzing the dot product, results indicated that part of the single particle mass spectra of two samples in the same size range are similar, which bring challenge to the future source apportionment activity by using single particle aerosol mass spectrometer. Results of this study will provide physicochemical information of important sources which contribute to particle pollution, and will support source apportionment activities.

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1. Introduction

As the world's second-largest economy, China has experienced severe haze pollution in recent years, with fine particulate matter reaching unprecedented levels in many large cities, such as the Jing-

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jin-ji Area (Zhang et al., 2015; Gao et al., 2015; Yuan et al., 2015; Wang et al., 2012; J. Wang et al., 2014), PRD (Wang et al., 2014a), Yangtze River Delta (Wang et al., 2014b), and Xi'an (Li et al., 2015). Particulate matter pollution has aroused worldwide concern during the past decade due to its significant adverse effects on visibility, public health and even global climate (Jimenez et al., 2009; Mimura et al., 2014; Keim et al., 2005). Therefore, investigating the emission sources of particulate matter is of great importance (Manousakas et al., 2015; Liu et al., 2015; Parworth et al., 2015; Xu et al., 2016). Many source apportionment studies have been conducted around the world (Manousakas et al., 2015; Liu et al., 2015; Parworth et al., 2015; Xu et al., 2016), among which, studies on fast source apportionment methods are becoming desirable, as they are enable of providing the characterization of particle emission sources, especially in haze pollution events (Peng et al., 2016; Gao et al., 2016; Zheng et al., 2016).

Single particle mass spectrometry, such as aerosol time-of-flight mass spectrometry (ATOFMS) and single particle aerosol mass spectrometry (SPAMS), have been extensively used to measure the aerodynamic size and chemical composition of individual aerosol particles (Bi et al., 2008; Ma, 2010; Toner et al., 2006; Shields et al., 2008; Spencer et al., 2007; Pratt and Prather, 2009). Currently, source apportionment methods based on single particle mass spectrometer have been studied extensively (Healy et al., 2012; Dall'Osto et al., 2009; Zhang et al., 2009; Dall'Osto et al., 2014; Giorio et al., 2015; Toner et al., 2008). For example, some studies applied ART-2a neural network algorithm to pick out fine particles containing specific species, such as lead and carbon, and to determine their original sources by analyzing the size distribution, chemical composition and temporal variation of ambient aerosols (Healy et al., 2012; Dall'Osto et al., 2009; Zhang et al., 2009). In addition, some researchers used clustering techniques to describe the majority of particle types, and to quantify or semi-quantify the source contributions by comparing the mass spectrum of generated particles types with the mass spectrum of sources (Dall'Osto et al., 2014). Furthermore, some studies also applied receptor models, such as PMF, to deconvolve the different pollutant sources which contribute to ambient particulate matter (Giorio et al., 2015). The source contribution can be quantified by analyzing the mass spectrum of each extracted factor. In addition, some studies use ART-2a neural network clustering algorithm matching method to match ambient particles with mass spectral fingerprints of sources (Toner et al., 2008). All the studies mentioned above were conducted based on background information of mass spectral fingerprints of different sources. Thus, study of individual particles emitted by various sources can potentially provide useful information for real time observation of particles in the atmosphere (Silva et al., 1999).

Some studies have been conducted to study the mass spectral signatures of particulate matter emission sources, such as soil dust (Silva et al., 2000), vehicle exhaust (Shields et al., 2007; Toner et al., 2006), brake wear (Beddows et al., 2016), tire dust (Dall'Osto et al., 2014), and biomass burning (Silva et al., 1999). Furthermore, many studies have also reported that single particle mass spectral signatures are complex. Shields reported spectral signatures from heavy-duty diesel vehicle emissions containing seven chemical classes (Shields et al., 2007). Toner also studied the single particle characterization of ultrafine and accumulation mode particles from heavy duty diesel vehicles using ATOFMS, and seven classes were extracted to illustrate the major signatures (Toner et al., 2006). Silva studied the single particle spectra of suspended soil dust from southern California, and four typical types were observed from the soil particle samples, and the chemical composition of soil dust showed even more variability (Silva et al., 2000). Therefore, more information on single particle mass spectral signatures is needed all over the world to characterize the mixing state, chemical composition of each emission sources, and which will also support source apportionment analysis.

In addition, biomass burning boiler was developed for high efficiency and clean combustion, which have been applied in many places all over the world, as well as in China. Biomass burning boiler has bright

and broad prospects in the future. But, its emission characteristic is not yet clear, thus the harm of its emission is unpredictable. Thus in this study, single particle spectral features of emitted particles by biomass burning boiler (BBB) and another worldwide used technology, coal combustion boiler (CBB) (Biswas et al., 2011; Sun et al., 2013), are studied to investigate the difference between them. As to the authors' knowledge, few studies have been conducted on BBB and CBB samples based on single particle mass spectrometry. Besides, most of the studies were performed in laboratory burning simulations (Wang et al., 2013; Silva et al., 1999; Dall'Osto et al., 2014), and studies on industry boiler emitted particles are very few.

The purpose of this paper is to obtain quantitative signatures for biomass burning boiler and coal burning boiler emitted particles. This analysis was performed on a single particle basis, measuring the size and chemical composition of each particle. The physical-chemical characteristics of the two boiler burning particles were investigated. A clustering method, Neutral network algorithm, ART-2a was applied to study the complex characteristics of biomass burning boiler particles and coal combustion boiler particles. An enrichment factor (EF) was proposed in this study to analyze the differences of specific mass-to-charge (m/z) intensities between source samples and ambient particles in the atmosphere. This study will provide both physical and chemical information of these two sources, and will help support further source apportionment studies based on single particle mass spectrometry.

2. Method

2.1. Sampling

Particle samples were collected from a sugar factory which uses a biomass burning boiler and from a power plant which uses a coal burning boiler in Guangxi Province, China. Guangxi Province is famous for sugar industry and has an abundance of biomass resources, and biomass burning boilers are promoted locally. The biomass burning boiler in this study burns bagasse from sugar cane to generate electricity. To consider the feasibility, samples were collected from bottom of electrostatic precipitators with a plastic brush (Kong et al., 2015).

2.2. Sample analysis

The experimental system consisted of a Resuspension Chamber, a single particle aerosol mass spectrometer (SPAMS, Hexin Analytical Instrument Co., Ltd., China), and an Electrical Low Pressure Impactor (ELPI) (Dekati Ltd., Tampere, Finland). All samples were in the form of a dry powder (before the analysis, samples were spread on clean paper in dry conditions at a room temperature of approximately 25 °C for over 24 h; the samples were then sieved). Before analysis, the resuspension machine was pre-cleaned and clean air was pumped into the machine continuously.

The Resuspension Chamber has been applied and described elsewhere (Chow et al., 1994; Kong et al., 2015). Briefly, a pre-dried and sieved sample is evenly spread out through a pump from the top of the chamber, which can simulate the process of the sample entering into the ambient environment. This method makes it possible to analyze the real size distribution and chemical components as they would be in the ambient environment.

The principle and design of the SPAMS instrument has been described in the literature (Li et al., 2011). Briefly, aerosols are introduced into the vacuum pumped SPAMS through a 0.1 mm critical orifice at a flow rate of 80 mL min⁻¹ due to the pressure drop from ~760 to ~2.2 Torr. They are then focused and accelerated to specific velocities characteristic of their aerodynamic diameter while passing through the aerodynamic lens, followed by the sizing region, where velocities of individual particles are determined by two continuous diode ND:YAG laser beams which operate at 532 nm and are separated by 6 cm. Desorption/ionization is applied subsequently by a 266 nm: YAG

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