Size-resolved chemical composition of atmospheric particles during a straw burning period at Mt. Huang (the Yellow Mountain) of China

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Highlights
- Size-resolved chemical composition of aerosol particles at a high mountain in southeastern China.
- K-Secondary and K-EC particles were the dominant particle types.
- Biomass burning has a significant impact on the chemical properties of aerosols.
- Long-range transport of pollutants from industrial emissions has a large responsibility to the K-EC rich particles.

Abstract
The particle size spectra and chemical composition of aerosol particles at Mt. Huang (the Yellow Mountain), a background site of southeastern China, were investigated using a single particle aerosol mass spectrometer (SPAMS) and other aerosol monitoring instruments. The field campaign was conducted from Sep. 29 to Oct. 9, 2012, to observe the influence of straw burning on the size distribution, chemical composition of atmospheric aerosols at a background site. Results showed that K-Secondary and K-EC particles were the dominant particle types during this period, with their number concentrations totally accounting for 74% of all the particles. From long-range transport analysis of air masses, six types of particles all contained high concentrations of 39[K]+ ion (known as the tracer for biomass burning) which indicate that biomass burning may represent as a significant source of aerosols for air masses originated from the north of Mt. Huang. The sampling period could be classified into three sub-periods according to the backward trajectories. During sub-period 1, the K-Secondary particles exhibited the highest concentrations, accounting for 74.2% and 55.4%, respectively, of the submicron and supermicron particles. In sub-period 3, K-EC dominated the submicron particles, indicating that more particles had anthropogenic sources, especially industrial emissions. The results obtained in this study will enrich the database of aerosol chemical composition in the background sites of southeast China and could be of important applications in environmental and climate research.

1. Introduction
Atmospheric particulate matter (PM) has a significant impact on human health, atmospheric visibility, and climate (Gard et al., 1998), but there are still big gaps in understanding their influence on the hydrological cycle, and their direct and indirect radiative forcing on climate (e.g., Knutti et al., 2002; Chen and Penner, 2005; Lohmann and Feichter, 2005; Moffet and Prather, 2005; Ramanathan et al., 2005; Dall’Osto and Harrison, 2006; Spencer et al., 2008). A key uncertainty is the lack of reliable information on the chemical composition and mixing state of aerosol particles (Jacobson, 2001; Chandra et al., 2004; Myhre et al., 2004; Schwartz, 2004; Lohmann and Feichter, 2005) which are essential for predicting the impact of aerosols on climate. Therefore, it is necessary to understand the dynamic nature, chemical composition and mixing state of aerosol particles in order to quantify their effects on...
climate and the environment (Gard et al., 1998; Babu and Moorhdy, 2001; Spencer et al., 2008).

Over the past three decades, rapid economic development and population growth has led to a general decline in environmental quality and widespread air pollution in southern and southeastern China (Hsu et al., 2010; Guo et al., 2010a,b; Ding et al., 2011; Han et al., 2011). The physical and chemical characteristics of atmospheric particles, especially the chemical composition and mixing state, became more complex in the region (Wu et al., 2006; Yi et al., 2007; Cheng et al., 2008; Guo et al., 2010a; Park et al., 2012).

Single particle mass spectrometer, such as aerosol time-of-flight mass spectrometer (ATOFMS) has emerged as a powerful tool for on-line characterization of aerodynamic size and chemical composition of particles (Gard et al., 1997, 1998; Guazzotti et al., 2001; Liu et al., 2003; Toner et al., 2006; Shields et al., 2008; Spencer et al., 2008; Pratt and Prather, 2009). Gard et al. (1997) described a portable ATOFMS for determining the size and chemical composition of ambient aerosols. Gard et al. (1998) observed the heterogeneous replacement of chloride by nitrate in individual sea-salt particles. These studies provide useful information on the mixing state and formation mechanisms of individual particles (Angelino et al., 2001; Moffet et al., 2004), essential for understanding the impact of aerosols on environment and climate (Freney et al., 2006; Denkenberger et al., 2007; Wang et al., 2009; Yang et al., 2009; Gaston et al., 2010).

In recent years, a number of studies have been carried out on characterization of chemical and physical properties of aerosol particles in urban areas of China using single-particle mass spectrometry (SPAMS) (e.g., Bi et al., 2011; Li et al., 2011, 2012; Tao et al., 2011; Zhang et al., 2012), but little attention has been paid on single aerosol particles in background regions of China, especially at remote mountainous regions where the chemical composition and mixing state of particles are influenced frequently by long-range transport, thus may reflect the atmospheric characteristics on a large-scale domain (Decesari et al., 2005; Wang et al., 2012).

In China, field burning of crop residues after harvest (June, July and October) is a common practice (Gao et al., 2011; Qu et al., 2012; Yang et al., 2006). Cao et al. (2008a) investigated the emission factors of particulate matter, element carbon, organic carbon, SO2, NOx, CO, CO2, and ten ions (Na+, NH4+, K+, Mg2+, Ca2+, F-, Cl-, NO2-, NO3-, SO42-) from the domestic burning of four types of commonly produced crop residues in rural China: rice straw, wheat straw, corn stover, and cotton stalk and found that K+ and Cl-, had the highest emission factors from all the crops. Meanwhile, they occupied by 3.39–16.23% and 7.0–16.02% of the particles. A number of studies indicated that this activity could significantly affect local and regional air quality, human health and even the climate (Yang et al., 2006; Li et al., 2007; Wang et al., 2007; Cao et al., 2008b; Zhang et al., 2008). Bi et al. (2011) studied the chemical composition and mixing state of biomass burning particles by SPAMS in Pearl River Delta region and indicated that biomass burning contributed to the formation of nitrate particles during transport. Yang et al. (2009) studied the impact of biomass burning particles on air quality in Shanghai. Significant influence of biomass burning emissions was observed with contributions of biomass smoke to OC in PM10 to be as high as 32% during a long-range transport episode at Guangzhou in July (Zhang et al., 2010). Cheng et al. (2009) also indicated that fine aerosol ion species including Cl-, K+ and NO3- increased 11.0, 6.7 and 5.5 times during agricultural burning periods compared with periods when agricultural waste burning was not performed and K+ was found mainly in the fine mode during agricultural burning period (June and November) in Taichung. However, few researches focused on the particle’s compositions in Chinese background site. The aerosol characteristics at background site can represent composition features of regional atmospheric particles and help clearly understand the interaction of the aerosol and cloud. Therefore, Mt. Huang (the background site for southeastern China) was selected in this study. An intensive measurement by SPAMS (Bi et al., 2011; Li et al., 2012; Zhang et al., 2012) and aerosol spectrometers was conducted during a straw burning period of 2012.

This work is one portion of a project aimed to investigate the changes in physical and chemical properties of atmospheric particles and cloud droplets and their interactions in southern mountainous area of China funded by China National Natural Science Foundation. The objectives of this work were: (1) to characterize the single particle size (0.2–2.5 μm) and real-time chemical compositions at a straw burning period; (2) to identify the source locations by back trajectories for aerosols at this background mountainous site; (3) to obtain the differences of chemical compositions for air masses transported from different directions.

2. Setup of the experiments

2.1. Site description and sampling period

Observations were conducted on the top of a building at the crest of Mt. Huang (30.12°N, 118.19°E, with altitude of about 900 m above mean sea level) as shown in Fig. 1. Outside the building valley which extends until the Bright peak (30.13°N, 118.15°E, 1840 m a. m. s. l.) Another observation site was located on the Bright peak. There were no obvious anthropogenic pollution sources for aerosols at the surrounding regions of the observational sites. Measurements of aerosol chemical composition were carried out from September 29 to October 9, 2012 using a SPAMS (Hexin Analytical Instrument Co., Ltd, China). A brief description of the measurements and the other instrument used during this campaign is as follows.

2.2. Sampling by SPAMS

Aerosols were collected through a stainless steel pump at a height of approximately 8 m above the ground level. The sampling mast entered into a temperature controlled room, which was kept at ambient temperature and relative humidity (RH) for collecting particles to prevent condensation or evaporation during particle measurements. A suite of instruments for monitoring the physical and chemical properties of particles were also set in the same room beside the SPAMS, including aerodynamic particle sizer (APS, TSI model 3321), mobility particle sizer (TSI model 3936), aerosol spectrometers was conducted during a straw burning period of 2012. Aerosols were collected through a stainless steel pump at a height of approximately 8 m above the ground level. The sampling mast entered into a temperature controlled room, which was kept at ambient temperature and relative humidity (RH) for collecting particles to prevent condensation or evaporation during particle measurements. A suite of instruments for monitoring the physical and chemical properties of particles were also set in the same room beside the SPAMS, including aerodynamic particle sizer (APS, TSI model 3321), mobility particle sizer (TSI model 3936), aethalometer (Magee Scientific) and nephelometer (TSI integrating nephelometer model 3563).

The design and performance of SPAMS have been reported by others (e.g., Bi et al., 2011; Li et al., 2011, 2012; Zhang et al., 2012). Briefly, ambient aerosol particles were introduced into the SPAMS by a vacuum-pumpthrough a 0.1 mm critical orifice at a flow of 80 mL min⁻¹ (the pressure drop from ~760 to ~2.2 Torr). Along with the pressure dropped from ~760 to ~2.2 Torr, particles achieved a terminal velocity in the supersonic expansion air flow. Particles were detected and aerodynamically sized by two continuous diode Nd: YAG laser beams (532 nm). They were then disrupted and ionized by a pulsed Nd: YAG laser (266 nm) triggered based on the velocity of a specific particle. The positive and negative ions which were produced and reached the detectors at different time were detected according to the different mass to charge ratios (m/z). The laser affecting the desorption/ionization was kept as low as 106 × 10⁸ W cm⁻². The instrument was routinely calibrated with polystyrene latex spheres of 0.2–2.5 μm in diameter (Nanosphere Size Standards, Duke Scientific Corp., Palo Alto).
3. Results and discussions

3.1. Number concentration of particles

A total of 692,362 particles were sized and chemically analyzed with TSI MS-Analyze software. The peak lists of particles sizes and mass spectral information were subsequently introduced into MATLAB 7.1 (Mathworks Inc.) and analyzed by YAADA 2.1 software (www.yaada.org) which was used for processing the single-particle mass spectral data with a MATLAB-based software toolkit. Peak identification described in this study was consistent with the most probable assignments for each specific value of mass-to-charge ratio (m/z). More details can be found in Murphy and Thomson (1997a; 1997b) and Liu et al. (2003). Single particle mass spectra were grouped by Adaptive Resonance Theory-based Neural Network, ART-2a (Hopke and Song, 1997; Song et al., 1999) with the vigilance factor as 0.7, learning rate as 0.05 and iterations as 20 times in this study. The grouping yielded 687 clusters by ART-2a analysis for about 692,000 unique particle mass spectra. The top 300 clusters represented over 90.8% of the mass spectra and were

Fig. 1. Location of the field campaign (the Huangshan Mountain is indicated with a black triangle, 30.12°N, 118.19°E).

Fig. 2. Comparison of the number concentrations obtained from APS and number count of SPAMS for particles within 0.5–1.0 μm (the top panel) and 1.0–2.5 μm (the bottom panel) collected from Mt. Huangshan at Sep. 29-Oct. 9, 2012.
used for the results presented here. Further manual classification was used to refine the aerosols into six chemical classes.

Fig. 2 compared the number concentrations of aerosol particles in the range of 0.5–2.5 μm in diameter, obtained from the SPAMS and the APS. The data are presented with a resolution of one hour. It can be seen from Fig. 2 that the SPAMS particle counts correlate well with the APS particle number concentrations during the whole observational period with the correlation coefficient ($R^2$) as 0.92 and 0.73 for particles within 0.5–1.0 μm and 1.0–2.5 μm, respectively.

From Oct. 5 to 8, 2012, both the SPAMS and APS detected a rapid increase in particle number concentrations. During this period, the air mass gradually drifted from the agricultural areas in North China Plain (NCP) to the industrial region, which is one of China’s most important economic and industrial zone, the Yangtze River Delta Region (as the back trajectory analysis shown in Section 3.3).

Table 1

<table>
<thead>
<tr>
<th>Type</th>
<th>K–Ca-rich</th>
<th>K-EC</th>
<th>K-ECOC</th>
<th>K-OC</th>
<th>K-secondary</th>
<th>Na–K-rich</th>
<th>Others</th>
</tr>
</thead>
<tbody>
<tr>
<td>Submicron</td>
<td>19,471</td>
<td>169,819</td>
<td>37,294</td>
<td>16,867</td>
<td>292,459</td>
<td>24,315</td>
<td>44,776</td>
</tr>
<tr>
<td>Super-micron</td>
<td>6395</td>
<td>11,820</td>
<td>3481</td>
<td>2581</td>
<td>37,666</td>
<td>6721</td>
<td>18,696</td>
</tr>
</tbody>
</table>

Fig. 3. Positive (+) and negative (−) ART-2a area vectors attributed to (a) K-Secondary; (b) K–Ca-rich; (c) K-EC; (d) K-ECOC; (e) K-OC and (f) Na–K-rich observed during the experimental period.
confirmed that the higher number concentration of particles were caused by both agricultural and industrial activities in these regions followed by long-range transport of air masses.

3.2. Major particle types from mass spectral analysis

3.2.1. Mass spectral classification

Totally 2,494,803 particles were collected, with 822,473 particles sized and recorded (with average hit rate as 28%). The hit fractions fluctuated in the ranges of 23%–51% and 15%–48% for submicron particles (0.2–1.0 μm in diameter) super-micron particles (1.0–2.5 μm in diameter), respectively. Both positive and negative ion spectra were analyzed for 692,362 particles. The hit rate was 28%.

Five major particle types were obtained as potassium with secondary organics (K-Secondary), potassium with calcium (K-Ca-rich), potassium with elemental carbon (K-EC), potassium with organic and elemental carbon (K-ECOC), potassium with organic carbon (K-OC) and sodium with potassium rich (Na–K-rich). Counts of particles in the six types of classified and unclassified particles (named as “others”) are shown in Table 1.

Area matrix for the above mentioned six particle types were shown in Fig. 3. During the observation period, 39 [K+] was always recognized as a good marker for biomass burning (Andreae, 1983; Bi et al., 2011), appeared in all the six types of particles. K-Secondary was always composed with 39 [K+], 97 [HSO4], nitrate ions (−46 [NO3]− and −62 [NO3]−) and sometimes with OC ions such as −26 [CN]− and −42 [CNO]− (Bi et al., 2011). K-Ca-rich was characterized by ion peaks of 39 [K+], 56 [CaO]+, and 97 [HSO4]. In addition to the major peaks, K-Ca-rich particles also contained abundant ions such as 23 [Na]+, −46 [NO3]−, and −62 [NO3]−, and phosphate (m/z −79 [PO4]3−). K-EC particles contained dual polarity carbon cluster ions, such as 12 [C]++, 24 [C2]++, ..., [C6]++ and 39 [K+], with an intense peak of sulfate −97 [HSO4]. While a few low intensity OC ions were detected, K-OC exhibited a clear signature of 39 [K+] internally mixed with OC ions and EC clusters. K-OC particles showed the least amounts, accounting for 2.81% of the total particles. It presented a strong peak at 39 [K+], and the peak values were likely related to the oxidized carbon compounds with m/z as 27 and 43. They were corresponding to [C2H3]++ and 39 [K+], sodium (m/z 23 [Na]+), calcium (m/z 56 [CaO]+), organics (like m/z 37 [C2H]+ and m/z 43 [C2H2O]+) and carbon cluster ions (such as m/z 12 [C]++, 24 [C2]++, ..., [C6]++). Six major particle types were obtained as potassium with secondary organics (K-Secondary), potassium with calcium (K-Ca-rich), potassium with elemental carbon (K-EC), potassium with organic and elemental carbon (K-ECOC), potassium with organic carbon (K-OC) and sodium with potassium rich (Na–K-rich). Counts of particles in the six types of classified and unclassified particles (named as “others”) are shown in Table 1.

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The sulfate ion as $m/z$ –97 [HSO₄]⁻ and potassium ion as $m/z$ +39 [K]⁺ were mixed with six types of particles as discussed above, which were similar to the results by using SPAMS (Bi et al., 2011). This intense signal of sulfate ion suggested that these particles had undergone a significant secondary processing during transport (Clarke et al., 2004). Potassium was the most abundant ion in the biomass burning particles (Spencer et al., 2008; Bi et al., 2011). Meanwhile, the fire points of field burning of crop straw were captured by satellite in the observation period (as Fig. 4). These results implied that particles were sourced from long-range transport of straw combustion-derived aerosols.

### 3.2.2. Size-resolved chemical composition of each particle type

Fig. 5 presents the percentages of the particle types as a function of aerodynamic diameter of aerosol particles (in 200 nm bins). The observed particles were classified into accumulation mode (from 0.2 to 2.0 μm) and coarse mode (from 2.0 to 2.5 μm) in this study. K-
ECOC and K-Secondary were the major two types for particles in 200–600 nm. With the particle size increasing, their contributions decreased. These particles were considered as effective cloud condensation nuclei (CCN) (Sullivan and Prather, 2005; Spencer et al., 2008; Bi et al., 2011). The mass spectra of the six particle types along with size distributions were suggestive for explaining the primary origin of these particles in accumulation mode. Our results also indicated that EC and sulfate mixed as internal form and kept for a long time when they were transported from the sources regions.

It could be seen from Fig. 5 that the proportion of Na–K-rich type of particles distributed in the accumulation mode. These particles may originate from industrial combustion and sea salt particles (Lin and Pan, 1998). However, the larger sea salt particles were scavenged and/or deposited in the transport process. Smaller sea-salt aerosols mixed with other compounds during long-range transport. For example, \( \text{SO}_4^{2-} \) was responsible for \( \text{Cl}^- \) depletion, and potassium particles were internally mixed with sea-salt aerosols (Harris et al., 2012; Kim and Park, 2012; Yao and Zhang, 2012). In larger particles (>2 \( \mu \)m in diameter), K–Ca-rich cluster dominated. There were also intensive sulfate ion signals implying that these particles have undergone secondary processing during transport. The particles were mixed internally as they transport from the source regions which was similar to that reported by Clarke et al. (2004).

In addition, particles of the six types interacted with each other by Pearson correlation analysis Table 2). There were significant correlations in the un-scaled SPAMS counts of certain particle types in one-hour resolution, except for K-OC and K–Ca-rich particles. The six types of particles could be divided into two groups: Group A includes K-Secondary, K-OC and Na–K-rich; Group B includes K–Ca-rich, K-EC and K-ECOC. It can be seen from Table 2 that negative correlation between group A and group B could be found while particles in both group A or group B exhibited positive correlations. The good positive correlation between these particle types in group A or group B suggested that they might have the similar sources or influenced by similar processes during transport. While the negative correlation between group A and group B indicated the particles types between them hold different sources or chemical processes.

3.3. Chemical composition of aerosols from different origins

Three-day air mass backward trajectories were employed to split the measurement campaign into three periods by using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model of NOAA (Fig. 6). Backward trajectories at 100 m, 500 m and 1000 m above ground level were adopted and three distinct periods were classified. Fig. 7 shows the time evolution of the chemical composition in particles during the observation period. Period 1 (from 05:00 Coordinated Universal Time UTC, 29 September to 8:00 UTC, 2 October) was influenced by continental air masses originated in the Siberia (long-range transport, with the ground level atmosphere originated from Inner Mongolia). Period 2 (from 9:00 UTC 2 October to 14:00 UTC 6 October) was influenced by a continental air mass which was sensitive to the emissions in the surroundings of Mt. Huang. Period 3 (from 15:00 UTC 6 October to 05:00 UTC 9 October) was influenced by both long-range transport continental and marine air masses.

Fig. 7a shows the time evolution of different chemical components for submicron particles. There were significant differences in the chemical composition of the submicron particles among the three distinct periods. For period 1, the site was impacted mainly by K-Secondary particles. This types of particles exhibited similar temporal trends indicating common or similar sources and/or they were subjected to similar processes. In period 1, there were abundant strong straw burning points in the NCP and the north of Mt. Huang (see Fig. 4a and b). When air mass passed the heavily populated areas influenced by straw burning in NCP, the contributions of K-Secondary particles increased substantially. Fig. 7b indicates that there were also similar distinct three periods for super-micron particles. It should be noted that about 9% of the particles analyzed by SPAMS were unclassified. They were composed of some non-calibrated particles and small numbers of particles with unique mass spectra. In particular, the K-Secondary was observed in 74.2% of the submicron particles during period 1 (Fig. 8a). K-Secondary particles have been attributed to biomass burning previously (Silva et al., 1999; Dall’Osto and Harrison, 2006; Bi et al., 2011). Previous studies showed that straw burning could produce potassium and chloride and that chlorine could convert into gas phase, while potassium kept in particular phase (e.g., Liu et al., 2000; Li et al., 2003). It was also indicated that the smoke particles could be processed very quickly if nitrate and/or sulfate were present (Li et al., 2003).

From period 1 to period 3, the percentages of the particle types including K-Secondary and K-OC decreased, while the contributions of K-EC, K-ECOC and K–Ca-rich increased for submicron particle, indicating that straw burning clearly impacted the major components of submicron particle during period 1. In periods 2 and 3, there were still straw burning events but its influence was
weaker. When air mass came from the surrounding or east of Mt. Huang (period 2), K-Secondary and K-EC dominated (accounting for 74.9%). In period 3, the percentages of K-EC and K-ECOC particles increased (Fig. 8c). For air mass originating from the industrial regions such as Bohai Bay surroundings and Yangtze River Delta Zone, an increase in particle types reflects the influence of industrial sources. Several researchers had also reported that EC and sulfate were from industrial sources (Moffet et al., 2008; Spencer et al., 2008; Decesari et al., 2011; Healy et al., 2012). EC and sulfate were major compounds in K-EC and K-ECOC particles, especially for K-EC component in submicron particles. In period 3, K-EC exhibited the highest mass percentages of the three periods, accounting for 40% of all the particles. A number of studies indicated that these particles (including EC and sulfate) may directly originate from biomass burning (Pratt and Prather, 2009; Li et al., 2010). Meanwhile, the straw burning was strictly forbidden at Mt. Huang. These particles may be related with the long-range transported primary emissions from human sources and secondary transform.

For super-micron particles (1.0–2.5 μm), the percentages of K-EC particles increased from 1.6% to 24.7% (Fig. 8d, e and f). The proportion of K-ECOC also increased with the change of the air mass transport routes, from 3.3% to 7.9%, which indicated that industrial sources may affect the local aerosols.

K-Ca-rich particles in super-micron size were few in the three periods, only accounting for 8.7–11.5%, higher than those in submicron size (2.8–4.6%). It indicated that they may be influenced by continental dust. However, the K-Ca-rich type particles were quite different from the naturally generated calcium-rich particles (Dall’Osto et al., 2010), as they were internally mixed with potassium and sulfate. It was important to note that atmospheric chemical compositions of Mt. Huang were similar to those in urban areas (Bi et al., 2011). It can also be concluded that particles at Mt. Huang showed a high fraction of K-containing particles in the sampling period indicating the influence of straw burning.

4. Summary and conclusions

In this study, SPAMS particle counts exhibited good agreement with those of APS data at Mt. Huang during the straw burning period, with the correlation coefficients (R²) as 0.92 and 0.73 for particles within 0.5–1.0 μm and 1.0–2.5 μm, respectively. A large proportion of particles could be classified into six types and they
were mostly concentrated in the accumulation mode with K–Ca- 
rich particles excluded. The relative mass fractions, total number 
concentrations and chemical compositions of particles changed 
with the directions and origins of the arriving air masses. For air 
masses passing across NCP, particles formed by straw burning rep-
resented a significant fraction of super- and submicron particles. 
The maximum proportion was K-Secondary for both submicron 
and super-micron size of particles, accounting for 74.2% and 24.7%, 
respectively. When the air mass shifted from northern to easterly 
direction, the fractions of K-Secondary particles decreased while 
the fraction of K-EC increased indicating the influence of industrial 
sources. While when the air mass came from Bohai Bay and across 
the industrial regions of Yangtze River Delta, the number concen-
trations for total and K-EC particles increased with the percentages 
of submicron and super-micron K-EC particles as 40% and 24.7%, 
respectively. Further studies will calculate the aerosol forcing in 
this region according to the size-resolved single particle chemical 
compositions and mixing state of aerosols.

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