

## Supplementary materials

**Table S1** Number fractions of the fresh SB particles containing specified ions

<i>m/z</i>	area	number fraction	
		rice-SB particles	corn-SB particle
7 (Li <sup>+</sup> )	> 50	0.08%	0.26%
23 (Na <sup>+</sup> )	> 50	43.7%	23.4%
39 (K <sup>+</sup> )	> 1000	90.0%	93.8%
56 (CaO <sup>+</sup> /Fe <sup>+</sup> )	> 50	4.1%	4.4%
113 (K <sub>2</sub> Cl <sup>+</sup> )	> 50	56.5%	65.5%
213 (K <sub>3</sub> SO <sub>4</sub> <sup>+</sup> )	> 50	71.1%	73.9%
-26 (CN <sup>-</sup> )	> 50	94.1%	97.1%
-35 (Cl <sup>-</sup> )	> 50	82.8%	94.6%
-42 (CNO <sup>-</sup> )	> 50	86.1%	95.9%
-46 (NO <sub>2</sub> <sup>-</sup> )	> 50	97.3%	98.7%
-62 (NO <sub>3</sub> <sup>-</sup> )	> 50	88.9%	98.7%
-97 (HSO <sub>4</sub> <sup>-</sup> )	> 50	96.1%	99.0%
27(C <sub>2</sub> H <sub>3</sub> <sup>+</sup> )	> 50	25.7%	38.3%
43(C <sub>2</sub> H <sub>3</sub> O <sup>+</sup> )	> 50	69.4%	75.7%
51(C <sub>4</sub> H <sub>3</sub> <sup>+</sup> )	> 50	38.6%	56.6%
63(C <sub>5</sub> H <sub>3</sub> <sup>+</sup> )	> 50	45.9%	61.9%
-25 (C <sub>2</sub> H <sup>-</sup> )	> 50	56.1%	87.6%
-45 (CHO <sub>2</sub> <sup>-</sup> )	> 50	23.9%	82.5%
-49 (C <sub>4</sub> H <sup>-</sup> )	> 50	52.2%	85.3%
-59 (C <sub>2</sub> H <sub>3</sub> O <sub>2</sub> <sup>-</sup> )	> 50	34.4%	88.5%
-71 (C <sub>3</sub> H <sub>3</sub> O <sub>2</sub> <sup>-</sup> )	> 50	13.8%	63.8%
-73 (C <sub>2</sub> H <sub>5</sub> COO <sup>-</sup> /C <sub>6</sub> H <sup>-</sup> )	> 50	39.5%	84.0%
-89 (C <sub>2</sub> HO <sub>4</sub> <sup>-</sup> )	> 50	8.7%	50.4%
-154(C <sub>12</sub> H <sub>10</sub> <sup>-</sup> )	> 50	16.4%	25.4%

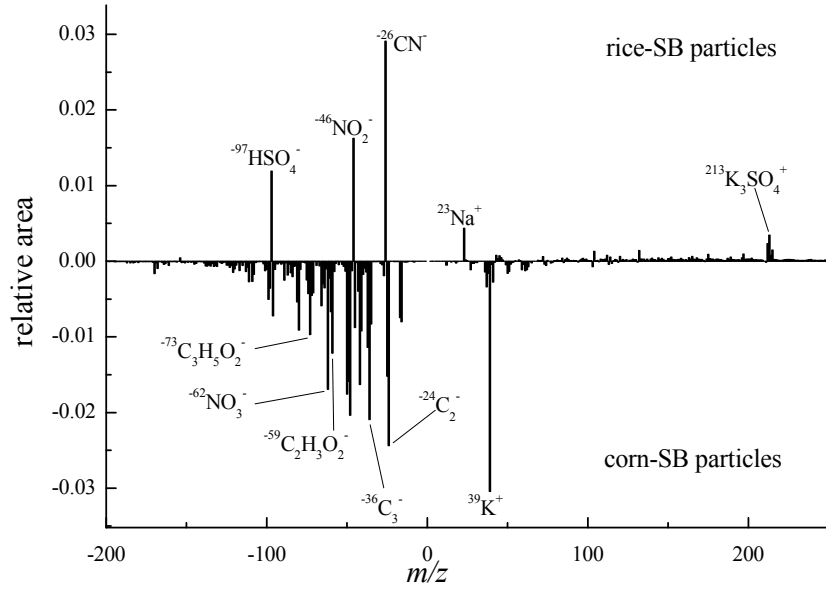
## Mass spectral analysis of the particles not included in the four major types

Biomass ashes, products of biomass burning, composed with silicate, oxides and hydroxides, sulphates, phosphates, carbonates, chlorides, nitrates and so on [1]. Based on high-temperature ash analyses, the oxide in crop straws ashes are  $\text{SiO}_2$ ,  $\text{K}_2\text{O}$ ,  $\text{CaO}$ ,  $\text{P}_2\text{O}_5$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{SO}_3$ ,  $\text{Na}_2\text{O}$ ,  $\text{MnO}$ ,  $\text{TiO}_2$  [2]. As the major substances, silicate, oxides ( $\text{K}_2\text{O}$ ,  $\text{CaO}$ ), phosphates, sulphates, nitrates were detected in the particles emitted from crop straws combustions by ATOFMS, as shown in Fig. S3. Depending on their compositions, the particles were divided into two types (ash1 and ash2). Furthermore, the ash1 particles also contained lead, which has toxic health effects.

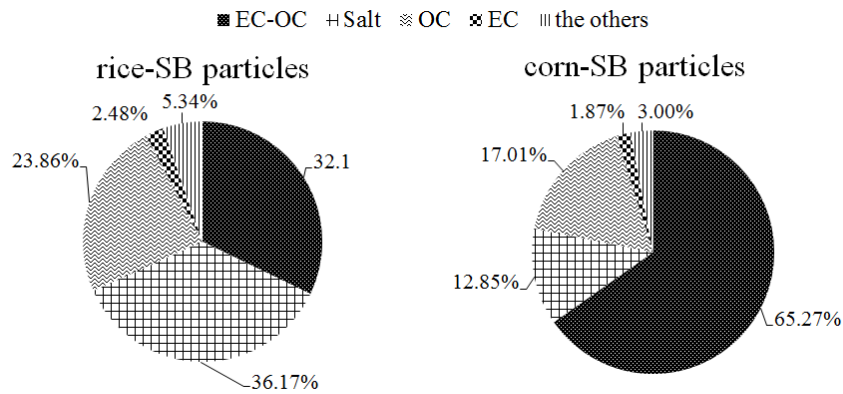
For the PAHs particles, the highest peak of PAHs was  $m/z$  252 $[\text{C}_{20}\text{H}_{12}^+]$  in positive mass spectra. In previous solid biofuel combustion study, the dominant PAH peaks were four-ring PAHs ( $m/z$  = 200–204, MW = 202) during start-up and five-ring PAHs ( $m/z$  = 250–255, MW = 252) during flaming effective combustion [3]. Because the crop straw combustion was flaming combustion, our result consisted with previous study. Besides, there were other PAHs, such as  $m/z$  168 $[\text{C}_{12}\text{H}_{24}^+]$  and 268 $[\text{C}_{21}\text{H}_{16}^+]$ . It was meaningful that the peak of  $m/z$ -154, -168, -170 were notably higher than other types.

As shown in Fig. S5, particles characterized with very high lead content ( $m/z$  206, 207, 208) were also detected by ATOFMS. Consistently, Silva et al. observed that 5% of the particles resulting from combustion of *Salsola tragus* contained lead [4]. In Zhang et al.'s research, they analyzed Pb-rich particles using ATOFMS and found that one category of the Pb-rich particles were emitted by biomass burning, but didn't figure out the kind of biomass [5]. According to our research, the combustions of rice and corn straws were two of the sources of Pb-rich particles, though the contribution was little. The presence of lead may attributed to particle deposition on the surface of the plant or an ability of the plant to absorb lead from the ground.

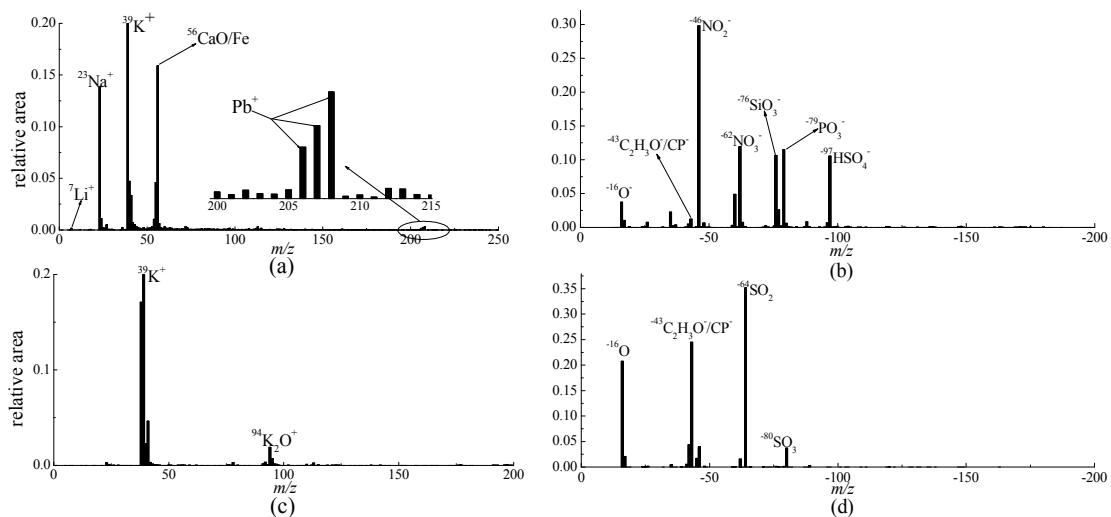
Nicotine, as marker for environmental tobacco smoke, was detected from crop straws combustions smokes using ATOFMS [6]. Crop straws combustions emitted some particles containing nicotine, as shown in Fig. S6.



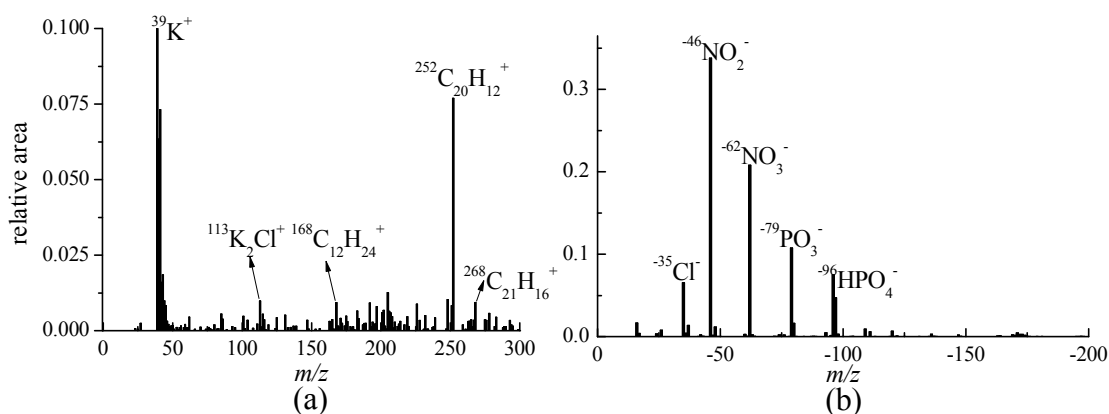
**Fig. S1** Subtraction mass spectra for rice-SB particles minus corn-SB particles



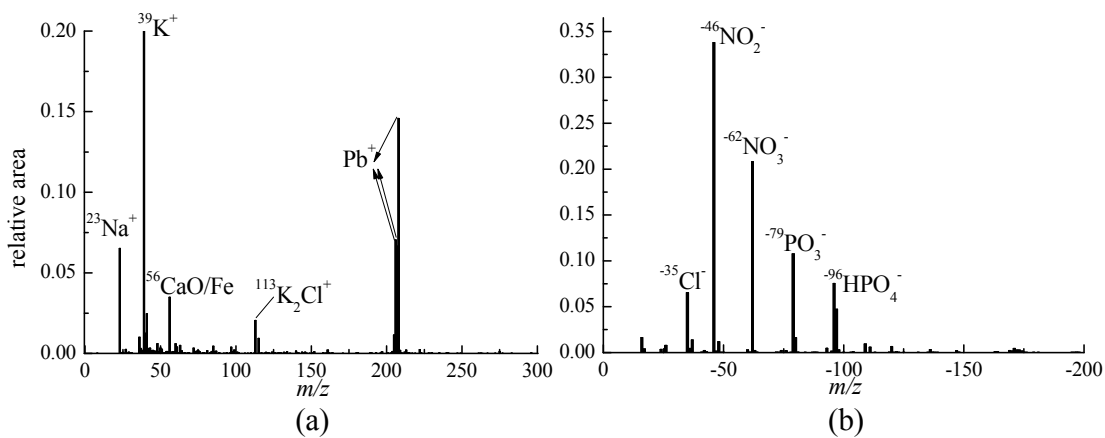
**Fig. S2** Distribution of four particle types in fresh SB particles



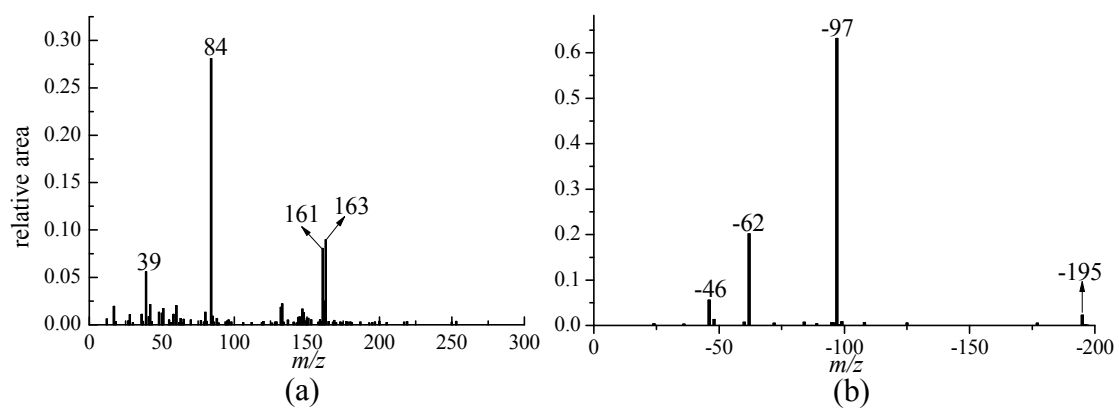
**Fig. S3** ATOFMS average mass spectra of ashes particles: (a) positive MS of ash1, (b) negative MS of ash1, (c) positive MS of ash2 and (d) negative MS of ash2



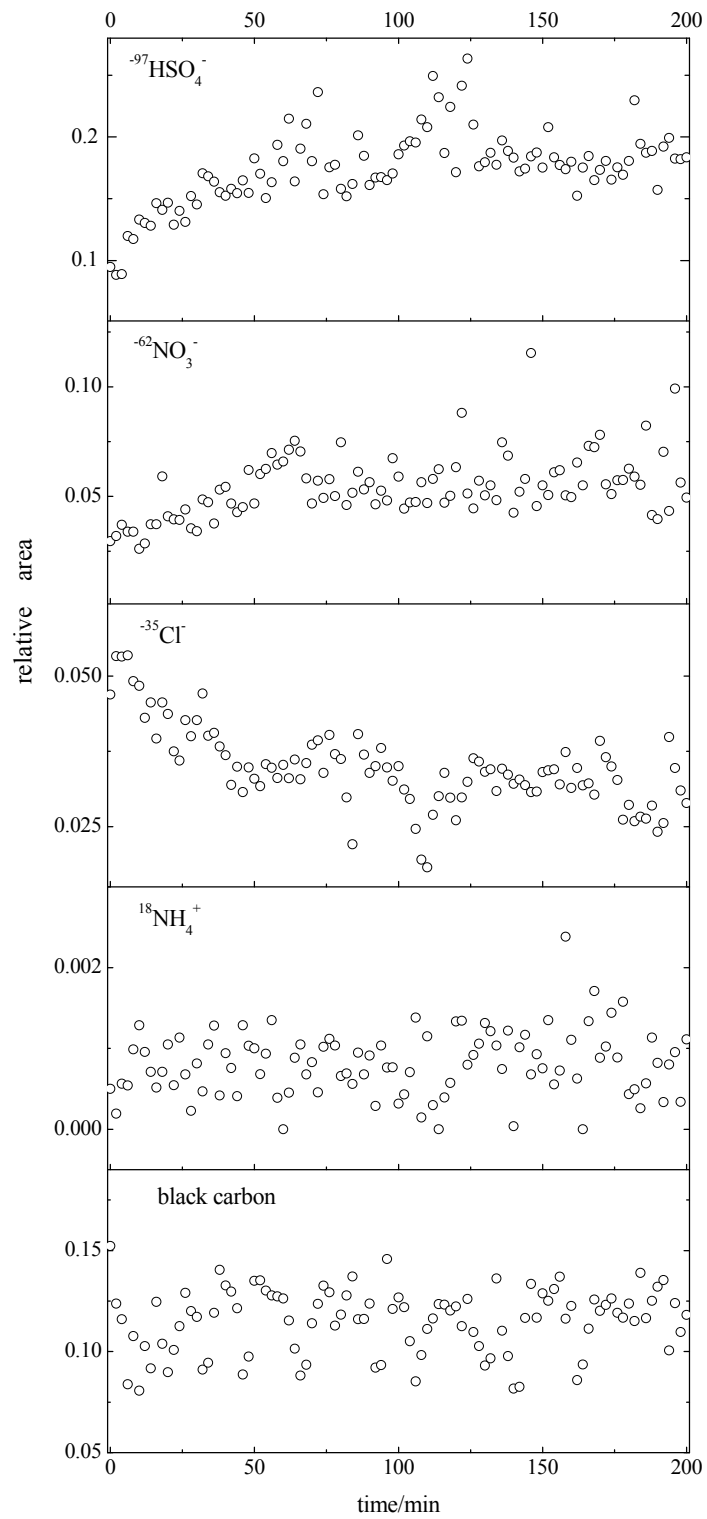
**Fig. S4** ATOFMS average mass spectra of PAHs particles: (a) positive MS and (b) negative MS



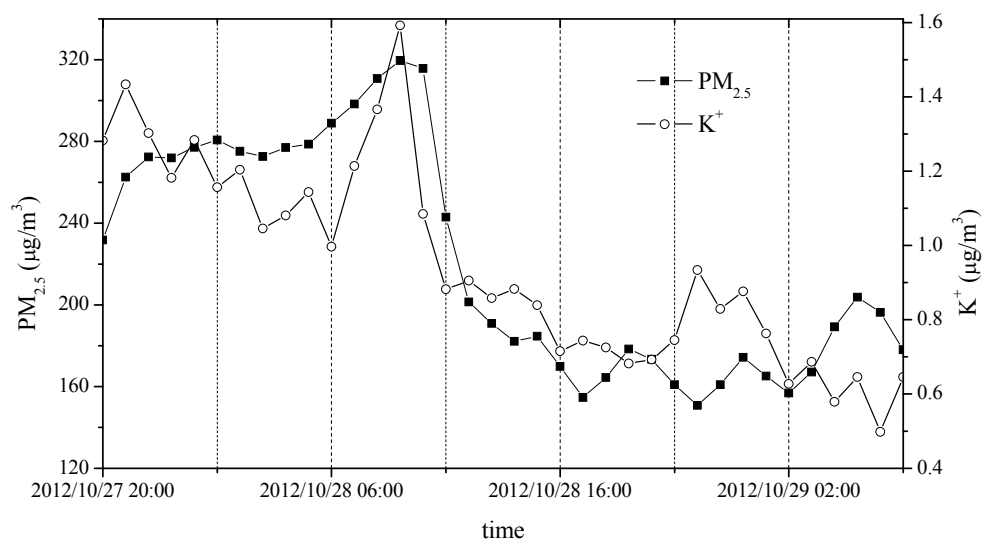
**Fig. S5** ATOFMS average mass spectra of heavy metal particles: (a) positive MS and (b) negative MS of Pb-rich particles



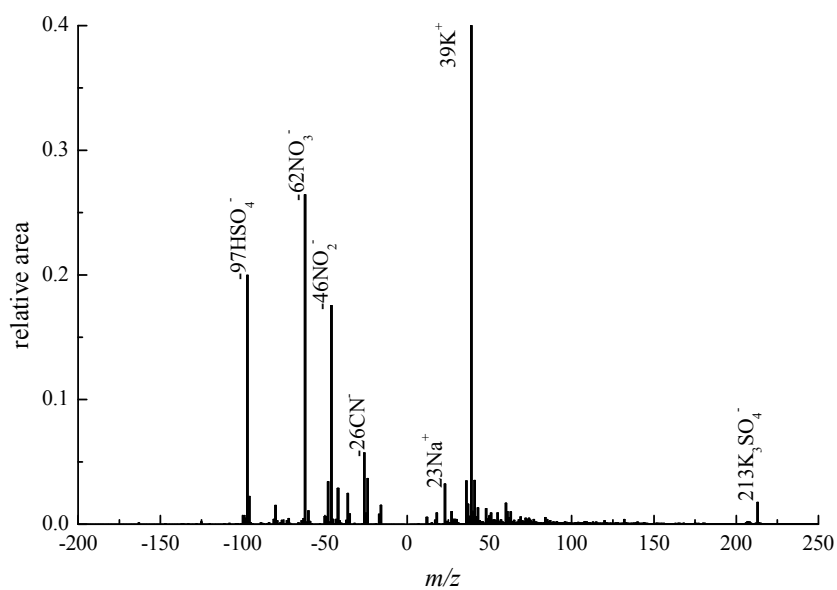
**Fig. S6** ATOFMS average mass spectra of nicotine particles: (a) positive MS and (b) negative MS



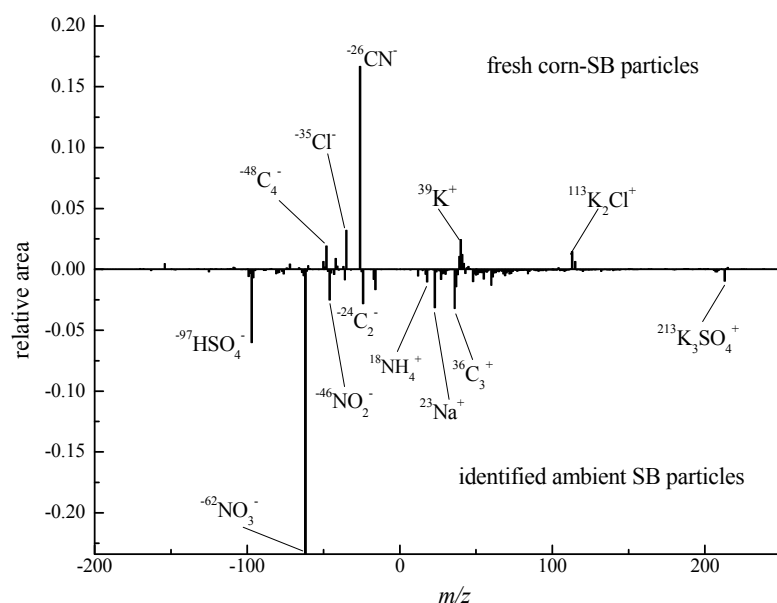
**Fig. S7** Variation of the average relative peak area for  $\text{HSO}_4^-$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{NH}_4^+$  and black carbon during the particle growth



**Fig. S8** PM<sub>2.5</sub> concentration and water-soluble K<sup>+</sup> concentration measured by MARGA



**Fig. S9** ATOFMS average mass spectrum of identified ambient SB particles during 21:00 27th Oct 2012 - 7:00 29th Oct. 2012



**Fig. S10** Subtraction mass spectra for laboratory-made fresh rice-SB particles minus identified ambient SB particles

## References

1. Vassilev S V, Baxter D, Andersen L K, Vassileva C G. An overview of the composition and application of biomass ash. Part 1. Phase-mineral and chemical composition and classification. *Fuel*, 2013, 105(0): 40–76 [doi:10.1016/j.fuel.2012.09.041](https://doi.org/10.1016/j.fuel.2012.09.041)
2. Vassilev S V, Baxter D, Andersen L K, Vassileva C G. An overview of the chemical composition of biomass. *Fuel*, 2010, 89(5): 913–933 [doi:10.1016/j.fuel.2009.10.022](https://doi.org/10.1016/j.fuel.2009.10.022)
3. Pagels J, Dutcher D D, Stolzenburg M R, McMurry P H, Gälli M E, Gross D S. Fine-particle emissions from solid biofuel combustion studied with single-particle mass spectrometry: Identification of markers for organics, soot, and ash components. *Journal of Geophysical Research, D, Atmospheres*, 2013, 118(2): 859–870 [doi:10.1029/2012JD018389](https://doi.org/10.1029/2012JD018389)
4. Silva P J, Liu D Y, Noble C A, Prather K A. Size and chemical characterization of individual particles resulting from biomass burning of local Southern California species. *Environmental Science & Technology*, 1999, 33(18): 3068–3076 [doi:10.1021/es980544p](https://doi.org/10.1021/es980544p)
5. Zhang Y, Wang X, Chen H, Yang X, Chen J, Allen J O. Source apportionment of lead-containing aerosol particles in Shanghai using single particle mass spectrometry. *Chemosphere*, 2009, 74(4): 501–507 [doi:10.1016/j.chemosphere.2008.10.004](https://doi.org/10.1016/j.chemosphere.2008.10.004) PMID:19027137
6. Hammond S K, Leaderer B P, Roche A C, Schenker M. Collection and analysis of Nicotine as a marker for environmental tobacco smoke. *Atmospheric Environment*, 1987, 21(2): 457–462