

Chemical characterization of size-resolved fine particles in human exhaled breath by using single particle aerosol mass spectrometry

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A number of scientific studies have suggested that PM_{2.5} exposure will cause a variety of significant health problems, including premature death (especially related to heart disease), cardiovascular effects, reduced lung development, chronic respiratory diseases, *etc* (USEPA, 2012). Health risks attributed to PM_{2.5} exposure can be evaluated based on the information of particle chemical composition, size and concentration (*e.g.*, particle number, mass per volume). In recent studies, analysis of particles in exhaled human breath has been successfully achieved by using scanning mobility particle sizer (SMPS) (Löndahl *et al.*, 2014) or inductively coupled plasma mass spectrometry (ICP-MS) (Bredberg *et al.*, 2014). However, for the methods aforementioned, chemical composition, size and concentration of particles could not be measured simultaneously. Single particle aerosol mass spectrometry (SPAMS) has been widely used to characterize the size and chemical composition of a single particle (Li *et al.*, 2011) and advanced the studies of source apportionment, aging process and optical properties of ambient aerosols. In contrast, the application of SPAMS to detect particles in exhalation has not been explored yet, and thus in this work, the potential of SPAMS for analyzing particles in human exhaled breath was studied.

The breath samples from 3 young healthy volunteers (24–35 years old) were analyzed by SPAMS (Figure 1). Take the results of one volunteer for example, 304 (\pm 98) particles L⁻¹ were detected in the exhaled breath samples. The particle number concentration is about 13% of that in the inhaled indoor air, suggesting that most inhaled PM_{2.5} have been retained within the human body. The exhaled particles were classified into nine types according to the characteristic peaks in the mass spectra obtained under both positive and negative ion detection modes, namely elemental carbon (EC), organic carbon (OC), aluminium (Al), potassium-rich (K), calcium (Ca), vanadium (V), iron (Fe), sodium- and K-rich (NaK) and amine (ON) types (Table 1). Interestingly, ON and NaK types are only observed in the exhaled breath and tentatively assumed to be endogenous aerosols.

Table 1 Number concentrations (L⁻¹) of each type of particle in exhaled breath and indoor air samples.

Sample	EC	OC	Al	K	Ca	V	Fe	ON	NaK
Breath	24	55	2	14	2	2	2	8	4
Indoor air	192	193	2	85	5	18	4	0	0

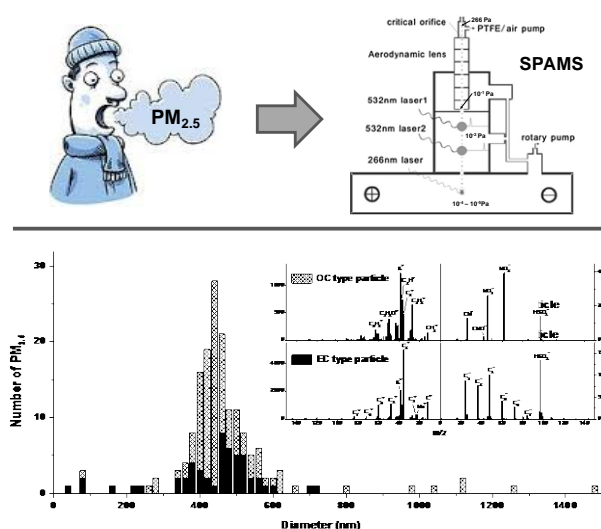


Figure 1. Schematic of monitoring particles in exhaled human breath by SPAMS (up); and sized resolved distribution of two dominant types of particles, *i.e.*, OC particle and EC particle (bottom).

For two dominant types of particles, EC and OC, the particle number concentrations in the exhaled breath are 13% and 28% of that in the inhaled indoor air, respectively (Table 1); meanwhile in the breath samples less peaks occur in the size range of 0.6–1.0 μm and the dominant peaks are around 0.5 μm (Figure 1). The change in the size-distribution pattern may be ascribed to both the deposition and hygroscopic growth of PM_{2.5} in human body.

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