



*Supplement of*

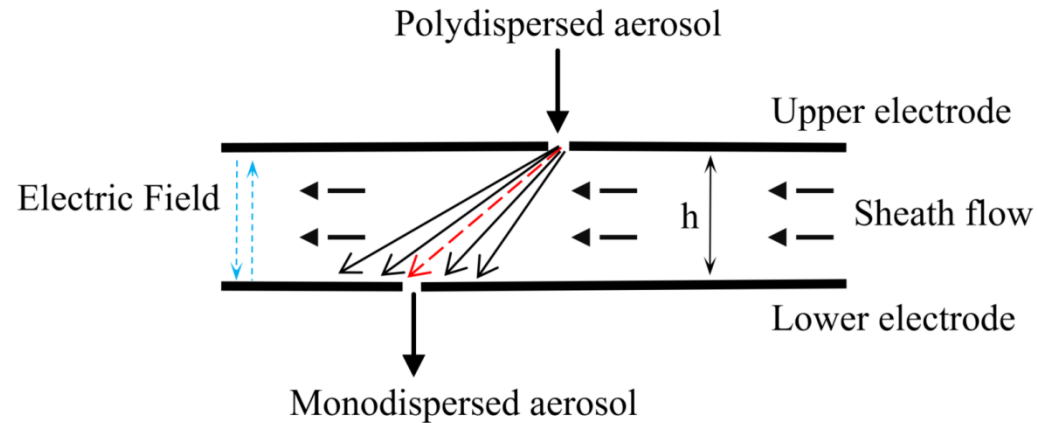
**Characterization of the planar differential mobility analyzer (DMA P5): resolving power, transmission efficiency and its application to atmospheric relevant cluster measurements**

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### S1: Theoretical derivation of the operation principle of planar DMA



**Figure S1 operation principle of planar DMA**

$V_{DMA}$ : Voltage between the two electrodes;

$h$ : Distance between the two electrodes;

The electric field between the two electrodes:  $E = \frac{V_{DMA}}{h}$  (S1)

During the scanning period of planar DMA, the electric field is applied on the  $z$ -direction, a laminar particle-free sheath flow is circulating thorough the capacitor along the  $x$ -direction at the flow rate of  $Q_{sh}$ , and the aerosol flow is fed into the capacitor thorough input slit located at the

top electrode at the flow rate of  $Q_a$ . The direction of aerosol flow is parallel to the electric field and perpendicular to the sheath flow.

The particle velocity in x-direction is given as:  $u_x(z) = \frac{dx}{dt}$  (S2)

The equation can be transformed as  $dx = u_x(z) \cdot dt$  (S3)

The particle velocity in z-direction is given as:

$$u_z = \frac{dz}{dt} = \frac{Q_a}{S_{slit}} + E \cdot Z_p = \frac{Q_a}{S_{slit}} + \frac{V_{DMA} \cdot Z_p}{h} \quad (S4)$$

where  $Z_p$  represent the electric mobility of the particle and  $S_{slit}$  represent the cross-section area of inlet slit.

Since  $\frac{Q_a}{S_{slit}}$  is much smaller than  $\frac{V_{DMA} \cdot Z_p}{h}$ , equation (S4) can be written as:  $u_z = \frac{dz}{dt} = \frac{V_{DMA} \cdot Z_p}{h}$  (S5)

Equation (S5) can be transformed as  $dt = \frac{h}{V_{DMA} \cdot Z_p} \cdot dz$  (S6)

Combined equation (S3) and (S6), we can get the relation that

$$dx = \frac{u_x(z) \cdot h}{V_{DMA} \cdot Z_p} \cdot dz \quad (S7)$$

Integrating equation (S7), we can get the equation that  $\int_0^L dx = \frac{h}{V_{DMA} \cdot Z_p} \int_0^h u_x(z) dz$  (S8)

where L represent the distance between the inlet slit and the monodispersed particle exit.

Assuming that  $\bar{u}_x(z) = \frac{Q_{sh}}{w \cdot h}$ , where w represents the width of the capacitor and w·h represent the cross-section area of the capacitor, the

integral equation can be transformed as  $\int_0^L dx = L = \frac{Q_{sh}}{w \cdot h} \cdot \frac{h}{V_{DMA} \cdot Z_p} \int_0^h dz = \frac{Q_{sh} \cdot h}{w \cdot V_{DMA} \cdot Z_p}$  (S9)

Equation (S9) can be written as  $Z_p = \frac{Q_{sh} \cdot h}{w \cdot U \cdot L}$ , and combined with the assumption that  $Q_{sh} = \bar{u}_x(z) \cdot w \cdot h$ , we can get the expression of

$$Z_p = \frac{\bar{u}_x(z) \cdot h^2}{V_{DMA} \cdot L} \quad (S10)$$

In equation (S10)  $\bar{u}_x(z)$  represent the average speed of sheath flow along z-direction; L and h represent the horizontal distance of inlet the exit and between the two electrodes, respectively;  $V_{DMA}$  represent the voltage applied between the two electrodes.

Account for the planar DMA P5, the sheath flow speed is uniform along z-direction ( $\bar{u}_x(z) = u_x$ ), the physical dimension of L and h are 40mm and 10mm, respectively. The relation of the electric mobility ( $Z_p$ ) and the voltage applied by planar DMA P5 ( $V_{DMA}$ ) can be expressed as:

$$Z_p = \frac{u_x \cdot h^2}{V_{DMA} \cdot L} \quad (S11)$$

## S2: Mobility diameter calculation

Calculation of diameter from mobility (Tammet, 1995; Wiedensohler et al., 2012)

$$Z_p = \frac{neC_c(D_p)}{3\pi\mu D_p} \quad (S12)$$

$$C_c = 1 + \frac{2\lambda}{D_p} (1.165 + 0.483 \exp(-0.997 \frac{D_p 2\lambda}{2\lambda})) \quad (S13)$$

$$\lambda = \lambda_0 \left(\frac{T}{T_0}\right)^2 \left(\frac{P_0}{P}\right) \left(\frac{T_0 + 110.4K}{T + 110.4K}\right) \quad (S14)$$

$$\mu = \mu_0 \left(\frac{T}{T_0}\right)^{3/2} \left(\frac{T_0+110.4K}{T+110.4K}\right) \quad (S15)$$

$n$  is Number of elementary charges on the particle;  $e$  is Elementary charge =  $1.60 \times 10^{-19}$  C;  $C_c$  is Cunningham slip correction;  $D_p$  is Mobility diameter;  $\mu$  is Dynamic gas viscosity;  $\lambda$  is Mean free path of gas;  $T$  is Temperature, and is set as 298.15 K;  $P$  is Pressure, assuming  $P$  equals to 1atm;  $T_0$  is Reference temperature (296.15 K);  $P_0$  is the Reference pressure = 1atm = 101325 Pa;  $\lambda_0$  is Mean free path at 296.15K and 1atm =  $67.3 \times 10^{-9}$  m;  $\mu_0$  is the gas viscosity at 296.15K and 1atm, which is equals to  $1.83245 \times 10^{-5}$  kg m<sup>-1</sup> s<sup>-1</sup>

### S3: Theoretical calculation of the resolving power of planar DMA

Assuming that variances are additive, the resolving power for a planar DMA is given by the following formula:

$$R^{-2} = \left(\frac{\Delta Z_{FWHM}}{Z}\right)^2 = \left(\frac{W}{L_{slit}} \frac{Q_{in}+Q_{out}}{Q_c+Q_m}\right)^2 + \left(\frac{2\sigma\sqrt{2\ln 2}}{L} \frac{\sqrt{L^2+h^2}}{h}\right)^2 \quad (S16)$$

Where  $W$  is the width of the DMA channel at the outlet slit (14.9 mm),  $L_{slit}$  is the monodisperse sampling slit length (6.5 mm),  $Q_{out}$  is the monodisperse sampling flow rate,  $Q_{in}$  is the polydisperse aerosol flow rate,  $Q_c$  is the flow rate at the DMA sheath gas inlet,  $Q_m$  is the flow rate at the DMA sheath gas outlet,  $2\sigma(2\ln 2)^{1/2}$  is the width at half maximum of a Gaussian distribution,  $L$  is the separation channel length,  $h$  is the normal distance between electrodes and  $((L^2+h^2)^{1/2})/h$  is the projection of the Gaussian distribution width into the outlet electrode. In this planar DMA, the polydisperse sample is electrically pushed through the inlet slit; indeed, a small counterflow (0.5 -1 L/min in this work) is exhausted through the inlet slit in order to prevent droplets from entering the DMA. Since  $Q_{in}$  and  $Q_{out}$  are much lower than  $Q_c$  and  $Q_m$ , the following simplification

may be assumed with little error:  $Q_c \sim Q_m \rightarrow Q_c = Q_m = Q$ , where  $Q$  is the sheath gas flow rate through the separation channel. Hereafter only  $Q$  will be considered.

The variance of a Gaussian distribution  $\sigma^2 = 2Dt$ , is controlled by the ion time of residence in the DMA  $t$  and the diffusion coefficient of the ions in the sheath gas  $D$ . The Einstein relation ( $D = ZkT/Ne$ ), relates  $D$  with the electrical mobility  $Z$ , the Boltzmann's constant  $k$ , the gas absolute temperature  $T$  and the net charge on the particle  $Ne$ . The time of residence  $t$  in the planar DMA can be expressed as follows:

$$t = \frac{L}{U} = \left( \frac{h}{ZE} \right) L = \frac{h^2}{ZV_{DMA}} \quad (S17)$$

Where  $E$  is the electric field between the DMA electrodes and  $V_{DMA}$  is the voltage between the electrodes. So  $\sigma^2$  can be expressed as:

$$\sigma^2 = 2Dt = 2 \frac{ZkT}{Ne} t = 2 \frac{ZkT}{Ne} \frac{h^2}{ZV_{DMA}} = \frac{2kTh^2}{V_{DMA}Ne} \quad (S18)$$

And  $Q$  can be expressed as a function of the Reynolds number:

$$Re = \frac{Uh}{\nu} = \frac{Q}{W\nu} \quad (S19)$$

Where  $U$  is the sheath gas velocity in the DMA channel and  $\nu$  is the kinematic viscosity of the gas. Then R can be rewritten as:

$$R^{-1} = \sqrt{\left[ \left( \frac{Q_{in} + Q_{out}}{L_{slit}^2 Re \nu} \right)^2 + \frac{16 \ln 2 kT}{V_{DMA} Ne} \left( 1 + \left[ \frac{h}{L} \right]^2 \right) \right]} \quad (S20)$$

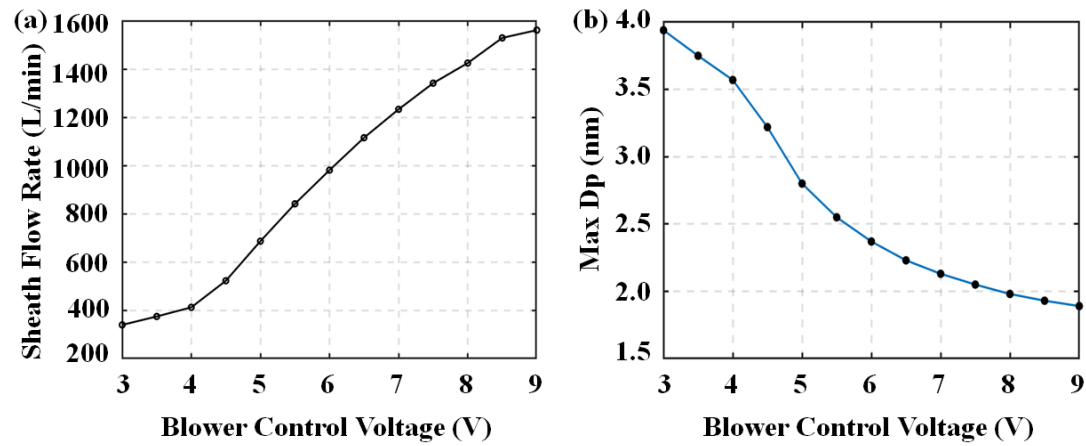
Convective diffusion problems at large Reynolds numbers are well known to be governed by the Peclet number  $Pe$  defined as:

$$Pe = Re \frac{\nu}{D} = \frac{Uh}{D} = \frac{ZV_{DMA}L}{hD} = \frac{V_{DMA}LNe}{hkT} \quad (S21)$$

Therefore, R can be expressed as a function of  $Pe$  number:

$$R^{-1} = \sqrt{\left[ \left( \frac{Q_{in} + Q_{out}}{L_{slit} 2 Re v} \right)^2 + \frac{16 \ln 2}{Pe} \left( \frac{L}{h} + \frac{h}{L} \right) \sqrt{\frac{16 \ln 2}{Pe} \left( \frac{L}{h} + \frac{h}{L} \right)} \right]} \quad (S22)$$

**S4: Supplementary figures and tables**



**Figure S2 The relation of blower control voltage with sheath flow rate and corresponding DMA P5 sizing range**

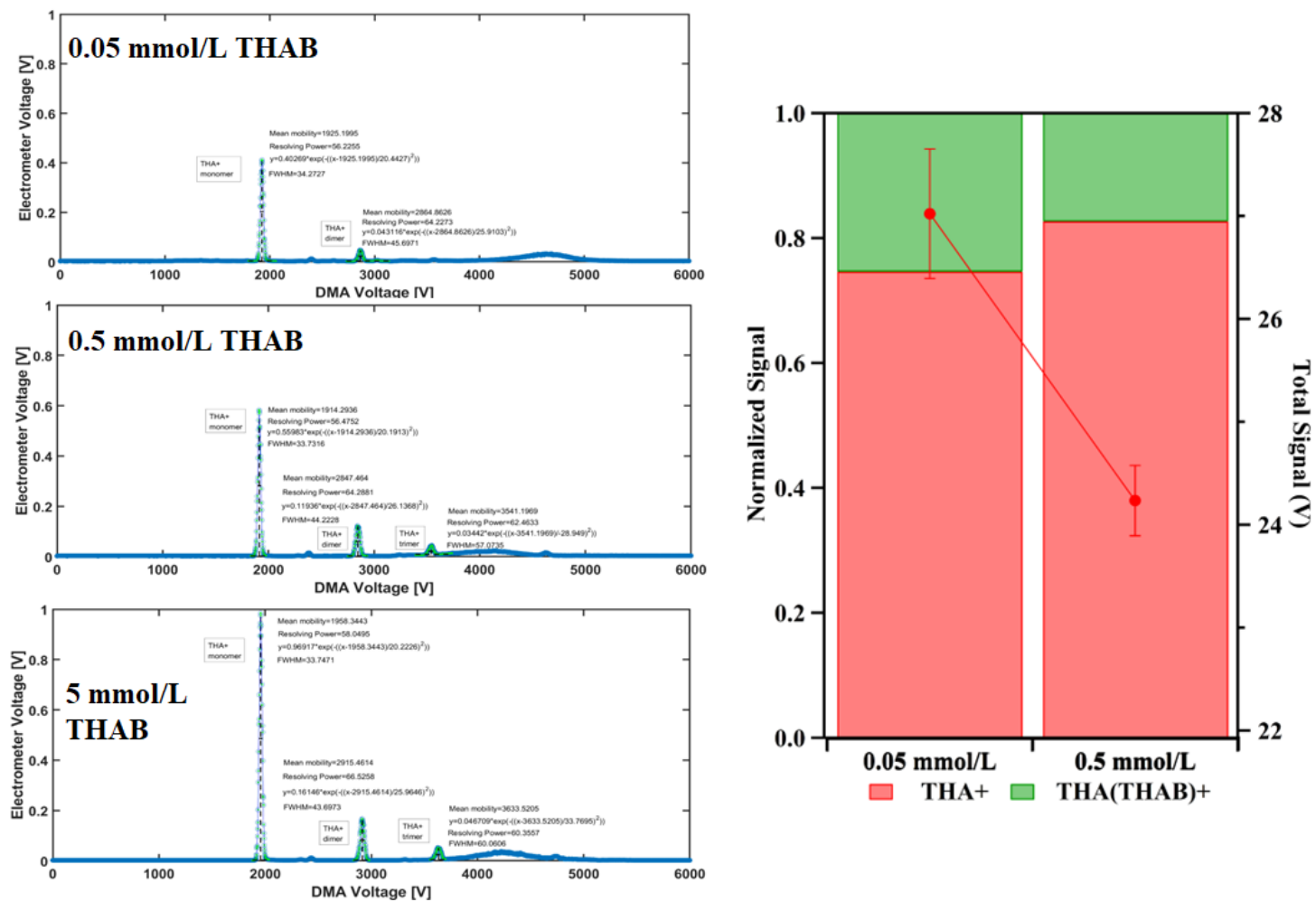


Figure S3 The positive ion mobility spectrum of THAB under suction mode ( $V_{\text{blower}} = 5\text{V}$ ,  $Q_{\text{in}} = 5\text{L/min}$ ,  $Q_{\text{out}} = 1.5\text{L/min}$ ) with different solution concentrations



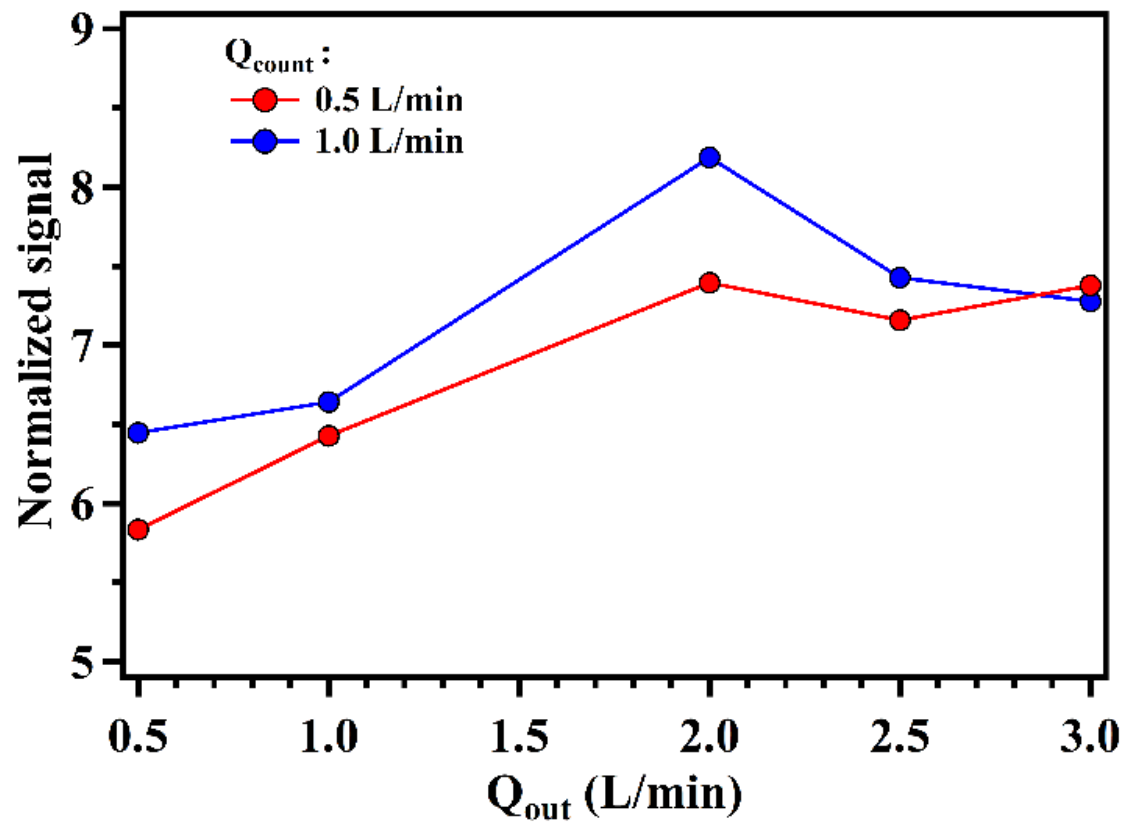


Figure S4 THA<sup>+</sup> Signal intensity normalized by monodispersed flow rate

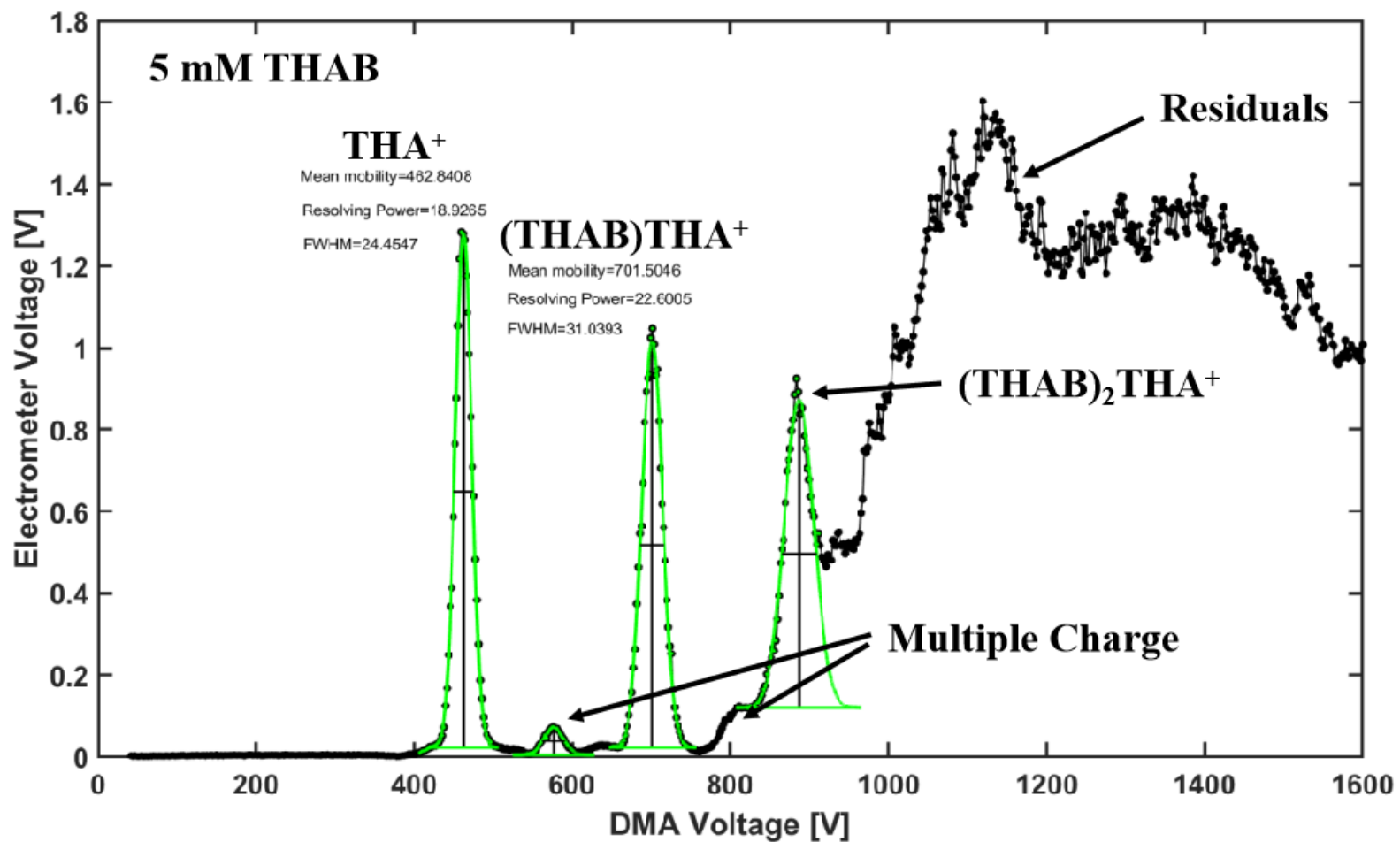


Figure S5 Positive ion mobility spectrum of electrospayed THAB solution obtained from HalfMini + Lynx E12

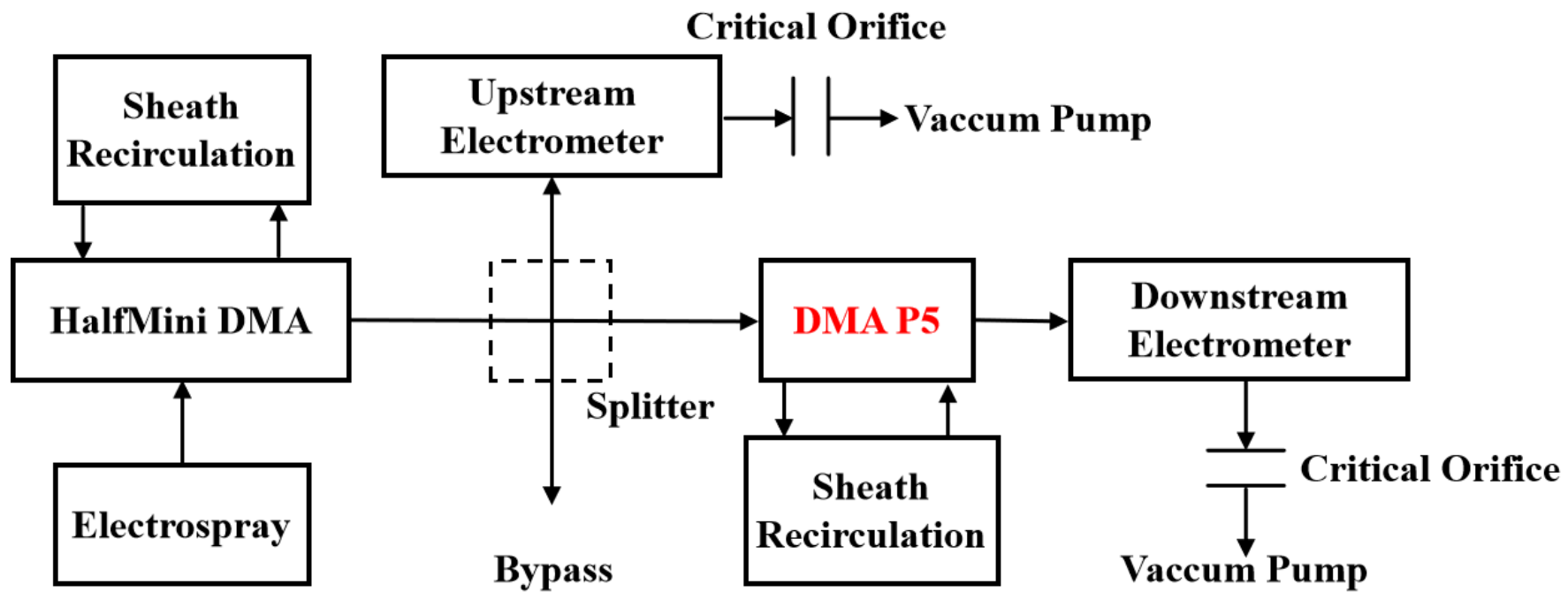


Figure S6 Schematic diagram of tandem DMA system

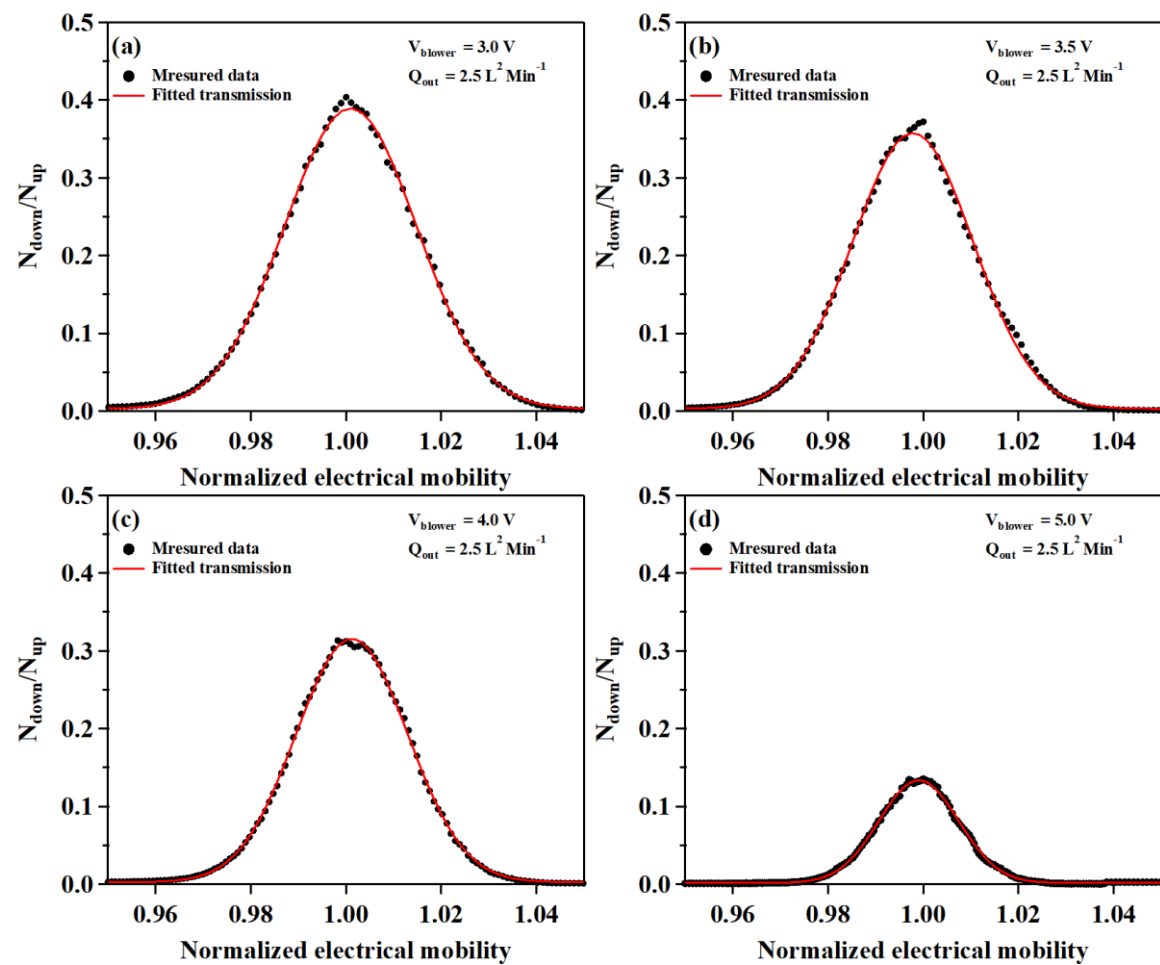


Figure S7 The distribution of transmission efficiency of the DMA P5 when classifying  $\text{THA}^+$  under different sheath flow rate, with  $Q_{\text{out}} = 2.5 \text{ L/min}$

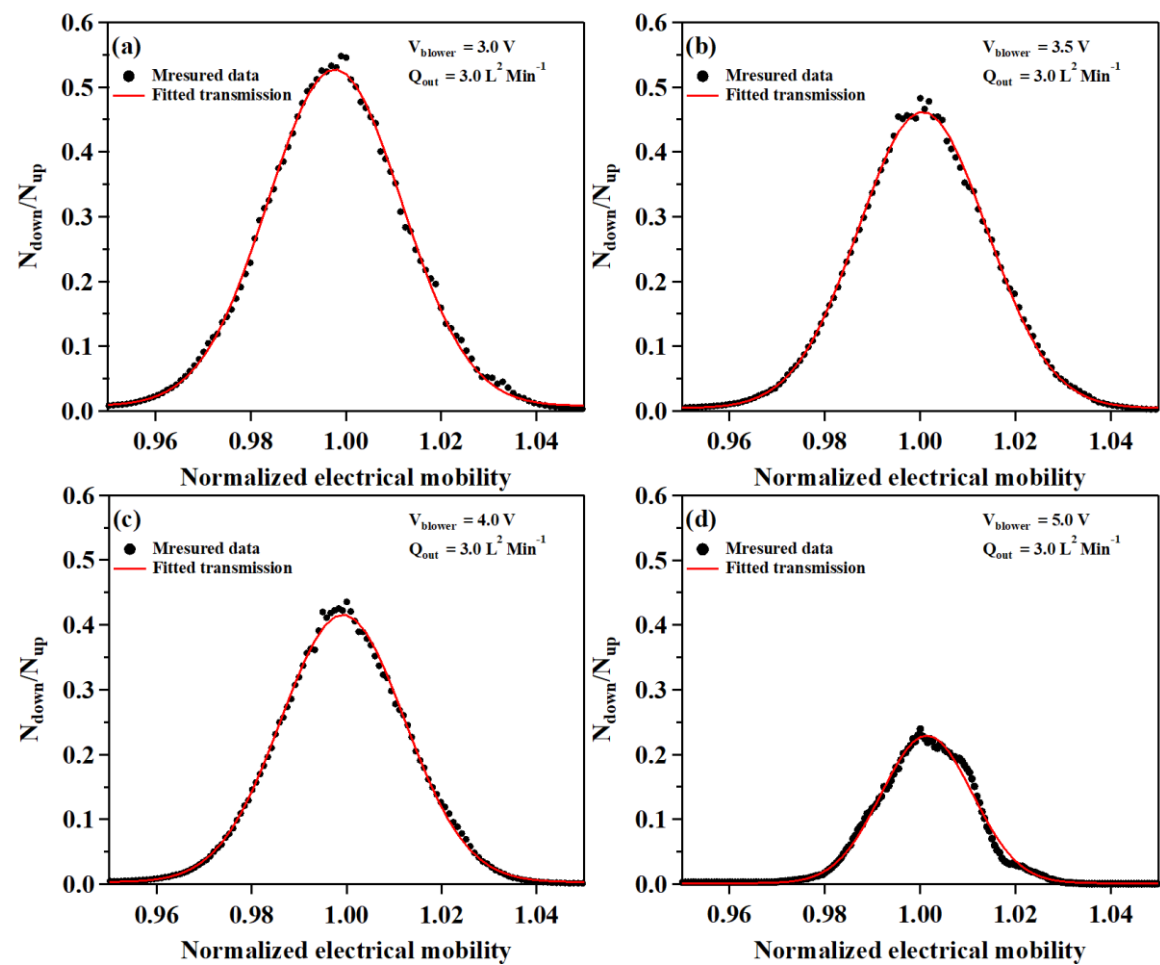


Figure S8 The distribution of transmission efficiency of the DMA P5 when classifying  $\text{THA}^+$  under different sheath flow rate, with  $Q_{\text{out}} = 3.0 \text{ L/min}$

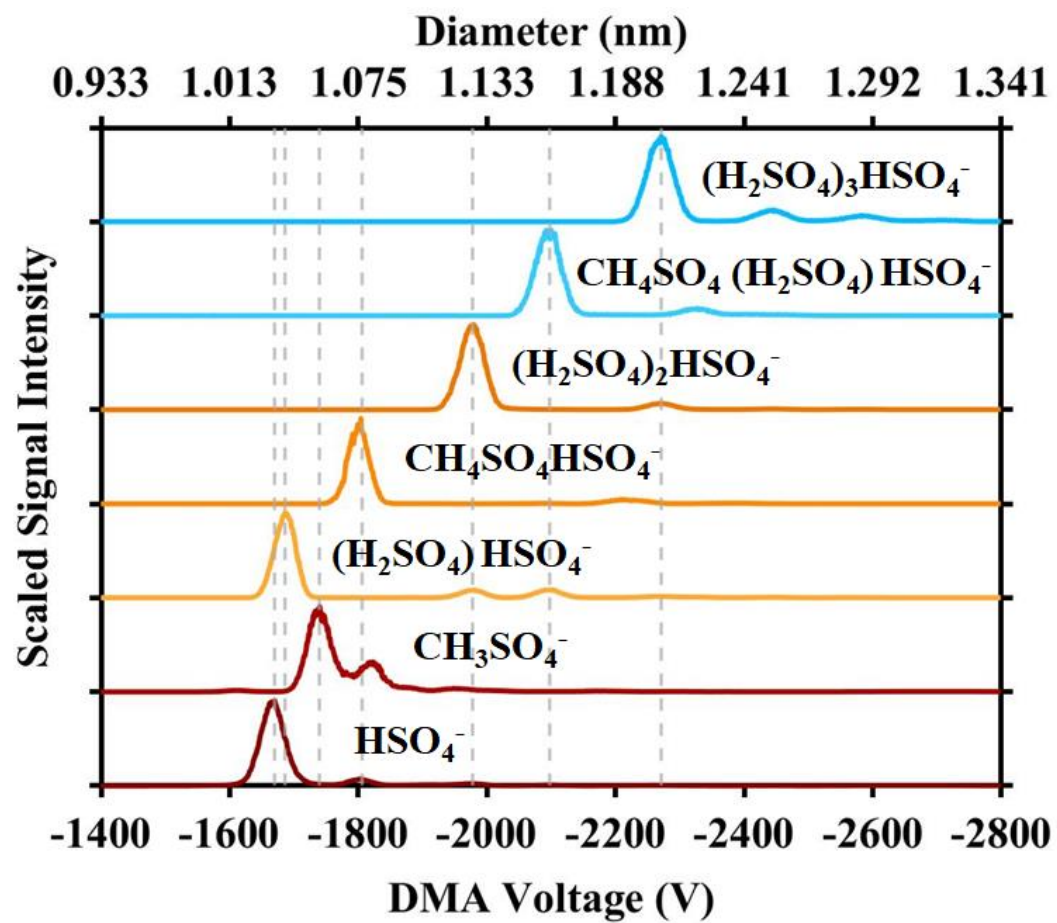


Figure S9 Ion mobility distribution of the main identified clusters. Table S1 Inverse mobilities  $1/Z$  ( $\text{V s/cm}^2$ ) for four tetra-alkyl ammonium positive ions

**Table S1 Inverse mobilities 1/Z (V s/cm<sup>2</sup>) for four tetra-alkyl ammonium positive ions**

Peak <sup>+</sup>	TMAI		TBAI		THAB		TDAB	
	this work	Ude et al.	this work	Ude et al.	this work	Ude et al.	this work	Ude et al.
		2005		2005		2005		2005
A <sup>+</sup>	0.458	0.459	0.723	0.718	1.03	1.03	1.269	1.285
A <sup>+</sup> (AB)	0.667	0.677	1.164	1.153	1.533	1.529	1.811	1.846
A <sup>+</sup> (AB) <sub>2</sub>	-		1.475	1.450	1.898	1.893	-	

**Table S2 Inverse mobilities 1/Z (V s/cm<sup>2</sup>) for four tetra-alkyl ammonium negative ions**

Peak <sup>-</sup>	TMAI	TBAI	THAB	TDAB
B <sup>-</sup>	0.423	0.422	0.436	0.436

**References**

Tammet, H.: SIZE AND MOBILITY OF NANOMETER PARTICLES, CLUSTERS AND IONS, J. Aerosol Sci., 6, (3), 459-475, [https://doi.org/10.1016/0021-8502\(94\)00121-E](https://doi.org/10.1016/0021-8502(94)00121-E), 1995.

Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., et al.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, Atmos. Meas. Tech., 5, (3), 657-685, 2012.