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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

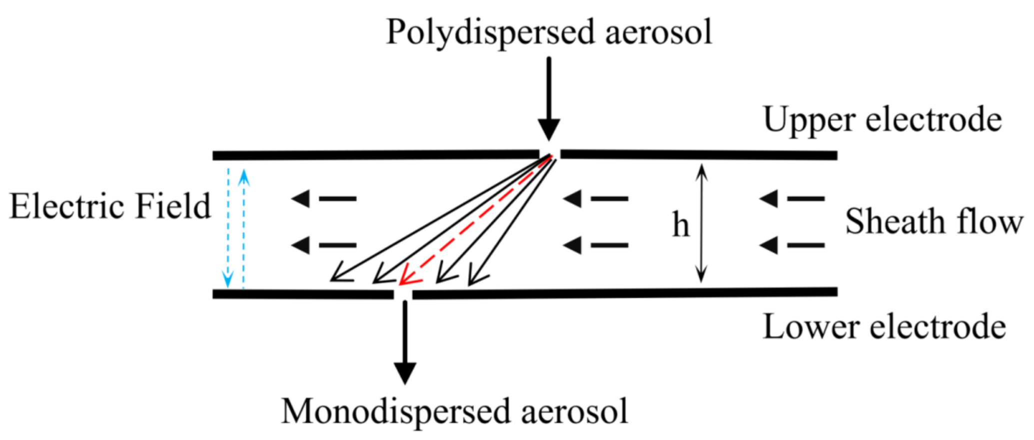

Figure S1 operation principle of planar DMA

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: $u_x(z) = \frac{d_x}{d_t}$ (S2)

The equation can be transformed as $d_x = u_x(\mathrm{z}) \cdot d_t$ (S3)

The particle velocity in z-direction is given as:

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

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 $d_t = \frac{h}{V_{DMA} \cdot Z_p} \cdot d_z$ (S6)

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

$$d_x = \frac{u_x(z) \cdot h}{V_{DMA} \cdot Z_p} \cdot d_z \quad \text{(S7)}$$

Integrating equation (S7), we can get the equation that $\int_0^L d_x = \frac{h}{V_{DMA} \cdot Z_p} \int_0^h u_x(z) d_z$ (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 $\int_0^L d_x = L = \frac{Q_{sh}}{w \cdot h} \cdot \frac{h}{V_{DMA} \cdot Z_p} \int_0^h d_z = \frac{Q_{sh} \cdot h}{w \cdot V_{DMA} \cdot Z_p}$ (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 $Q_{sh} = \bar{u}_x(z) \cdot w$ ·h, we can get the expression of

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

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：

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

S2: Mobility diameter calculation

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

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

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

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

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

n is Number of elementary charges on the particle; e is Elementary charge = 1.60×10^{-19} C; C_c is Cunningham slip correction; D_p is Mobility diameter; μ is Dynamic gas viscosity; λ 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; λ_0 is Mean free path at 296.15K and 1atm = 67.3 $\times$ 10^{-9} m; μ_0 is the gas viscosity at 296.15K and 1atm, which is equals to 1.83245 $\times$ 10^{-5} kg m^{-1} s^{-1}

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:

$$R^{-2} = \left(\frac{\Delta Z_{FWHM}}{Z}\right)^2 = \left(\frac{W}{L_{slit}} \frac{Q_{in}+Q_{out}}{Q_c+Q_m}\right)^2 + \left(\frac{2\sigma\sqrt{2ln2}}{L} \frac{\sqrt{L^2+h^2}}{h}\right)^2 \qquad \text{(S16)}$$

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, $2\sigma(2ln2)^{1/2}$ 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 ($D = ZkT/N_e$), 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:

$$t = \frac{L}{U} = \left(\frac{h}{ZEL}\right) L = \frac{h^2}{ZV_{DMA}} \qquad \text{(S17)}$$

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

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

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

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

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

$$R^{-1} = \sqrt{\left[\left(\frac{Q_{in}+Q_{out}}{L_{slit}2\,Re\,\nu}\right)^2 + \frac{16ln2kT}{V_{DMA}N_e}\left(1+\left[\frac{h}{L}\right]^2\right)\right]} \qquad \text{(S20)}$$

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

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

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

$$R^{-1} = \sqrt{\left[\left(\frac{Q_{in}+Q_{out}}{L_{slit}2\,Re\,\nu}\right)^2 + \frac{16ln2}{Pe}\left(\frac{L}{h}+\frac{h}{L}\right)\sqrt{\frac{16ln2}{Pe}\left(\frac{L}{h}+\frac{h}{L}\right)}\right]} \qquad \text{(S22)}$$

S4: Supplementary figures and tables

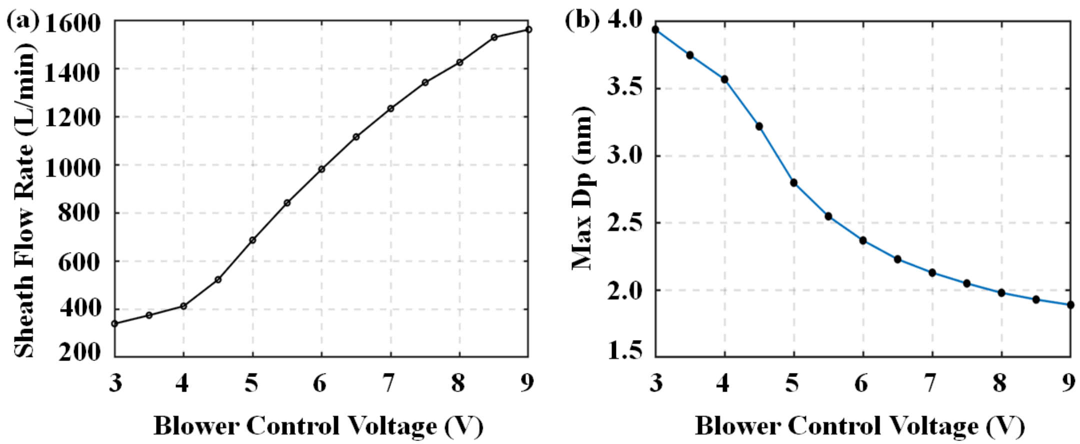

Figure S2 The relation of blower control voltage with sheath flow rate and corresponding DMA P5 sizing range

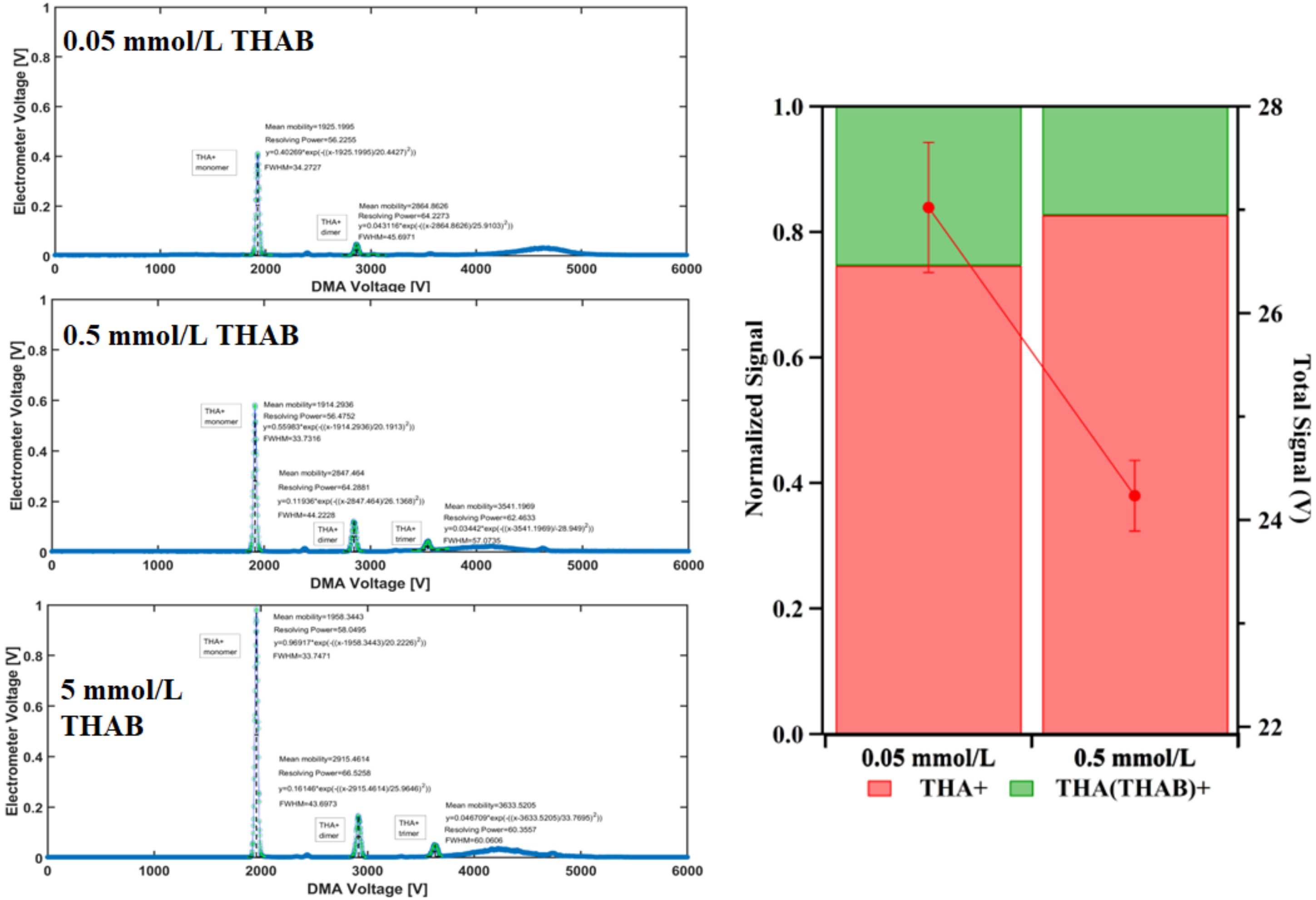

Figure S3 The positive ion mobility spectrum of THAB under suction mode (V_{blower} = 5V, Q_{in}= 5L/min, Q_{out}= 1.5L/min) with different solution concentrations

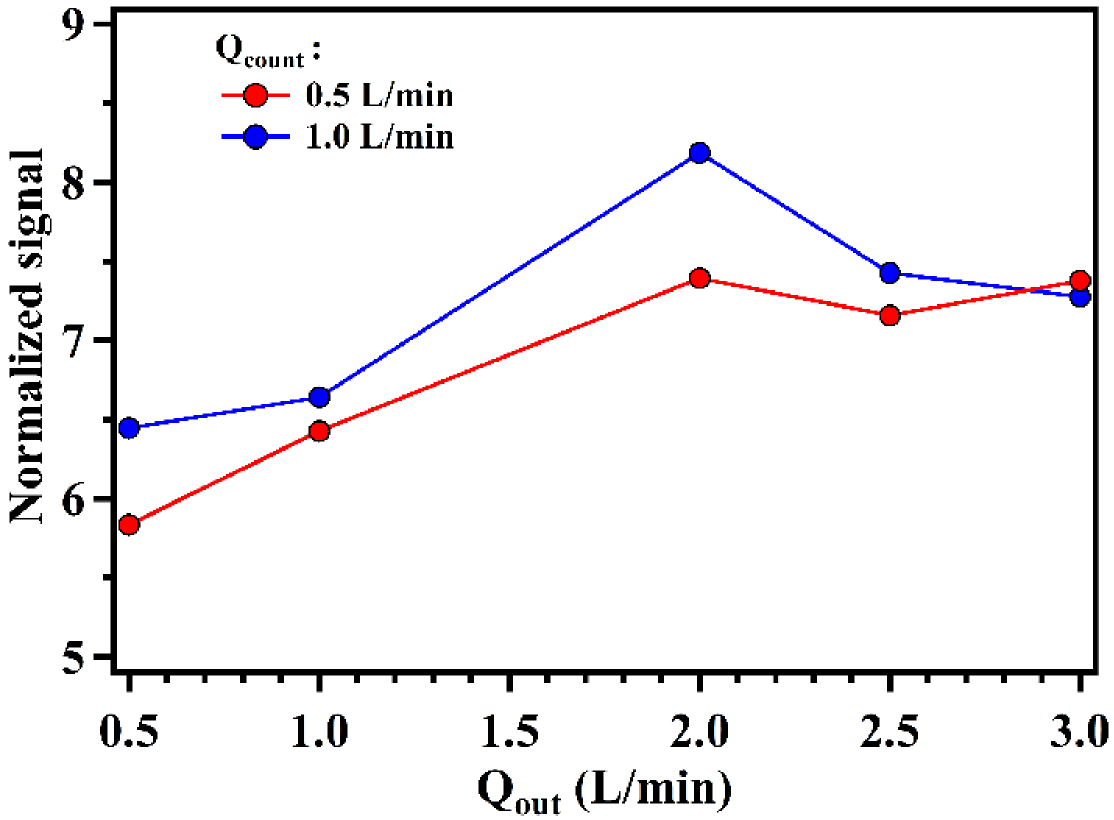

Figure S4 THA^+ Signal intensity normalized by monodispersed flow rate

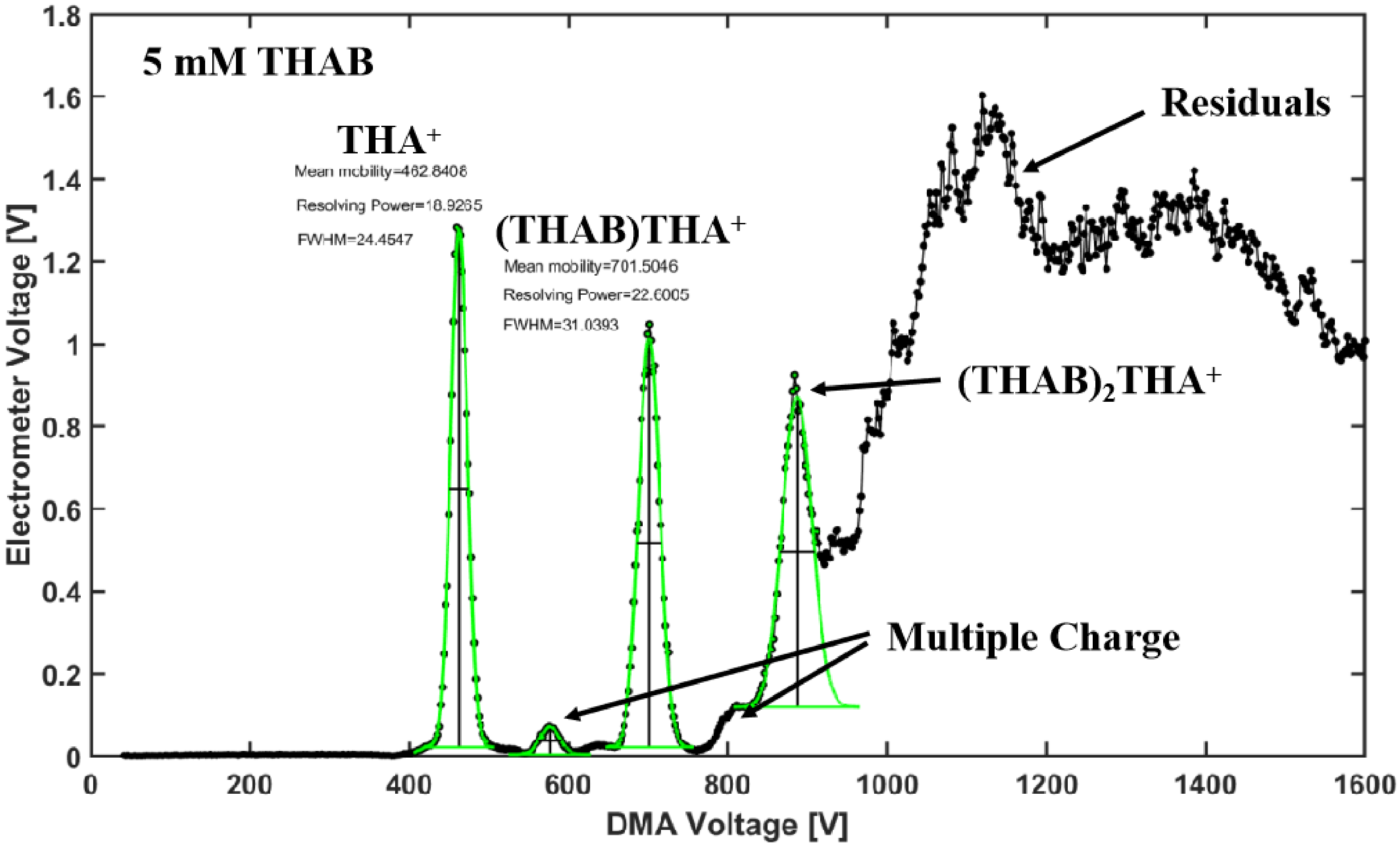

Figure S5 Positive ion mobility spectrum of electrospraied THAB solution obtained from HalfMini + Lynx E12

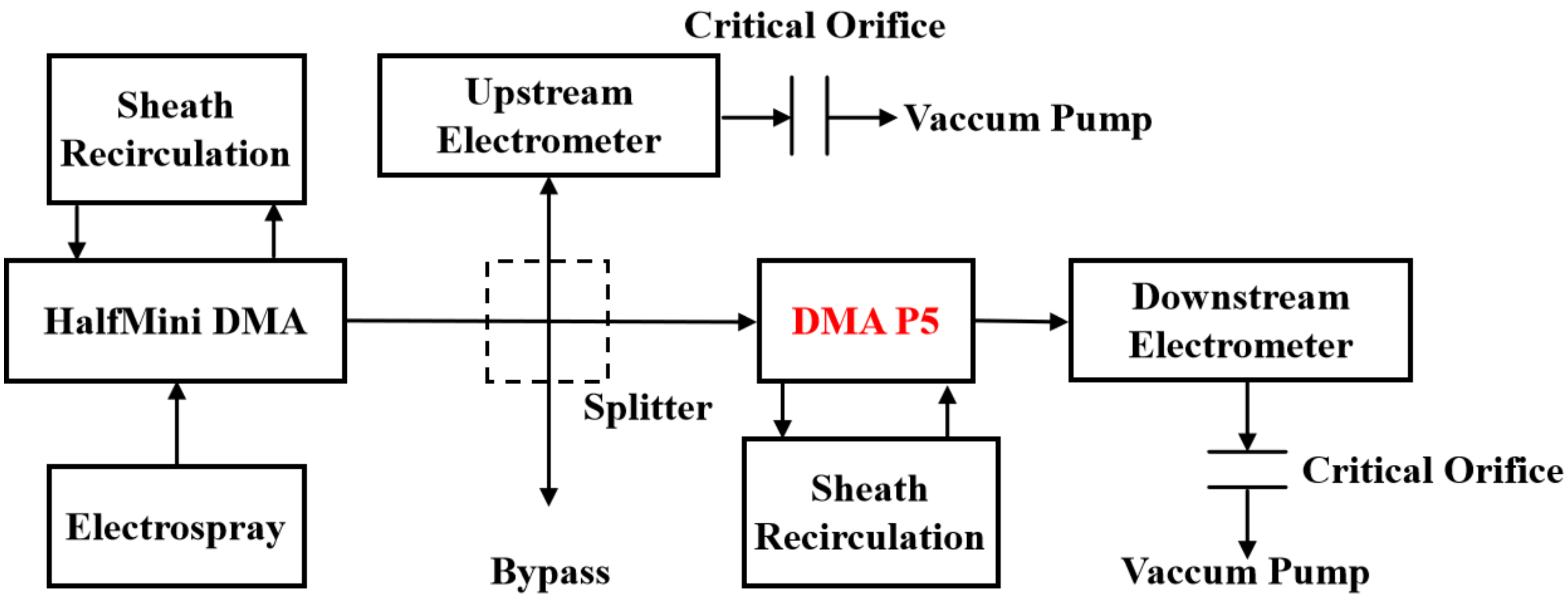

Figure S6 Schematic diagram of tandem DMA system

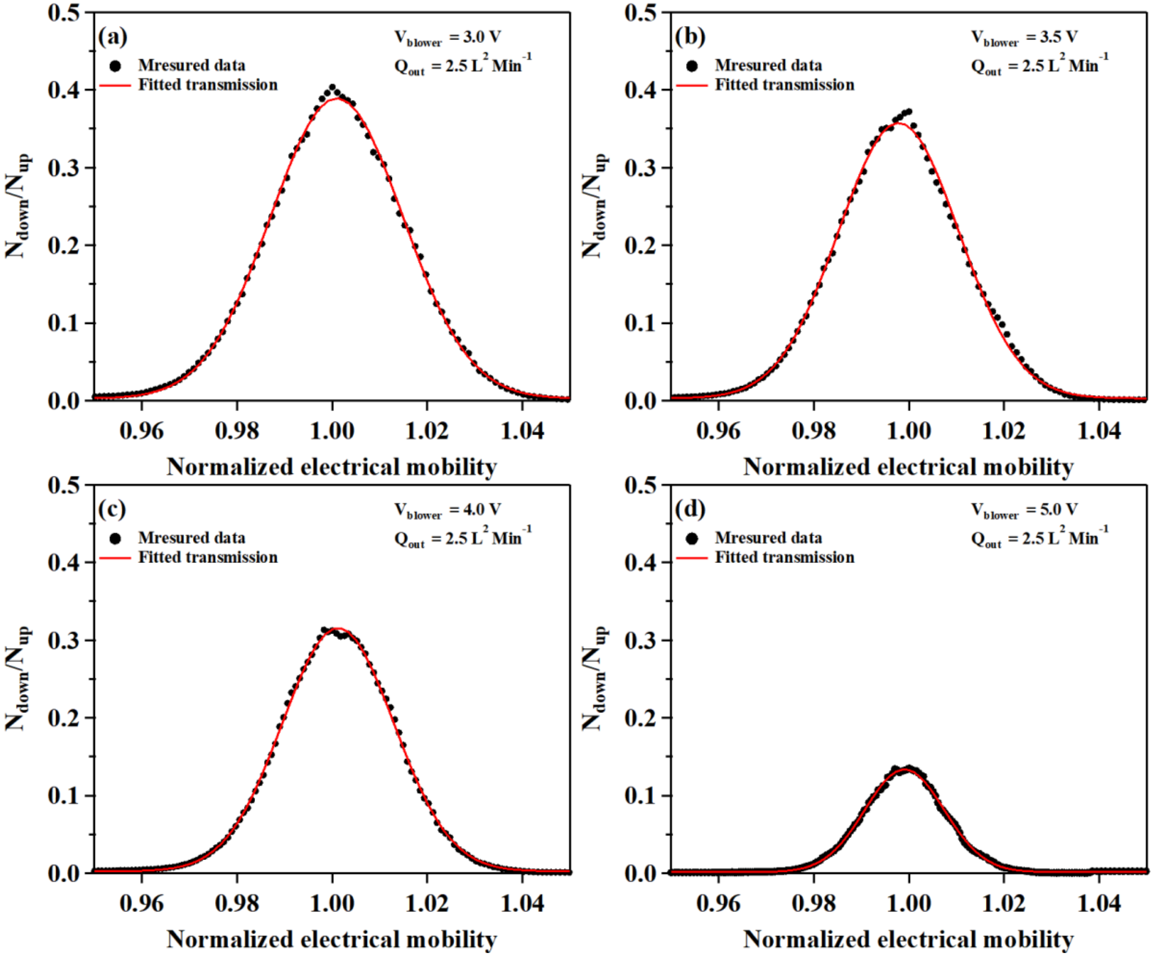

Figure S7 The distribution of transmission efficiency of the DMA P5 when classifying THA^+ _under different sheath flow rate, with Q_{out}= 2.5 L/min

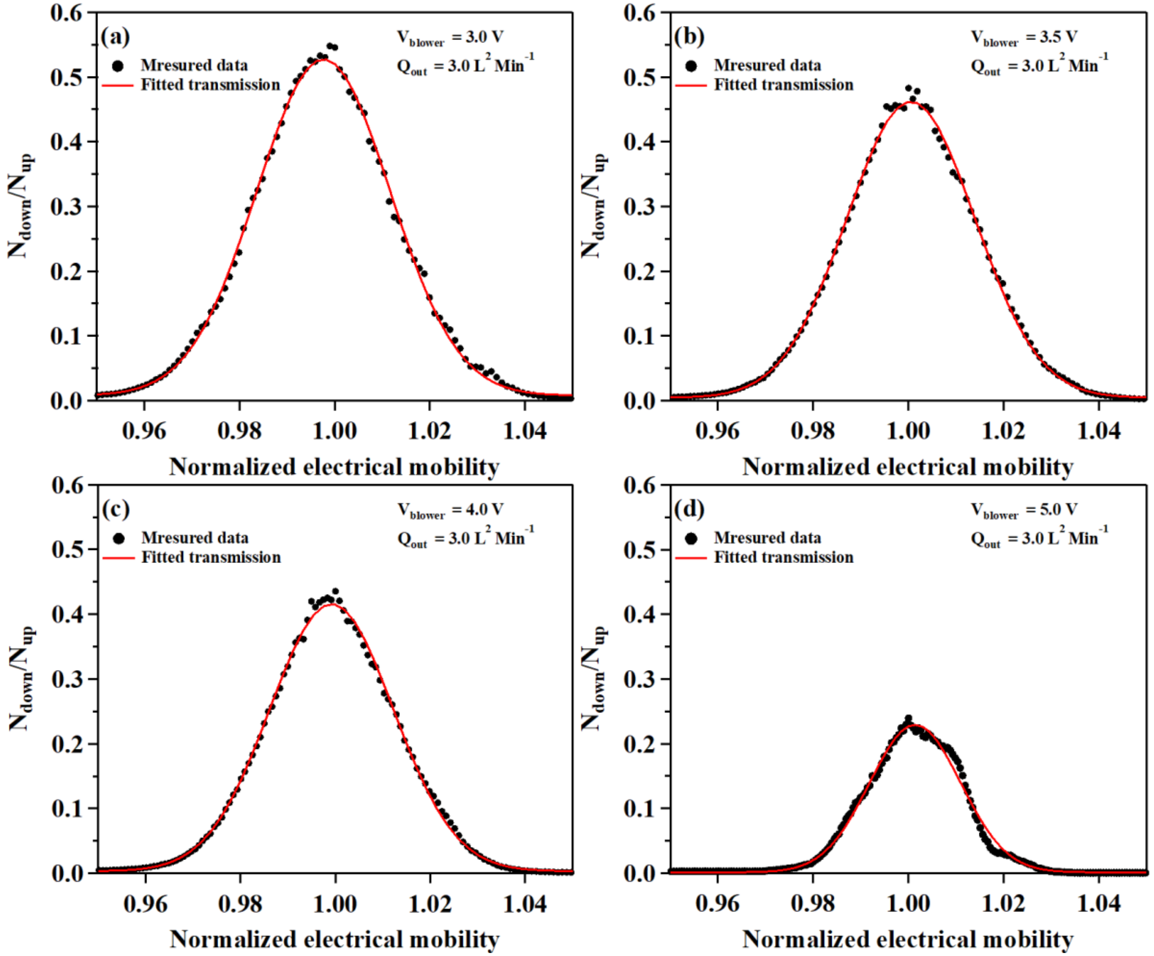

Figure S8 The distribution of transmission efficiency of the DMA P5 when classifying THA^+ _under different sheath flow rate, with Q_{out}= 3.0 L/min

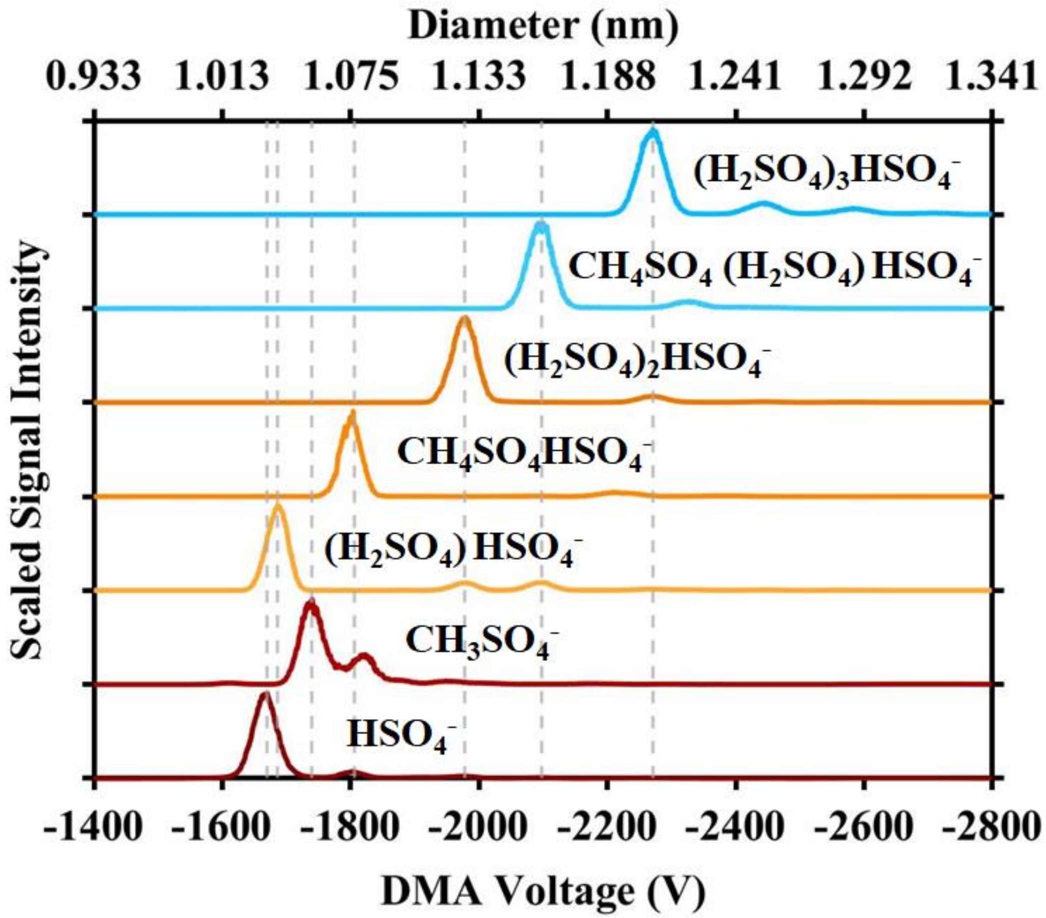

Figure S9 Ion mobility distribution of the main identified clusters.Table S1 Inverse mobilities 1/Z (V s/cm^2) for four tetra-alkyl ammonium positive ions

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

Peak$^+$	TMAI		TBAI		THAB		TDAB	
	this work	Ude et al. 2005	this work	Ude et al. 2005	this work	Ude et al. 2005	this work	Ude et al. 2005
A^+	0.458	0.459	0.723	0.718	1.03	1.03	1.269	1.285
$A^+(AB)$	0.667	0.677	1.164	1.153	1.533	1.529	1.811	1.846
$A^+(AB)_2$	-		1.475	1.450	1.898	1.893	-	

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

Peak$^-$	TMAI	TBAI	THAB	TDAB
B^-	0.423	0.422	0.436	0.436

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