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Effects of multi-charge on aerosol hygroscopicity measurement by a HTDMA

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1. The procedures of calculating the number ratio of particles carrying different charges.

Step 1. Calculate the particle charge distribution.

The particle charge distribution at each size is based on a theoretical model developed by Wiedensohler et al. (1986). To calculate the fraction of particles carrying zero, one or two charges, use the equation below:

$$f(D_p, N) = 10^{\left[\sum_{i=0}^{5} a_i(N)(log\frac{D_p}{nm})^i\right]}$$

a_i(N)	N=-2	N=-1	N=0	N=1	N=2
a_0	-26.3328	-2.3197	-0.0003	-2.3484	-44.4756
a_1	35.9044	0.6175	-0.1014	0.6044	79.3772
a_2	-21.4608	0.6201	0.3073	0.4800	-62.8900
a_3	7.0867	-0.1105	-0.3372	0.0013	26.4492
a_4	-1.3088	-0.1260	0.1023	-0.1553	-5.7480
a_5	0.1051	0.0297	-0.0105	0.0320	0.5049

For the fraction of particles carrying three or more charges, use the equation below:

$$f(D_p, N) = \frac{e}{\sqrt{4\pi^2\varepsilon_0 D_p \kappa T}} \exp \frac{-\left[N - \frac{2\pi\varepsilon_0 D_p \kappa T}{e^2} \ln(\frac{Z_{i+}}{Z_{i-}})\right]^2}{2\frac{2\pi\varepsilon_0 D_p \kappa T}{e^2}}$$

where e is the elementary charge of 1.60217733E-19 coulomb; ε_0 is the dielectric constant of 8.854187817E-12 farad/m; the D_p is the particle diameter in [m]; κ is the Boltzmann's constant of 1.380658E-23 joule/K; T is the temperature in [K]; N is the number of elementary charge units; $\frac{Z_{i+}}{Z_{i-}}$ is the ion mobility ratio of 0.875.

Here we present bipolar particle charge distribution with number of charges up to 4 over the size range of 10-1000 nm.

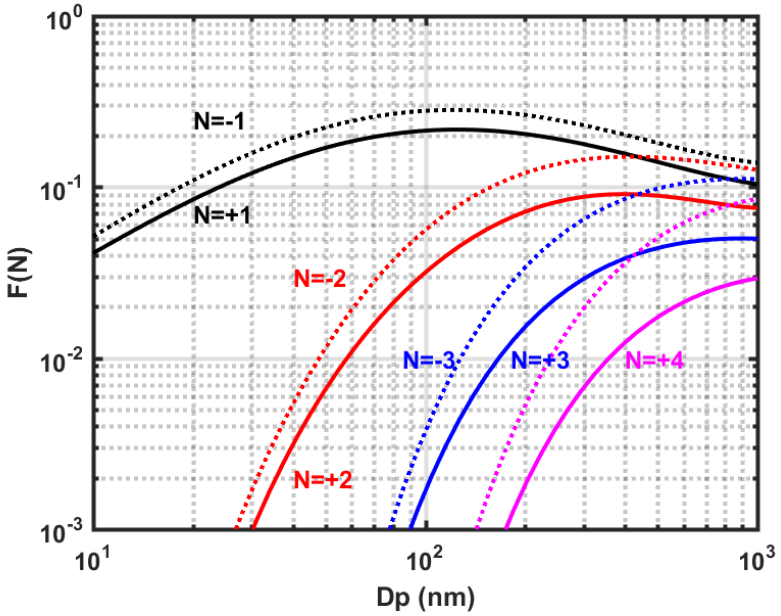

Fig.S1: Calculated number fraction of bipolar charged particles as a function of particle size. Curves decreasing in maximum charged fraction from left to right represent particles with 1 through 4 charges respectively.

Step 2: Calculate the DMA transfer function and Kernel function for each size set at DMA. In our field measurement, the DMA (BMI, Model 2100) selected those negatively charged particles. The sample/sheath ratio of 0.75/4 is used to calculate the transfer function. Then the Kernel function can be obtained from:

$$\mathrm{G}\left(D_p^*, x\right) = \sum_{v=1}^{\infty} F(x, N)\Omega(x, N, D_p^*)$$

The following is an example for the DMA set size of 100 nm.

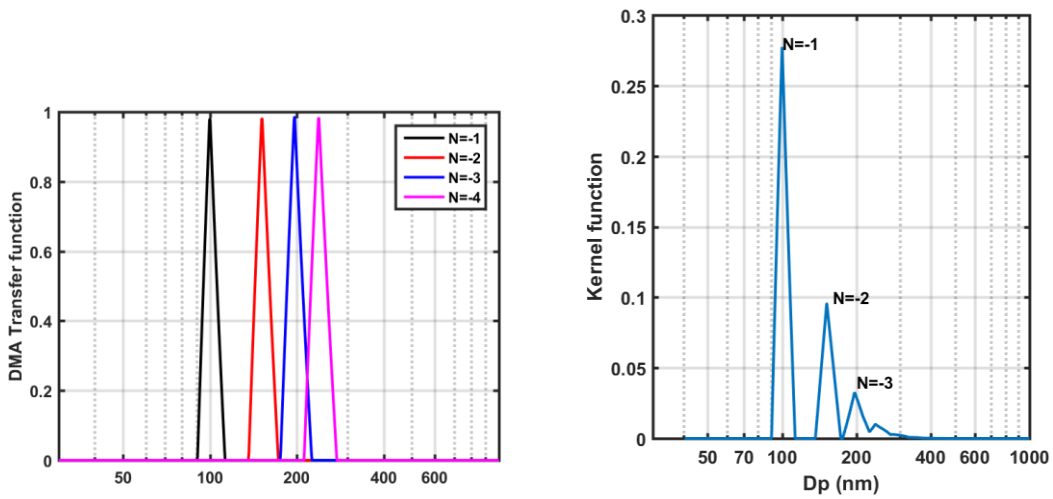

Fig.S2: The left figure is the calculated DMA transfer function for the DMA set size of 100 nm for different charges carried. The right figure is the calculated Kernel function of particles as a function of particle size. Triangle peaks from left to right represent particles with increasing number of charges.

Step 3. Calculate the (1) the total number concentration of particles that can pass through the DMA and (2) the number concentration of particles carrying v charges. In this step, particle number size distribution data is needed.

$$\mathrm{N}(D_p^*) = \int_0^{\infty} G\left(D_p^*, x\right) n(x) dx$$

$$N_v(D_p^*) = \int_0^{\infty} G_v\left(D_p^*, x\right) n(x) dx$$

Here we present a particle number size distribution data during the relatively polluted period in our field measurement. When combined with the total kernel function or charge-resolved kernel function, we can obtain the corresponding number concentration of particles that can pass through the DMA. Fig.S3 shows an example when the DMA set size is 100 nm. It can be seen that when the accumulation mode particles increase, the doubly or triply charged particles also increase greatly. In this case, when integrated over the whole size range, the singly charged particles only constitute 55% of all the particles that can pass through the DMA.

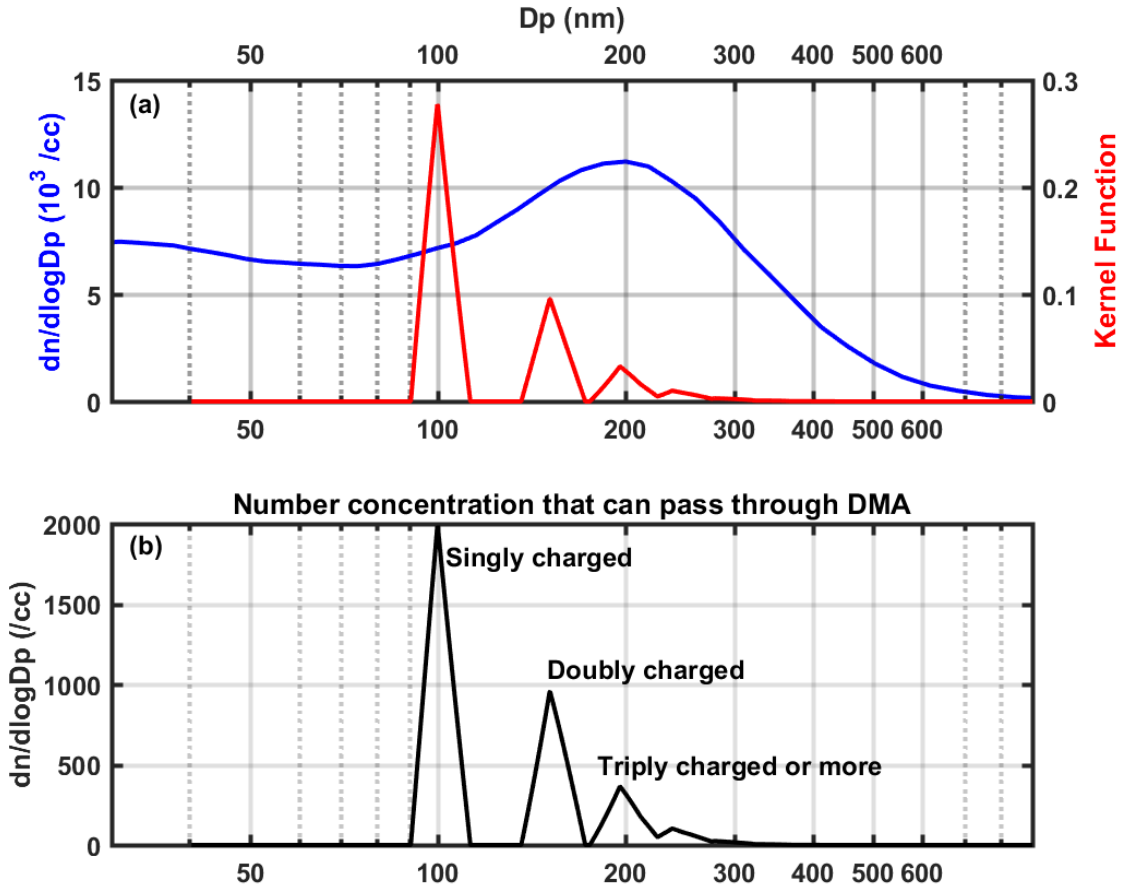

Fig.S3: (a) The blue line represents the measured particle number size distribution. The red line represents the calculated kernel function for DMA set size of 100 nm. (b) Calculated number concentration of particles that can pass through the DMA when the DMA set diameter is 100 nm. Triangle peaks from left to right represent particles with increasing number of charges.

2. The calculation and properties of f function

In the paper, f function is defined as:

$$D_p^{\nu} = f\left(D_p^*, \nu\right)$$

It describes the physical diameter of charged particles with the known electrical mobility diameter (D_p^*) and number of charges (ν).

We don't have an analytical expression for f function, but it can be calculated through two steps:

(1) Calculate the electrical mobility Z_p from D_p^* using the following equation:

$$Z_p = \frac{eC(D_p^*)}{3\pi\mu D_p^*}$$

(2) Solve the following nonlinear equation and get the best fit D_p^{ν} through the optimization method.

$$Z_p = \frac{\nu e\, C(D_p^{\nu})}{3\pi\mu\, D_p^{\nu}} = \frac{eC(D_p^*)}{3\pi\mu D_p^*}$$

It will simplify into:

$$\frac{\nu\, C(D_p^{\nu})}{D_p^{\nu}} = \frac{C(D_p^*)}{D_p^*}$$

The Cunningham slip correction C can be calculated as:

$$C = 1 + Kn[\alpha + \beta \exp(\frac{-\gamma}{Kn})]$$

where $\alpha = 1.142$, β=0.558, γ=0.999 (Allen & Raabe, 1985). Kn is the Knudsen Number of $2\lambda/D_p$, and λ is the gas mean free path with the expression of $\lambda_r(\frac{P_r}{P})(\frac{T}{T_r})(\frac{1+S/T_r}{1+S/T})$. S is the Sutherland constant of 110.4 K; T is the temperature in [K] and T_r is the reference temperature in [K].

Here, we give an example of the Cunningham slip correction C with the temperature of 25 °C and pressure of 101300 Pa. $f(D_p^*, \nu)$ curves are also shown with 1 through 4 charges respectively. It can be seen that when $\nu = 1$, the D_p^ν is equal to D_p^*. When $\nu > 1$, the D_p^ν is larger than D_p^*.

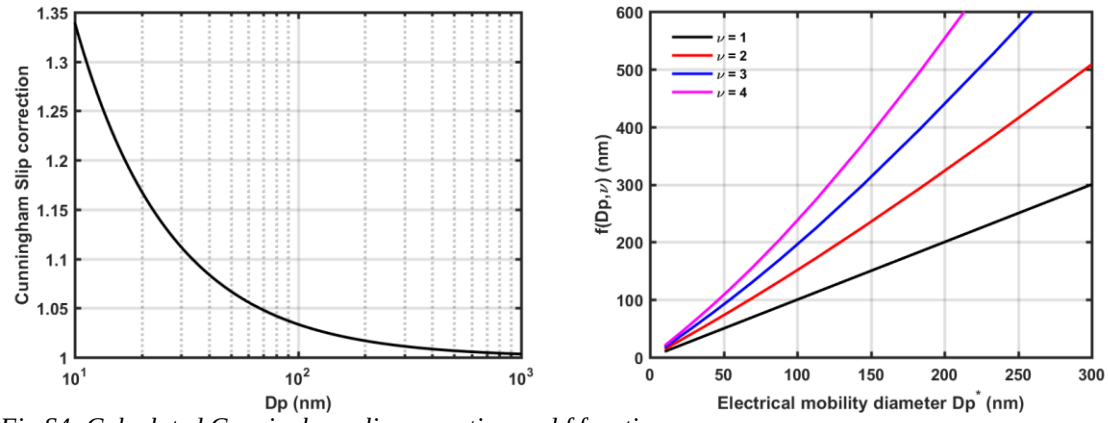

Fig.S4: Calculated Cunningham slip correction and f function curve.

Reference:

Wiedensohler, A., Lütkemeier, E., Feldpausch, M., and Helsper, C.: Investigation of the bipolar charge distribution at various gas conditions, J Aerosol Sci, 17, 413-416, https://doi.org/10.1016/0021-8502(86)90118-7, 1986.