Inversion of Differential Mobility Particle Sizer (DMPS)

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1 Governing Equations

Differential mobility analyzer (DMA) measures particle electrical mobility, Z, the migration velocity per unit of electric field strength in the low field limit, which is a monotonic function of particle size (Seinfeld and Pandis 2016):

$$Z = \frac{qC_c}{3\pi\mu D_p}, \tag{1}$$

where q is the charge number, $C_c = 1 + \frac{2\lambda}{D_p}\left[a + b\exp\left(-\frac{cD_p}{2\lambda}\right)\right]$ is the slip correction factor (a = 1.231, b = 0.4695, and c = 1.1783, Hutchins et al. 1995), λ (= 67.3 nm) and μ (= 1.8325×10^{-5} kg m^{-1} s^{-1}) are the mean free path and viscosity of air at 760 torr and 296.15 K, and D_p is the particle diameter. When DMA is operated in static mode, the relationship between the central electrical mobility Z^* and the voltage V (assume positive voltage) is:

$$Z^* = -\frac{Q_{sh} + Q_{ex}}{4\pi LV}\ln\frac{R_2}{R_1}, \tag{2}$$

where Q_{sh} and Q_{ex} are sheath and excess flow rates, L is the effective length of DMA column, R_1 and R_2 are the inner and outer diameters of the annular space inside DMA. Differential mobility particle sizer (DMPS) operates DMA in stepping mode, which leaves the transmission time long enough for the system to reach steady state at one specific voltage. Thus the Stolzerburg equation (Stolzenburg 1988) can be used to evaluate the DMPS transfer function:

$$\Omega(\zeta) = \frac{\sigma}{\sqrt{2}\beta(1-\delta)}\left\{\mathcal{E}\left(\frac{\zeta-(1-\beta)}{\sqrt{2}\sigma}\right) + \mathcal{E}\left(\frac{\zeta-(1+\beta)}{\sqrt{2}\sigma}\right) - \mathcal{E}\left(\frac{\zeta-(1-\beta\delta)}{\sqrt{2}\sigma}\right) - \mathcal{E}\left(\frac{\zeta-(1+\beta\delta)}{\sqrt{2}\sigma}\right)\right\}, \tag{3}$$

where $\mathcal{E}(x) = x\mathrm{erf}(x) + \frac{e^{-x^2}}{\sqrt{\pi}}$, $\zeta = \frac{Z}{Z^*}$, $\beta = \frac{Q_a+Q_c}{Q_{sh}+Q_{ex}}$, $\delta = \frac{Q_c-Q_a}{Q_c+Q_a}$ (Q_a and Q_c are aerosol sampling and classified flow rates), and $\sigma^2 = G \cdot \tilde{D}$, where

$$\tilde{D} = \frac{4\pi L k_B T Z^*}{qe(Q_m + Q_c)},$$

where k_B is Boltzmann constant ($= 1.38 \times 10^{-23}$ m^2 kg s^{-2} K^{-1}), T is the temperature (K), and e is the electron coulomb ($= 1.6 \times 10^{-19}$ C),

$$G = 4\frac{(1+\beta)^2}{1-\gamma}\left[I_\gamma(\gamma) + (2(1+\beta)\kappa)^{-2}\right],$$

where

$$I_\gamma(\gamma) = \frac{\frac{1}{4}(1-\gamma^2)(1-\gamma)^2 + \frac{5}{18}(1-\gamma^3)(1-\gamma)\ln\gamma + \frac{1}{12}(1-\gamma^4)\ln^2\gamma}{(1-\gamma)\left[-\frac{1}{2}(1+\gamma)\ln\gamma - (1-\gamma)\right]^2},$$

$\gamma = \left(\frac{R_1}{R_2}\right)^2$, and $\kappa = \frac{L}{R_2}\frac{1}{1-\gamma}$. Note that this G is not corrected by the radial-direction flow at the inlet and outlet of the DMA column as there is no more geometric information about Hauke-type DMA.

Except the transfer function of the DMA column, the charging probability of the particles $f(q)$, the detection efficiency of the condensation particle counter (CPC) η_{cpc}, as well as the penetration efficiency η_{tube} due to particle diffusion loss in the sampling tube have to be considered.

The charging probability distribution follows the Wiedensohler's method (Wiedensohler 1988):

$$f(q) = 10^{\left[\sum_{i=0}^{5} a(q) \cdot \left(\log \frac{D_p}{\mathrm{nm}}\right)^i\right]}, \tag{4}$$

where D_p is the particle diameter in nm, q is the charge number (> 0 means positively charged and < 0 is negatively charged), and $a_i(q)$ is from Table 1 (Wiedensohler 1988):

Table 1: Approximation coefficients $a_i(q)$

$a_i(q)$	q				
	-2	-1	0	1	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	0.5059

Eq. 4 is applied for the following ranges: 1 nm $\leq D_p \leq$ 1000 nm for $q = -1, 0, 1$; 20 nm $\leq D_p \leq$ 1000 nm for $q = -2, 2$. For particles larger than 70 nm with multiple charges ($q \geq 3$), Eq. 5 (Gunn and Woessner

1956) is used:

$$f(q) = \frac{e}{\sqrt{4\pi^2\epsilon_0 D_p k_B T}} \cdot \exp \frac{-\left[|q| + \frac{q}{|q|}\frac{2\pi\epsilon_0 D_p k_B T}{e^2}\ln\left(\frac{c_+ Z_+}{c_- Z_-}\right)\right]^2}{2\frac{2\pi\epsilon_0 D_p k_B T}{e^2}} \tag{5}$$

The detection efficiency of two types of CPC applies Eq. 6 (Mertes et al. 1995):

$$\eta_{\mathrm{cpc}} = \begin{cases} 1 - a(1 + \mathrm{e}^{(D_p - D_1)/D_2})^{-1} & D_p \geq D_0 \\ 0 & D_p < D_0 \end{cases}, \tag{6}$$

where $D_0 = D_2 \ln(a-1) + D_1$. For CPC-3010, $a = 1.7$, $D_1 = 4.3$, and $D_2 = 1.5$ (Mertes et al. 1995), while for CPC-3025, $a = 3.27$, $D_1 = 0.06$, and $D_2 = 2.22$ (by fitting Eq. 6 to Fig. 7 in Mordas et al. 2008).

The penetration efficiency η_{tube} is evaluated by Eq. 7 (Gormley and Kennedy 1948):

$$\eta_{\mathrm{tube}} = \begin{cases} 0.81905 \cdot \exp(-3.6568\mu) + 0.09753 \cdot \exp(-22.305\mu) + 0.03250 \cdot \exp(-56.961\mu) & \\ \quad +0.01544 \cdot \exp(-107.62\mu) & \mu \geq 0.02 \\ 1 - 2.5638\mu^{2/3} + 1.2\mu + 0.1767\mu^{4/3} & \mu < 0.02 \end{cases}, \tag{7}$$

where $\mu = \frac{\pi D_p L_{\mathrm{tube}}}{Q_{\mathrm{sample}}}$, L_{tube} is the length of the sampling tube, and Q_{sample} is the sampling flow rate through the sampling tube.

2 Inversion

The inversion strategy is to find the relationship between the counts (R_i, the ith measurement) by CPC, the voltage (V_i, assume positive) applied to the DMA column, and the diameter (D_p) of the particles. The continuous form of this relationship can be written as:

$$R_i = Q_a t_c \sum_{q=-1}^{q_{max}} \int_{-\infty}^{\infty} \Omega(Z(q, D_p), Z^*(V_i)) \cdot f(q, D_p) \cdot \eta_{\mathrm{cpc}}(D_p) \cdot \eta_{\mathrm{tube}}(D_p) \cdot n(\log D_p) d\log D_p,$$

where t_c is the counting time of CPC. In practice, the integral form can be written in a discrete form:

$$R_i = Q_a t_c \sum_{q=-1}^{q_{max}} \sum_{j=0}^{J} \Omega(Z(q, D_{p,j}), Z^*(V_i)) \cdot f(q, D_{p,j}) \cdot \eta_{\mathrm{cpc}}(D_{p,j}) \cdot \eta_{\mathrm{tube}}(D_{p,j}) \cdot n(\log D_{p,j}) \log\left(\frac{D_{p,j+\frac{1}{2}}}{D_{p,j-\frac{1}{2}}}\right),$$

where $D_{p,j}$ is the geometric mean diameter of the jth bin, and J is the total size bin number. This equation can be represented in matrix form:

$$\mathbf{R} = \mathbf{\Gamma} \cdot \mathbf{N} \tag{8}$$

where $\mathbf{R}$ is the array of raw data of counts, R_i ($i = 1, 2, ..., I$, I is the total measured channel number), $\mathbf{N}$ is the array of the discrete particle number size distribution (PNSD), $N_j = n(\log D_{p,j})$ ($j = 1, 2, ..., J$), and $\mathbf{\Gamma}$ is the response matrix, where

$$\Gamma_{ij} = Q_a t_c \log\left(\frac{D_{p,j+\frac{1}{2}}}{D_{p,j-\frac{1}{2}}}\right) \sum_{q=-1}^{q_{max}} \Omega(Z(q, D_{p,j}), Z^*(V_i)) \cdot f(q, D_{p,j}) \cdot \eta_{\text{cpc}}(D_{p,j}) \cdot \eta_{\text{tube}}(D_{p,j}).$$

Eqs. 2 - 7 can be used to generate the response matrix $\mathbf{\Gamma}$. Thus the PNSD $\mathbf{N}$ can be found by:

$$\mathbf{N} = \mathbf{\Gamma}^{-1} \cdot \mathbf{R} \tag{9}$$

where the total non-negative least square method (TNNLS, Merritt and Zhang 2005) is applied to find the optimized size distribution.

3 Uncertainty Propagation

Assuming there is no correlation between each measurement, the uncertainty of $\mathbf{N}$, Var($\mathbf{N}$), can be calculated from Eq. 9 in a simplified form:

$$\text{Var}(\mathbf{N}) = \mathbf{\Gamma}^{-1} \cdot \text{Var}(\mathbf{R}) \cdot (\mathbf{\Gamma}^{-1})^T, \tag{10}$$

where Var($\mathbf{N}$) and Var($\mathbf{R}$) are the variances of the size distribution and the measurement (both are matrices). Assuming flow rates and voltages introduce no errors, the error of measurement is due to the counting statistics (Poisson distribution) of CPC, so that $\sigma_{\mathbf{R}} = \sqrt{\mathbf{R}}$ and Var($\mathbf{R}$) is a diagonal matrix. Since Var($\mathbf{N}$) is usually a full matrix, we will only adopt the diagonal elements of Var($\mathbf{N}$) as the variance of $\mathbf{N}$ by ignoring the covariance terms.

The above calculation of Var($\mathbf{N}$) is from $\mathbf{R}$ to $\mathbf{N}$. If $\mathbf{N}$ is known but $\mathbf{R}$ is unknown, Eq. 8 can be used to get $\mathbf{R}$ first and then Eq. 10 to calculate Var($\mathbf{N}$) assuming Poisson distribution.

4 MATLAB Scripts

There are comments on all MATLAB scripts, so you can get a general idea about what the purpose of each script. The scripts are tested in MATLAB 2018b. Note that this script only applies to ".DAT" format from Jorma Joutsensaari. You can use the 110618 data to test the script.

DMPS_run.m is the main script to run the inversion from raw data;

DMPS_erun.m runs **DMPS_err.m** to return the uncertainty of PNSD based on PNSD;

DMPS_prep.m prepares the working conditions of DMA and CPC;

DMPS_stlzbg_TF.m returns the Stolzenburg transfer function;

DMPS_IM.m calculates the response matrix;

DMPS_inv.m applies **DMPS_TNNLS.m** to do the inversion;

DMPS_vDp.m returns the diameter at a specific voltage based on the DMA type;

DMPS_getDp.m returns the diameter with the input of electrical mobility.

References

Gormley, P. G., Kennedy, M., 1948. Diffusion from a Stream Flowing through a Cylindrical Tube. Proc. R. Ir. Acad., Sect. A 52, 163–169.

Gunn, R., Woessner, R. H., 1956. Measurements of the systematic electrification of aerosols. J. Colloid Sci. 11 (3), 254–259.

Hutchins, D. K., Harper, M. H., Felder, R. L., 1995. Slip Correction Measurements for Solid Spherical Particles by Modulated Dynamic Light Scattering. Aerosol Sci. Technol. 22 (2), 202–218.

Merritt, M., Zhang, Y., 2005. Interior-Point Gradient Method for Large-Scale Totally Nonnegative Least Squares Problems. J. Optim. Theory Appl. 126 (1), 191–202.

Mertes, S., Schröder, F., Wiedensohler, A., 1995. The Particle Detection Efficiency Curve of the TSI-3010 CPC as a Function of the Temperature Difference between Saturator and Condenser. Aerosol Sci. Technol. 23 (2), 257–261.

Mordas, G., Manninen, H. E., Petäjä, T., Aalto, P. P., Hämeri, K., Kulmala, M., 2008. On Operation of the Ultra-Fine Water-Based CPC TSI 3786 and Comparison with Other TSI Models (TSI 3776, TSI 3772, TSI 3025, TSI 3010, TSI 3007). Aerosol Sci. Technol. 42 (2), 152–158.

Seinfeld, J. H., Pandis, S. N., 2016. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 3rd Edition. John Wiley & Sons, Inc., Hoboken, New Jersey.

Stolzenburg, M. R., 1988. An Ultrafine Aerosol Size Distribution Measuring System. Ph.D. thesis, University of Minnesota.

Wiedensohler, A., 1988. An approximation of the bipolar charge distribution for particles in the submicron size range. J. Aerosol Sci. 19 (3), 387–389.