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Particle Classification by the Tandem Differential Mobility Analyzer – Particle Mass Analyzer System

by

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Abstract

Particle mass analyzers, such as the aerosol particle mass analyzer (APM) and the Couette centrifugal particle mass analyzer (CPMA), are frequently combined with a differential mobility analyzer (DMA) to measure particle mass m_p and effective density ρ_{eff} distributions of particles with a specific electrical mobility diameter d_m. Combinations of these instruments, which are referred as the DMA-APM or DMA-CPMA system, are also used to quantify the fractal dimension D_f of non-spherical particles, as well as to eliminate multiply charged particles. This study investigates the transfer functions of these setups, focusing especially on the DMA-APM system. The transfer function of the DMA-APM system was derived by multiplying the transfer functions of the DMA and APM. The APM transfer function can be calculated using either the uniform or parabolic flow models. The uniform flow model provides an analytical function, while the parabolic flow model is more accurate. The resulting DMA-APM transfer functions were plotted on $log(m_p)$- $log(d_p)$ space. A theoretical analysis of the DMA-APM transfer function demonstrated that the resolution of the setup is maintained when the rotation speed ω of the APM is scanned to measure distribution. In addition, an equation was derived to numerically calculate the minimum values of the APM resolution parameter λ_c for eliminating multiply charged particles.

1. Introduction

Particle classification is a key technique for investigating aerosol particles (Hinds 1999; McMurry 2000). Particle classification instruments, such as the differential mobility analyzer (DMA), have been widely employed throughout all areas of aerosol research (Knutson and Whitby 1975; Stolzenburg and McMurry 2008). Most of these instruments, including the DMA, classify particles based on diameter d_p, using the dynamics of the particles as classification principles (Hinds 1999).

The particle mass analyzer (PMA), which includes both the aerosol particle mass analyzer (APM) and Couette centrifugal particle mass analyzer (CPMA), is becoming a popular tool to classify particle mass (Ehara et al. 1996; Olfert 2005; Tajima et al. 2011). The concept of the APM was firstly introduced by Ehara et al. (1996), and the CPMA was proposed by Olfert and Collings (2005). The PMA consists of two rotating cylinders, and a voltage is applied in between them. This design allows the PMA to classify particles based on the balance between the centrifugal and electrostatic forces. Since centrifugal force is proportional to particle mass m_p, the PMA is capable of classifying particles based on their mass. In the case of the APM, two cylinders rotate at the same angular velocity for accurate mass classification (Ehara et al. 1996). On the other hand, the rotation speeds of the two cylinders are different for the CPMA, which allows the instrument to have a higher particle transmission than the APM (Olfert 2005).

In many cases, the PMA is combined with the DMA in tandem (McMurry et al. 2002; Kuwata et al. 2009; Cross et al. 2010). Examples of these setups include the DMA-APM, DMA-CPMA, and APM-scanning mobility particle sizer (SMPS) systems (McMurry et al. 2002;

Malloy et al. 2009; Cross et al. 2010). In these setups, particles are classified by both electrical mobility diameter d_m and m_p, which allows for the quantification of important physical parameters, such as effective density ρ_{eff}, dynamic shape factor, and mass-mobility exponent D_f (Park et al. 2003; Kuwata and Kondo 2009; Zangmeister et al. 2014). The combination of these two techniques is useful in eliminating multiply charged particles because the classification regions for multiple charged particles of the DMA and PMA do not overlap (Pagels et al. 2009; Shiraiwa et al. 2010).

However, the instrumental responses of these setups, which are useful in optimizing experimental conditions, have not been evaluated theoretically. This study develops the transfer functions of the DMA-PMA setup by focusing on the APM. Implications of the theoretically derived transfer functions on actual operation will also be discussed.

2. Mass-mobility relationship

2.1 Effective density and mass-mobility exponent

The relationship between m_p, d_m, and ρ_{eff} is shown by the following equation (McMurry et al. 2002; DeCarlo et al. 2004).

$$m_p = \frac{1}{6}\pi\rho_{eff} d_m^3 \qquad (1)$$

The equation is rewritten as follows in the logarithmic scale

$$\log(m_p) = \log\left(\frac{1}{6}\pi\right) + \log(\rho_{eff}) + 3\log(d_m) \qquad (2).$$

Equation 2 has the advantage of considering particle classification by both m_p and d_m since the relationship is linear in the $log(d_m)$- $log(m_p)$ space. This equation can be equally applied to both spherical and non-spherical particles because ρ_{eff} depends both on the material density and morphology of the particles (Park et al. 2003; Kuwata and Kondo 2009). Figure 11a plots the relationship between m_p, d_m, and ρ_{eff} in the $log(d_m)$- $log(m_p)$ space. This space is convenient for deriving the DMA-PMA transfer function because the DMA and PMA classify particles by d_m and m_p, respectively.

The $log(d_m)$- $log(m_p)$ relationship can also be represented using other metrics, such as the mass-mobility exponent (D_f), which is calculated by the following equation (DeCarlo et al. 2004; Cross et al. 2010; Sorensen 2011; Zangmeister et al. 2014).

$$m_p = \rho_f d_m^{D_f} \Leftrightarrow \log(m_p) = \log(\rho_f) + D_f \log(d_m) \qquad (3)$$

Completely spherical particles have $D_f = 3$, while the value is smaller for non-spherical particles. Although the definition of D_f is similar to that of the fractal dimension, these two parameters are not equivalent (Sorensen 2011). Examples of $log(d_m)$- $log(m_p)$ relationships for different values of D_f are shown in figure 11b. As indicated by the logarithmic form of equation 3, D_f corresponds to the value of the slope in the $log(d_m)$- $log(m_p)$ space. This metric is especially useful for characterizing the structure of aggregate particles, such as soot (Park et al. 2003; DeCarlo et al. 2004; Zangmeister et al. 2014). The values of ρ_f and $1/6\pi\rho_{eff}$ are equivalent when D_f is equal to three (equations 2 and 3), meaning that equation 3 may be considered as a generalized form of equations 2.

2.2 Particle population on the *log*(d_m)- *log*(m_p) space

Particles can populate on the $log(d_m)$ –$log(m_p)$ space in different ways, depending on their morphology and mixing state. Three different types of particle populations are considered here, namely spherical (or nearly spherical) particles with a constant value of ρ_{eff}, aggregated non-spherical particles with a certain value of D_f, and a mixture of spherical and non-spherical particles with a range of ρ_{eff} (Figure 22).

Examples of spherical/nearly spherical particles with a constant value of ρ_{eff} include oil droplets, ammonium sulfate, and sodium chloride (Kuwata and Kondo 2009; Tajima et al. 2011; Tajima et al. 2013). In these cases, particles populate only on a line in the $log(d_m)$ –$log(m_p)$ space, which has an intercept of $\log\left(\frac{1}{6}\pi\right)+\log\left(\rho_{eff}\right)$ and a slope of three (equation 2). The intercept is dependent on both the particle morphology and chemical composition, since these parameters determine ρ_{eff}. An example for this case is shown in figure 22a, in which ρ_{eff} is assumed to be 1000 kg m^{-3}. In this case, the particles can only populate on the black solid line in the figure.

Figure 22b presents an example of the second case, which corresponds to a constant value of D_f. As discussed in section 2.12.1, the value of the slope in the $log(d_m)$ –$log(m_p)$ space is smaller than three for aggregated particles, such as soot (Park et al. 2003; Cross et al. 2010; Zangmeister et al. 2014). In figure 22b, a mass-mobility relationship measured by Park et al. (2003) is shown as an example. The particles populate only on the black solid line. The line is not parallel to the isodensity lines because the mass-mobility exponent is smaller than three. As a result, ρ_{eff} is smaller for larger particles (Park et al. 2003).

Figure 22c illustrates an example of an area for particle population for a mixture of spherical and non-spherical particles with a range of ρ_{eff} (i.e., external mixture of various types of particles). For example, ρ_{eff} of atmospheric sub-micron particles can have broad distributions because many different types of particles, such as non-spherical soot particles, primary organic aerosol particles, and secondary particles exist in the atmosphere (McMurry et al. 2002; Kuwata and Kondo 2009). The upper limit of ρ_{eff} is determined by the material density of the heaviest compound in the particles, and the lower limit of ρ_{eff} depends on both the material density of the lightest species and the particle morphology.

3. Theory

3.1. Differential mobility analyzer (DMA) transfer function

The DMA classifies particles based on electrical mobility $Z_p\left(d_{m,q}\right)$, which is defined by the following equation (Knutson and Whitby 1975; Stolzenburg and McMurry 2008)

$$Z_p\left(d_{m,q}\right) = \frac{qeC_c\left(d_{m,q}\right)}{3\pi\mu d_{m,q}} \quad (4)$$

where q is the particle charge, e is the elemental charge, and μ is the viscosity of a fluid (air). The suffix d_m (i.e., q) indicates the number of charges on a particle. $C_c(d_{m,q})$ is the slip correction factor, which is calculated using the mean free path of air l as

$C_c\left(d_{m,q}\right) = 1 + \left(2l/d_{m,q}\right)\left[1.142 + 0.558\exp\left(-0.999 d_{m,q}/\left(2l\right)\right)\right]$ (Allen and Raabe 1985). In a certain DMA operating condition, the mode mobility of the classified particles Z_p^* is calculated as (Knutson and Whitby 1975; Stolzenburg and McMurry 2008)

$$Z_p^* = \frac{Q_{sh}\ln\left(r_{2\_DMA}/r_{1\_DMA}\right)}{2\pi V_{DMA} L_{DMA}} \quad (5).$$

In this equation, r_{1_DMA} and r_{2_DMA} denote the inner and outer radii of the DMA, and L_{DMA} is the length of the DMA. V_{DMA} stands for the DMA voltage, and Q_{sh} is the sheath flow rate. Q_{sh} is typically controlled as equal to the excess flow rate of DMA (Wiedensohler et al. 2012). This condition is assumed when deriving equation 5 and is employed throughout this study.

The DMA transfer function (Ω) is calculated by the following equation when particle diffusion is negligible (Knutson and Whitby 1975; Stolzenburg and McMurry 2008).

$$\Omega\left(Z_p,\beta\right)=\frac{1}{2\beta}\left[\left|Z_p\left(d_{m,q}\right)-(1+\beta)\right|+\left|Z_p\left(d_{m,q}\right)-(1-\beta)\right|-2\left|Z_p\left(d_{m,q}\right)-1\right|\right] \quad (6)$$

In equation 6, $Z_p\left(d_{m,q}\right)=Z_p\left(d_{m,q}\right)/Z_p^*$ and β represent the ratio of the sample and the sheath flow rates. An example of the non-diffusing DMA transfer function is shown in figure 33a. Although Ω is symmetric in the electrical mobility space, the shape of the function is skewed in the diameter space because of $C_c(d_m)$. The minimum, central, and maximum electrical mobility diameters for particle classification are denoted as $d_{min,q}$, $d_{c,q}$ and $d_{max,q}$ (figure 33a). These values are calculated by the following equations

$$Z_p\left(d_{\min,q}\right)=(1+\beta) \quad (7)$$

$$Z_p\left(d_{c,q}\right)=1 \quad (8)$$

$$Z_p\left(d_{\max,q}\right)=(1-\beta) \quad (9).$$

As shown in figure 33a, Ω is separated into three regions by $d_{min,q}$, $d_{c,q}$ and $d_{max,q}$. In regions 1 ($d_m < d_{min,q}$) and 3 ($d_m > d_{max,q}$), no particles are classified. The particles in region 3 ($d_{min,q} \leq d_m \leq d_{max,q}$) can pass through the DMA.

3.2. APM transfer function

This section briefly introduces the transfer function of the APM, which was derived by Ehara et al. (1996). The APM transfer function could be considered a special case of the CPMA transfer function, as discussed by Olfert (2005). The APM transfer function has an analytical solution, which facilitates the theoretical analysis of the DMA-APM response (section 3.3~~3.3~~).

The APM classifies particles based on the balance between the centrifugal and electrostatic forces, which is expressed by the following equation

$$s_c = \frac{m_{c,q}}{qe} = \frac{V_{APM}}{r_{c\_APM}{}^2 \varpi^2 \ln\left(r_{2\_APM} / r_{1\_APM}\right)}$$
$$\Leftrightarrow\ m_{c,q}\varpi^2 = V_{APM} \frac{qe}{r_{c\_APM}{}^2 \ln\left(r_{2\_APM} / r_{1\_APM}\right)} \qquad (10).$$

Specific mass *s*, which is calculated as *m*/*qe*, is a useful parameter for deriving the APM transfer function. Suffix *c* indicates the central values of *m* and *s* of particles classified by the APM, and suffix *q* corresponds to the number of particle charges. r_{1_APM}, r_{c_APM}, and r_{2_APM} denote the inner, center, and outer radii of the APM operating space, respectively. V_{APM} and ω are the voltage and rotation speed of the APM. Equation 10 shows that both ω and V_{APM} can be adjusted to select s_c or m_c. Ehara et al. (1996) has further demonstrated that the classification performance parameter λ of the APM, which is defined by equation 11, plays a critical role in determining the APM transfer function (Tajima et al. 2011).

$$\lambda = \frac{2\tau\varpi^2 L_{APM}}{\bar{v}} = \frac{2 m_p \varpi^2 Z_p\left(d_{m,q}\right) L_{APM}\, \pi \left(r_{2\_APM}^2 - r_{1\_APM}^2\right)}{qeQ_{APM}} \qquad (11).$$

This parameter is calculated as a function of relaxation time τ, ω, length of the APM operating space L_{APM}, and the average flow velocity $\bar{v}$. λ depends on m_p, Z_p, and the APM flow rate Q_{APM}, since τ and $\bar{v}$ are calculated as $\tau = m_p C_c(d_{m,q})/(3\pi\mu d_{m,q}) = m_p Z_p(d_{m,q})/(qe)$ (Seinfeld and Pandis 2006) and $\bar{v} = Q_{APM}/\left(\pi\left(r_{2\_APM}^2 - r_{1\_APM}^2\right)\right)$, respectively. λ calculated for m_c is specifically named λ_c. The APM transfer function is conserved for a specific value of λ_c when it is plotted as a function of normalized specific mass (s^*/s) (Ehara et al. 1996).

The APM transfer function can be calculated either by the uniform or parabolic flow model (Ehara et al. 1996). The uniform flow model has an analytical solution, which is advantageous in theoretical analyses. On the other hand, the parabolic flow model provides a more accurate form of the APM transfer function. Figure 3b shows APM transfer functions that were calculated using these two models.

Uniform flow model

The uniform flow APM transfer function is separated into five regions by four parameters ($m_1^{\pm}$ and $m_2^{\pm}$). The uniform flow APM transfer function has a maximum value of $\exp(-\lambda_c)$ at $m_2^- \le m_p \le m_2^+$ (region C). It monotonically increases/decreases in the regions of $m_1^- \le m_p \le m_2^-$ (region B)/ $m_2^+ \le m_p \le m_1^+$ (region D), respectively. The transfer function is zero for $m_p \le m_1^-$ (region A) and $m_1^+ < m_p$ (Region E). Table 1 summarizes the functional form for the APM transfer function.

The values of $m_1^{\pm}$ and $m_2^{\pm}$ are calculated using the following equations.

$$\frac{s_2^{\pm}}{s_c} = \frac{m_{2,q}^{\pm}}{m_{c,q}} = \frac{1}{\left(1 \mp \delta / r_{c\_APM}\right)^2} \quad (12)$$

$$\Leftrightarrow \ln\left(s_2^{\pm}\right) - \ln\left(s_c\right) = \ln\left(m_{2,q}^{\pm}\right) - \ln\left(m_{c,q}\right) = -2\ln\left(1 \mp \delta / r_{c\_APM}\right)$$

$$\frac{s_1^{\pm}}{s_c} = \frac{m_{1,q}^{\pm}}{m_{c,q}} = \frac{1}{\left[1 \mp \left(\delta / r_{c\_APM}\right)\coth\left(\lambda_c / 2\right)\right]^2} \quad (13)$$

$$\Leftrightarrow \ln\left(s_1^{\pm}\right) - \ln\left(s_c\right) = \ln\left(m_{1,q}^{\pm}\right) - \ln\left(m_{c,q}\right) = -2\ln\left(1 \mp \left(\delta / r_{c\_APM}\right)\coth\left(\lambda_c / 2\right)\right)$$

In these equations, δ is calculated as $\delta = \left(r_{2\_APM} - r_{1\_APM}\right)/2$. The ratios of $m_{2,q}^{\pm} / m_{c,q}$ are determined by the instrumental design, and the $m_{1,q}^{\pm} / m_{c,q}$ ratios depend both on the instrumental design and operating conditions, which is characterized by λ_c. Neither the APM transfer function nor the mass resolution ratio changed for a specific instrumental design as long as λ_c is conserved.

Figure 4 plots the values of λ_c, $m_1^{\pm}$ and $m_2^{\pm}$ in the *log* (m_p)- *log* (d_p) space. The value of λ_c is smaller for larger particles because Z_p is smaller (equation 11). This diameter dependence leads to a broader APM resolution for larger particles, which also affects the DMA-APM transfer function.

Parabolic flow model

A detailed description of the parabolic flow model APM transfer function is provided in the Supplemental Information. For this model, $m_{2,q}^{\pm}$ are calculated using equation 12, while numerical calculations are required to obtain $m_{1,q}^{\pm}$. Numerical computation is also needed to acquire the APM transfer function using the parabolic flow model.

3.3. Transfer function of the DMA-APM system

The transfer function of the tandem DMA-APM system (*Φ*) is calculated by overlaying the transfer functions of both the DMA and APM (Radney et al. 2013).

$$\Phi\left(m_{p,q}, d_{m,q}\right) = \Omega\left(m_{p,q}, d_{m,q}\right)\Psi\left(d_{m,q}\right) \tag{14}$$

This equation can also be employed for the APM-DMA system because $\Omega\left(m_{p,q}, d_{m,q}\right)\Psi\left(d_{m,q}\right)$ is equivalent to $\Psi\left(d_{m,q}\right)\Omega\left(m_{p,q}, d_{m,q}\right)$ (Malloy et al. 2009). The properties of this equation are examined in the following sections. The DMA-APM transfer function can be calculated for seven different regions in the *log*(d_m)- *log*(m_p) space, as shown in figure 55 and table 33.

Region 1 ($d_m < d_{min}$)

This region corresponds to region 1 in the DMA transfer function, meaning that no particles in this region can pass through the DMA-APM system (i.e., $\Phi\left(m_{p,q}, d_{m,q}\right) = 0$).

Region 2 ($d_{min} \le d_m \le d_{max}$)

The particles in this size range are classified by the DMA. Particle transmittance in this region depends both on the DMA and APM transfer functions.

Region 2A **(**$d_{min} \le d_m \le d_{max}$**,** $m_p < m_1^-$**)**

This region corresponds to region A in the APM transfer function, meaning that no particles in this range can pass through the APM.

Region 2B **(**$d_{min} \le d_m \le d_{max}$**,** $m_1^- \le m_p < m_2^-$**)**

This range of m_p conforms to region B of the APM transfer function. Since both the DMA and APM transfer functions are positive in this range, the DMA-APM transfer function is positive in this region.

Region 2C **(**$d_{min} \le d_m \le d_{max}$**,** $m_2^- \le m_p < m_2^+$**)**

In this area, region C of the APM transfer function overlaps with region 2 in the DMA transfer function. Region C has the highest particle transmittance in the APM transfer function, meaning the DMA-APM transfer function has the highest value in this region. The maximum value is found at $\{d_m, m_p\}=\{d_c, m_c\}$. The corresponding value of Φ is $\exp(-\lambda_c)$ when the uniform flow model is employed to calculate the APM transfer function.

Region 2D **(**$d_{min} \le d_m \le d_{max}$**,** $m_2^+ \le m_p < m_1^+$**)**

The particles in this region pass through the APM, meaning that the DMA-APM transfer function is positive.

Region 2E **(**$d_{min} \le d_m \le d_{max}, m_1^+ \le m_p$**)**

The particles in this region cannot pass through the APM. Therefore, the DMA-APM transfer function is zero in this region.

Region 3 ($d_m < d_{max}$**)**

This region corresponds to region 3 in the DMA transfer function. No particles in this region can pass through the DMA-APM system (i.e., $\Phi\left(m_{p,q}, d_{m,q}\right) = 0$).

Examples of the DMA-APM transfer functions are shown in figure 66. An example of the uniform flow model for the APM is shown in figure 66a, and figure 66b demonstrates a result for the parabolic flow model. These two transfer functions calculated using two different models resemble each other, since the APM transfer functions for the corresponding operating conditions are similar (figure 33b). In the following section, the characteristics of the DMA-APM transfer function are mainly analyzed using the uniform flow APM model because the analytical solution for the model facilitates detailed analyses.

3.4. Resolution of the DMA-APM system

The DMA-APM transfer function is surrounded by four points, which are denoted as P_1 ~P_4 in figure 66. These points are located at $P_1\{d_{min}, m_1^+\}$, $P_2\{d_{max}, m_1^+\}$, $P_3\ \{d_{min}, m_1^-\}$, and $P_4\{d_{max}, m_1^-\}$. The maximum mass of the classified particle m_{max} is observed at P_2, while P_4 corresponds to the minimum value of particle mass m_{min}. Both of those two points are located at

d_{max} because λ_c is smaller for larger particles, which have smaller electrical mobility (equation 11). The masses at these points are calculated by equation 13, using d_{max} in calculating λ_c.

$$\frac{m_{\max,q}}{m_{c,q}} = \frac{1}{\left[1-\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{\max,q}}/2\right)\right]^2}$$
$$\frac{m_{\min,q}}{m_{c,q}} = \frac{1}{\left[1+\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{\max,q}}/2\right)\right]^2} \qquad (15).$$

This equation demonstrates that the mass resolution of the DMA-APM system is determined by the instrumental design of the APM and λ_c at d_{max}.

Points P_1 and P_4 correspond to the minimum and maximum values of ρ_{eff} ($\rho_{eff\_min,q}$, and $\rho_{eff\_max,q}$)

$$\rho_{eff\_\min,q} = \frac{6m^{-}_{1,d_{\max,q}}}{\pi d^3_{\max,q}} = \frac{6m_{c,q}}{\pi d^3_{\max,q}} \frac{1}{\left[1+\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{\max,q}}/2\right)\right]^2}$$

$$\rho_{eff\_\max,q} = \frac{6m^{+}_{1,d_{\min,q}}}{\pi d^3_{\min,q}} = \frac{6m_{c,q}}{\pi d^3_{\min,q}} \frac{1}{\left[1-\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{\min,q}}/2\right)\right]^2} \qquad (16)$$

These equations are rewritten as

$$\frac{\rho_{eff\_\max,q}}{\rho_{eff\_\min,q}} = \frac{\rho_{eff\_c,d_{\min,q}}}{\rho_{eff\_c,d_{\max,q}}} \left(\frac{\left[1+\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{\max,q}}/2\right)\right]}{\left[1-\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{\min,q}}/2\right)\right]} \right)^2 \qquad (17).$$

In this equation, $\rho_{eff\_c,d_{p,q}}$ corresponds to the ρ_{eff} of the particles with $\{d_p, m_p\} = \{d_{p,q}, m_{c,q}\}$. These equations demonstrate that the density resolution of the DMA-APM system is determined by both the DMA and APM resolutions. The density ratio of points A and B in figure 66, which is calculated as $\rho_{eff\_c,d_{min,q}} / \rho_{eff\_c,d_{max,q}}$, corresponds to the density resolution derived solely from the DMA resolution. The rest of the term in equation 17 $\left(\left[1+\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{max,q}} / 2\right)\right] / \left[1-\left(\delta / r_{c\_APM}\right)\coth\left(\lambda_{c,d_{min,q}} / 2\right)\right]\right)^2$ matches the APM resolution.

3.5. Apparent diameter resolution of the DMA-APM system

The m_p resolution of the APM can be converted to d_m resolution when the particles have a uniform mass-mobility relationship (i.e., figures 22a and 22b), since m_p can be converted easily into d_m when the relationship between these two parameters is uniquely known. In such cases, the values of d_m, which correspond to the minimum (d_{APMmin}) and maximum (d_{APMmax}) values of m_p classified by the APM, can be calculated using equation 3 as

$$d_{APM\min} = \left(\frac{m^{-}_{2,d_{APM\min}}}{\rho_f}\right)^{\frac{1}{D_f}} \quad (18)$$

and

$$d_{APM\max} = \left(\frac{m^{+}_{2,d_{APM\max}}}{\rho_f}\right)^{\frac{1}{D_f}} \quad (19).$$

Depending on the design and operating condition of the APM, d_{APMmin} may be larger than d_{min}, and d_{APMmax} may be smaller than d_{max}. In this case, the apparent diameter resolution of the DMA-APM system is determined by the APM rather than the DMA.

$$\frac{d_{max}}{d_{min}} > \frac{d_{APM\ max}}{d_{APM\ min}} = \left(\frac{m^{+}_{1,d_{APM\ max}}}{m^{-}_{1,d_{APM\ min}}} \right)^{\frac{1}{D_f}} \quad (20).$$

Figure 7~~7~~ provides an example. The particles populate on regions 2A and 2E in figure 7~~7~~ (figure 5~~5~~). In these regions, the particles cannot pass through the DMA-APM system even though the DMA selects them because these areas are located outside of the APM classification region. This situation occurs when the slope of the line connecting P_2 and P_3 is smaller than D_f (figure 7~~7~~). This condition is written as

$$\frac{\log\left(m^{+}_{2,d_{max}}\right) - \log\left(m^{-}_{2,d_{min}}\right)}{\log\left(d_{max}\right) - \log\left(d_{min}\right)} < D_f$$
$$or \quad (21).$$
$$\frac{d_{max}}{d_{min}} > \left(\frac{m^{+}_{1,d_{max}}}{m^{-}_{1,d_{min}}} \right)^{\frac{1}{D_f}}$$

It should be noted that even when the d_m resolution of the DMA-APM system appears to be controlled by the APM, the area for particle classification by the DMA-APM in the $log(d_m)$-$log(m_p)$ at a certain operating condition is still regulated by the DMA and APM (figure 7~~7~~). Although the apparent d_m resolution of the DMA-APM system can be higher than the DMA resolution, the actual d_m resolution of the DMA-APM system is still controlled by the DMA.

When the particles have a broad distribution in the $log(d_m)$- $log(m_p)$ space (figure 2~~2~~c), the diameter resolution of the DMA-APM system is predominantly determined by the DMA resolution (d_{min} and d_{max}) because the particles distribute across the entire areas of 2A~2E.

4. Implication for instrumental operation

4.1. Operating the DMA-APM to investigate d_m-m_p relationships

The DMA-APM transfer function would ideally be maintained as a constant shape while scanning the $log(d_m)$- $log(m_p)$ space in order to minimize skewness induced by the instrument on measurements (Lall et al. 2009). In most of the DMA-APM operations, one operating parameter of either the DMA or the APM (e.g., V_{DMA}, V_{APM}, or ω) is scanned to measure the particle population in the $log(d_m)$- $log(m_p)$ space. This is done in order to obtain the values or distributions of ρ_{eff} and D_f (McMurry et al. 2002; Park et al. 2003; Malloy et al. 2009; Zangmeister et al. 2014). An example of DMA voltage scanning is the APM-SMPS measurement (Malloy et al. 2009). The shape of the DMA-APM transfer function cannot be maintained in this case because (1) the DMA transfer function continuously changes in the $log(d_m)$ space due to C_c (d_p) and (2) λ_c also changes with d_m (equation 11). The inversion of the DMA-APM data, which incorporates the DMA-APM transfer function, would be required to resolve this issue.

In many applications of the DMA-APM system, an operating parameter of the APM is scanned to measure the particle population of $log(d_m)$- $log(m_p)$ while the DMA operating condition is fixed (McMurry et al. 2002; Radney et al. 2013; Zangmeister et al. 2014). An advantage of this scanning method is that the DMA transfer function is maintained throughout the operation, which allows us to focus on the APM transfer function. The resolution and the shape of the APM transfer function should be kept constant during scanning, which is satisfied by keeping λ_c constant (table 1 and equations 15 and 17). Equations 15 and 17 suggest that the resolution stays the same in the logarithmic scale as long as λ_c for a certain diameter is kept constant. λ_c is determined by several parameters, including Q_{APM}, the dimensions of the APM, Z_p (d_p), and $m_c\omega^2$ (equation 11). Q_{APM} is not scanned for most of the APM operations, and the dimensions of the APM, such as L_{APM}, cannot be changed during operation. In the case of the DMA-APM system, Z_p (d_p) can also be considered a constant because the particles are already prescribed by the DMA.

A constant λ_c value can be achieved by keeping $m\omega^2$ constant. Particle classification by the APM is controlled by both V_{APM} and ω (equation 10), meaning that m_c can be scanned by changing one of them. Equation 10 demonstrates that $m_c\omega^2$ is preserved as long as V_{APM} and the physical dimensions of the APM are maintained, meaning that λ_c does not vary when ω is changed to scan m_c for a fixed value of V_{APM}.

Figure 88 compares the DMA-APM transfer functions for the ω scan (fixed V_{APM}) and the V_{APM} scan (fixed ω). The shape of the DMA-APM transfer function does not change during the ω scan in the *log* (d_p)- *log* (m_p) space. On the other hand, the DMA-APM transfer function is narrower for higher values of m_c when V_{APM} is scanned because λ_c is proportional to m_c (equation 11). In conclusion, the ω scan has the advantage of maintaining the DMA-APM resolution compared with the V_{APM} scan.

A caveat for the above discussion is that λ_c depends on $Z_p(d_m)$, even though the range of $Z_p(d_m)$ is narrow following particle classification by the DMA (figure 77). For this reason, the DMA-APM transfer function does not have a rectangular shape in the *log*(d_m)- *log*(m_p) space. This d_m dependence in the DMA-APM transfer function needs to be carefully considered when interpreting data, especially when a particle population has a uniform mass-mobility relationship (i.e., figures 22a and 22b). When m_c is close to $\frac{1}{6}\pi\rho_{eff}d_{\min}^{3}$ or $\rho_f d_{\min}^{D_f}$, the d_m of the particles classified by the DMA-APM system is close to d_{min}. On the other hand, the d_m of the classified particles is close to d_{max} when m_c is around $\frac{1}{6}\pi\rho_{eff}d_{\max}^{3}$ or $\rho_f d_{\max}^{D_f}$. Even if V_{APM} is fixed when scanning m_c to maintain λ_c for a certain diameter, the λ_c corresponding to the classified particles by the DMA-APM system could change due to the fact that the d_m of the classified particles depends both on the DMA and the APM. For this reason, the distribution of m_p or ρ_{eff} , as measured by the DMA-APM system, may not be symmetric, even if V_{APM} is fixed as a constant. Ideally, the V_{APM} should be slightly adjusted when scanning m_c so that λ_c is maintained at a certain constant value for the classified particles. However, such an operation requires prior knowledge regarding the mass-mobility exponent.

4.2. Operating the DMA-APM system to remove multiply charge particles

The DMA-APM system is used in some applications as a tool to eliminate multiply charged particles (Pagels et al. 2009; Shiraiwa et al. 2010). In these cases, the DMA-APM transfer function should have a high particle transmittance in region 2C in order to effectively classify the particles of interest. The DMA-APM transfer function, on other other hand, must be sufficiently narrow in order to remove multiply charged particles. However, these two conditions contradict each other. The maximum value of the DMA-APM transfer function is higher for smaller values of λ_c (Table 33), while the resolution of the DMA-APM transfer function is narrower for higher values of λ_c (section 3.3~~3.3~~). A method to obtain the maximum value of λ_c that satisfies these conditions is discussed in the following section, assuming that particle population has a narrow distribution of mass-mobility relationship (figures 2~~2~~a and 2~~2~~b).

When operating the DMA-APM system to remove multiply charged particles, the central part of the DMA-APM transfer function, which is located at $\{d_p, m_p\} = \{d_{c,+1}, m_{c,+1}\}$, is adjusted to classify the desired particles. The ρ_{eff} corresponding to this point is denoted as $\rho_{eff\_c,+1}$. The maximum value of ρ_{eff} for multiply charged particles, which is located at P_1 for +2 charge particles ($P_{1,+2}$), must be smaller than $\rho_{eff\_c,+1}$ to completely remove the multiply charged particles (Figure 9~~9~~). These conditions lead to the following equation

$$\rho_c\left(d_{c,+1}\right) > \rho_{eff\_\max,+2}$$
$$\Rightarrow \coth\left(\lambda_c\left(d_{\min,+2}\right)/2\right) < \frac{r_{c\_APM}}{\delta}\left(1-\sqrt{2\left(\frac{d_{c,+1}}{d_{\min,+2}}\right)^3}\right) \quad (22).$$

This equation determines the minimum value of λ_c to eliminate multiply charged particles, since *coth* (λ_c /2) monotonically decreases for higher values of λ_c.

Figure 9~~9~~ shows an example of a condition that satisfies equation 22. The uniform flow model was used for figure 9~~9~~a, while the parabolic flow model was employed to calculate the APM transfer function in figure 9~~9~~b. In both figures, $\rho_c\left(d_{c,+1}\right)$ is equal to 930 kg m^{-3}, and $d_{c,+1}$ is set at 100 nm. In this case, the doubly charged particles with 930 kg m^{-3} of ρ_{eff} cannot pass through the DMA-APM system because the classification region for +2 particles ($d_{c,+2}$ = 150.9 nm) does not overlap with the area for particle population, which is on the line of of ρ_{eff} = 930 kg m^{-3}.

This condition can be further generalized to non-spherical fractal particles. In that case, $m^{+}_{1,+2}$ of the APM at $d_{min,+2}$ must be smaller than the mass of particles of interest, which equals $\rho_f d_{\min,+2}^{D_f}$ (equation 3)

$$
\begin{aligned}
& m^{+}_{1,+2}\left(d_{\min,+2}\right) < \rho_f d_{\min,+2}^{D_f} \\
& \Rightarrow \coth\left(\lambda_c\left(d_{\min,+2}\right)/2\right) < \frac{r_{c\_APM}}{\delta}\left(1-\sqrt{2\left(\frac{d_{c,+1}}{d_{\min,+2}}\right)^{D_f}}\right)
\end{aligned}
\qquad (23)
$$

where $m^{+}_{2,+2}$ is assumed to be smaller than $\rho_f d_{\min,+2}^{D_f}$ in deriving this equation. Since $m^{+}_{1,+2}$ is always larger than $m^{+}_{2,+2}$ (equation 13), no solution is available for equation 23 when this assumption is invalid. Equation 23 is more general than equation 22 because these two equations are equivalent for spherical particles (D_f = 3). This equation will be useful for experiments where generation of monodisperse fractal particles is needed, such as a study on the optical properties of soot particles.

Interestingly, λ_c for single and multiple charged particles are the same for the DMA-APM system (equation 11) because their Z_p values are the same as long as they are classified by the same DMA (equations 4 and 5). Similarly, m_c/qe does not depend on the particle charge (equation 10). An implication of this interesting fact is that the minimum value of λ_c does not depend on m_p or ρ_{eff}, as long as d_c and d_{min} are the same.

This phenomenon is also useful in considering the elimination of highly (q > 2) charged particles. As evident in figure 9, the condition to eliminate multiply charged particles requires the slope of a line connecting $\{d_p, m_p\}$ =$\{\{d_{c,+1}, m_{c,+1}\},\{d_{c,+2}, m_{c,+2}\}\}$ to be smaller than D_f in the *log*

(d_p)- *log* (m_p) space. The distance between {$d_{c,+n}$, $m_{c,+n}$} ($n \geq 3$) and the line for particle population is further than that for doubly charged particles, while λ_c does not depend on the particle charge (figure S1). The implication is that highly charged particles ($q \geq 3$) are always removed by the DMA-APM system when it is being used to eliminate doubly charged particles from the system.

5. Conclusions

The transfer function of the DMA-APM system was developed by overlapping that of the DMA and the APM, and mapped on the *log*(m_p)- *log*(d_p) space. The APM transfer function was calculated using either the uniform or parabolic flow models. The uniform flow model has an analytical expression that is favorable for investigating the instrumental response theoretically. On the other hand, the parabolic flow model provides the APM transfer function more accurately. The m_p and ρ_{eff} resolutions of the DMA-APM system were theoretically investigated using the derived transfer function. The resolution of the DMA-APM system was also evaluated theoretically.

The DMA-APM system is frequently used to measure the ρ_{eff} distribution of particles and is occasionally used to eliminate multiply charged particles. The ideal operations of the DMA-APM system for these applications were also discussed. In measuring the m_p or ρ_{eff} distributions, the system would provide accurate data when the rotation speed of the APM is scanned to measure the distributions because the APM resolution parameter λ_c does not vary in that case. In eliminating multiply charged particles, the minimum value of λ_c for that application can be calculated using a derived equation.

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Table 1

The APM transfer function for the uniform flow model (Ehara et al. 1996). $\rho(s)$ is defined as

$$\rho(s) = \frac{1}{\delta}\left(\sqrt{\frac{V_{APM}}{s\omega^2 \ln\left(r_{2\_APM} / r_{1\_APM}\right)}} - r_{c\_APM} \right).$$

	Range	APM transfer function Ω (the uniform flow model)
Region A	$s < s_1^-$	0
Region B	$s_1^- \leq s < s_2^-$	$\left\{\left(1-\rho(s)\right)+\left(1+\rho(s)\right)\exp\left(-\lambda_c\right)\right\}/2$
Region C	$s_2^- \leq s < s_2^+$	$\exp\left(-\lambda_c\right)$
Region D	$s_2^+ \leq s < s_1^+$	$\left\{\left(1+\rho(s)\right)+\left(1-\rho(s)\right)\exp\left(-\lambda_c\right)\right\}/2$
Region E	$s_1^+ \leq s$	0

Table 2

Dimensions of the APM used for calculations in this study. These values are taken from the design values of APM-3600 (KANOMAX Japan, Inc.)

Parameter	Size (m)
r_{1_APM}	0.05
r_{2_APM}	0.052
L_{APM}	0.25

Table 3

The DMA-APM transfer function for the uniform flow model. $\rho(m_p)$ is defined as

$$\rho\left(m_p\right)=\frac{1}{\delta}\left(\sqrt{\frac{qeV_{APM}}{m_p\omega^2\ln\left(r_{2\_APM}/r_{1\_APM}\right)}}-r_{c\_APM}\right).$$

	DMA-APM transfer function $\Phi\left(m_{p,}d_p\right)$ (the uniform flow model)
Region 1	0 (no particle passes through the DMA)
Region 2A	0 (no particle passes through the APM)
Region 2B	$\frac{1}{2\beta}\left[\left\|Z_p\left(d_{p,q},d_{c,q}\right)-(1+\beta)\right\|+\left\|Z_p\left(d_{p,q},d_{c,q}\right)-(1-\beta)\right\|-2\left\|Z_p\left(d_{p,q},d_{c,q}\right)-1\right\|\right]\left[\frac{\left(1-\rho\left(m_{p,q}\right)\right)+\left(1+\rho\left(m_{p,q}\right)\right)\exp\left(-\lambda_c\right)}{2}\right]$
Region 2C	$\frac{1}{2\beta}\left[\left\|Z_p\left(d_{p,q},d_{c,q}\right)-(1+\beta)\right\|+\left\|Z_p\left(d_{p,q},d_{c,q}\right)-(1-\beta)\right\|-2\left\|Z_p\left(d_{p,q},d_{c,q}\right)-1\right\|\right]\exp\left(-\lambda_c\right)$
Region 2D	$\frac{1}{2\beta}\left[\left\|Z_p\left(d_{p,q},d_{c,q}\right)-(1+\beta)\right\|+\left\|Z_p\left(d_{p,q},d_{c,q}\right)-(1-\beta)\right\|-2\left\|Z_p\left(d_{p,q},d_{c,q}\right)-1\right\|\right]\left[\frac{\left(1+\rho\left(m_{p,q}\right)\right)+\left(1-\rho\left(m_{p,q}\right)\right)\exp\left(-\lambda_c\right)}{2}\right]$
Region 2E	0 (no particle passes through the APM)
Region 3	0 (no particle passes through the DMA)

Figure captions

Figure 1. The $log(m_p)$- $log(d_m)$ relationships for the particles. (a) ρ_{eff} ; (b) D_f.

Figure 2. Examples of areas for particle population in the $log(m_p)$- $log(d_m)$ space. (a) Spherical (or nearly spherical) particles with a constant value of ρ_{eff}. ρ_{eff} was assumed to be 1000 kg m^{-3}. Particles can only populate on the black solid line; (b) Aggregated non-spherical particles with a certain value of D_f (e.g., soot). The black solid line on which particles can populate was calculated as $m_p = 6\times10^{-6} d_m^{2.41}$ based on Park et al. (2003); (c) A mixture of spherical and non-spherical particles with a range of ρ_{eff}. Particles may populate in the shaded area.

Figure 3. Examples of (a) the DMA and (b) the APM transfer functions. The DMA transfer function was calculated at d_c = 100 nm for β = 0.1. The following parameter set was used to calculate the APM transfer function: V_{APM} = 100 V, ω = 523.599 rad s^{-1} (equivalent as 5000 rpm), Q_{APM} = 1.67×10^{-5} m^3 s^{-1} (equivalent as 1 l min^{-1}), q=1, and d_m = 100 nm.

Figure 4. Diameter dependences of (a) m_c , $m_1^{\pm}$, and $m_2^{\pm}$, and (b) λ_c. The following parameter set was employed to obtain these values: V_{APM} = 100 V, ω = 523.599 rad s^{-1}, Q_{APM} = 1.67×10^{-5} m^3 s^{-1}, and q=1.

Figure 5. Seven different regions for the DMA-APM transfer function.

Figure 6. Examples of the DMA-APM transfer functions calculated using (a) the uniform flow model and (b) the parabolic flow model. The following parameter set was employed for the calculations: V_{APM} = 100 V, ω = 523.599 rad s^{-1}, Q_{APM} = 1.67×10^{-5} m^3 s^{-1}, q=1, d_c = 100 nm and β = 0.1.

Figure 7. Comparison of diameter resolutions of the DMA and APM for particles with a uniform value of ρ_{eff}. Grey dash lines show important values for the transfer functions of the DMA and the APM, including m_c, $m_1^{\pm}$, d_c, d_{min}, and d_{max}. A black dashed line for ρ_{eff}, which corresponds to the value in the central part of the classification region (ρ_{eff} = 930 kg m^{-3}), is also shown. If all the particles populate on the line of ρ_{eff} = 930 kg m^{-3}, then particles with $m_1^{-} \leq m_p \leq m_1^{+}$ can be classified by the system. The corresponding diameter range ($d_{APM\,\min} \leq d_m \leq d_{APM\,\max}$) is narrower than the particle classification range by the DMA ($d_{\min} \leq d_m \leq d_{\max}$). The colored area represents the DMA-APM transfer function, which is calculated at V_{APM} = 85 V, ω = 523.599 rad s^{-1}, Q_{APM} = 5.0×10^{-6} m^3 s^{-1}, q=1, d_c = 100 nm and β = 0.1. See the text for further details.

Figure 8. Comparisons of the APM scanning methods. (a~c) shows the DMA-APM transfer functions for rotation speed scanning and (d~f) corresponds to voltage scanning. These transfer functions were calculated for Q_{APM} = 1.67×10^{-5} m^3 s^{-1}, q=1, d_c = 100 nm and β = 0.1. The parameter sets of {V_{APM}, ω} are (a) {100 V, 641.274 rad s^{-1}}, (b) {100 V, 523.599 rad s^{-1}}, (c) {100 V, 427.516 rad s^{-1}}, (d) {66.667 V, 641.274 rad s^{-1}}, (e) {100V, 641.274 rad s^{-1}}, and (f) {150 V, 641.274 rad s^{-1}}.

Figure 9. Elimination of multiply charged particles by the DMA-APM system. (a) the uniform and (b) the parabolic flow models were used for the calculation. The following parameter set was employed for the calculations: V_{APM} = 85 V, ω = 523.599 rad s^{-1}, Q_{APM} = 3.33×10^{-5} m^3 s^{-1}(equivalent as 2 l min^{-1}), $d_{c,+1}$ = 100 nm and β = 0.1. The DMA-APM transfer function for +2 particles does not overlap with the line for ρ_c of +1 particle (930 kg m^{-3}).