ESTIMATION OF MASS WITH THE MODEL 3321 APS™ SPECTROMETER

APPLICATION NOTE APS-001

Introduction

Particle size and mass are two important parameters that are often used to characterize an aerosol. Ambient air quality standards (PM_{10} and $PM_{2.5}$), exposure assessment of inhalable particulate matter and FDA guidelines for pharmaceutical aerosol characterization are all based on mass and aerodynamic size of the particles. Most of these applications will benefit from real-time measurement of mass-weighted aerodynamic size distributions as this will drastically reduce the time needed to characterize the aerosols along with providing a higher resolution of particle size data. Currently, there is a lack of methodologies to directly measure mass-weighted aerodynamic size distribution of an aerosol in real-time. In the absence of such instrumentation, measurements of count-weighted aerodynamic size distributions can be used as surrogates to estimate the mass-weighted aerodynamic size distributions.

The Aerodynamic Particle Sizer® (APS™) spectrometer is the only commercially available instrument that measures count-weighted aerodynamic size distributions in real-time. A complete particle size distribution in the range of 0.5 to 20 micrometers may be obtained in a matter of seconds, making the APS the instrument of choice for a broad range of applications. This application note discusses the requirements that need to be fulfilled for accurate estimation of mass-weighted size distributions from APS measurements. Selected case studies are presented below to investigate applicability of the APS as a surrogate for mass measurement. The first case study discusses APS™ spectrometer estimated ambient coarse mode PM mass concentrations and comparisons with Federal Reference Method (FRM) samplers at various locations in the US. The second case study discusses the estimation of Mass Median Aerodynamic Diameter (MMAD) of Metered Dose Inhalers (MDIs) by the APS™ spectrometer. A discussion on the measurement of liquid droplets and determination of their mass concentration by the APS™ spectrometer follows these case studies.

Mass-weighted aerodynamic size distribution from APS™ spectrometer measurements

Calculations

Differential mass concentration for a given aerodynamic diameter (dM_{Dae}) can be calculated for each channel of the APS™ spectrometer as:

$$dM_{D_{ae}} = dN_{D_{ae}} \frac{\pi}{6} D_{ve}^3 \rho_p \qquad \text{Equation 1}$$

Where, D_{ae} is the aerodynamic diameter measured by the APS™ spectrometer, dN_{Dae} is APS™ spectrometer measured differential number concentration for a given aerodynamic diameter, ρ_p is the density of the particle and D_{ve} is the volumetric equivalent diameter which can be calculated from the definition of terminal velocity (Hinds, 1998) as:

$$D_{ve} = D_{ae} \sqrt{\frac{\rho_0 C_{ae} \chi}{\rho_p C_{ve}}} \qquad \text{Equation 2}$$

Where ρ_0 is unit density (1 g cm^{-3}) and χ is the aerodynamic shape factor. C_{ae} is the Cunningham correction factor associated with the aerodynamic diameter, and C_{ve} is the Cunningham correction factor associated with the volume equivalent diameter.

Substituting eq. 2 into eq. 1, the following expression for differential mass is obtained:

$$dM_{D_{ae}} = dN_{D_{ae}} \frac{\pi}{6} D_{ae}^3 \left(\frac{\rho_0 C_{ae} \chi}{C_{ve}} \right)^{3/2} \frac{1}{(\rho_p)^{1/2}} \qquad \text{Equation 3}$$

As an illustrative example, figure 1 displays a mass-weighted aerodynamic size distribution estimated from APS measured count-weighted aerodynamic size distribution using eq. 3.

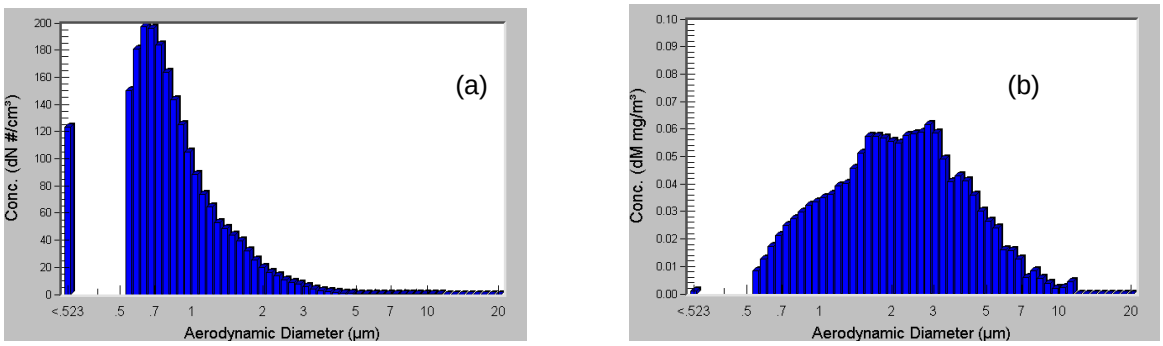

Figure 1. Polydispersed salt (NaCl) aerosol size distributions. (a) APS-measured count-weighted aerodynamic size distribution (b) APS-estimated mass-weighted aerodynamic size distribution using χ =1 and ρ_p=2.16 g cm^{-3}.

Discussion

Eq. 3 demonstrates that, a function of particle diameter cubed, APS-estimated mass is strongly affected by sizing of the aerosol. It is progressively less strongly affected by shape factor (to the power 3/2), counts (to the power 1), and particle density (to the power ½). Thus, in order to estimate accurate mass-weighted size distributions from APS™ spectrometer measurements, following conditions should be fulfilled:

(i) **Accurate sizing.** The APS™ spectrometer uses a patented* time-of-flight (TOF) technique for aerodynamic sizing of particles which involves acceleration of aerosol particles in response to the accelerated flow of the sample aerosol through a nozzle. As particles exit the nozzle, the TOF between two laser beams is recorded and converted to aerodynamic diameter. Generally, sizing accuracy has not been a problem with the APS™ spectrometer, except in the case where large liquid droplets with low viscosity and surface tension may deform during acceleration through the nozzle. Large liquid droplets when flattened in the acceleration zone, experience greater drag than a spherical particle, and are, in turn, sized too small (Baron, 1986). The droplet correction factor (Baron et al., 2004) should be applied to account for deformation of liquid droplets in the acceleration nozzle of the APS™ spectrometer to obtain accurate sizing.

(ii) **Knowledge of particle shape factor and density**. The particle's shape factor and density depend on particle composition, type and configuration. Therefore, the most successful applications using the APS as a mass-estimating device are those where the particle characteristics are well known.

(iii) **Representative sampling**. For real-life applications of the APS™ spectrometer, representative sampling of an aerosol is essential to avoid either an overestimation or an underestimation of particle concentrations of the sampling environment. Representative sampling can be ensured by configuring the APS™ spectrometer to sample isokinetically from an airstream.

(iv) **Efficient counting**. Counting efficiencies of the APS™ spectrometer for various particle sizes and types can be characterized. For example, Volckens and Peters (2005) found that the Model 3321 APS™ spectrometer counts solid particles with near 100% efficiency. For liquid particles, the APS™ spectrometer counting efficiencies were found to be substantially less than 100%. A correction for counting efficiencies should be made where necessary to obtain accurate mass concentrations from the APS™ spectrometer.

* United States Patent Number 5,561,515

Selected case studies

Ambient coarse PM mass

Peters (2006) applied simple assumptions of shape factor and density (shape factor =1.4, density =2.0 g cm^{-3}) to estimate the mass concentration of ambient coarse mode particulate ($PM_{10-2.5}$) with data from the Model 3321 APS™ spectrometer. These estimates compared well with collocated, time-integrated filter based federal reference method (FRM) samplers in Phoenix (AZ) and Riverside (CA). However, the mass concentration estimated with the APS™ spectrometer was approximately 57% of that measured with FRM samplers in Gary (IN). Similar results to those found in Gary were found in a recent study in Birmingham (AL). Recent work suggests that removing moisture from the air entering the inlet of the APS™ spectrometer improves the correlation between APS™ spectrometer -estimated and FRM-measured mass, but does not improve the bias. This bias may relate to differences in aerosol shape factor and density among sites in the US. The shape factor and density assumed by Peters may be applicable in western US sites (Phoenix and Riverside) that are in close proximity to the desert and have a dry climate, but not in eastern US sites (Birmingham and Gary).

Figure 2 (data from Phoenix in January 2004) illustrates the importance of applying shape factor and density in eq. 3 when estimating mass concentration with the APS™ spectrometer. The slope of the relationship between APS™ spectrometer –estimated $PM_{10-2.5}$ and that measured by the FRM improved from 0.87 to 0.99 by adjusting both shape factor and density. Interestingly, correction of density only caused the slope to be reduced to 0.60.

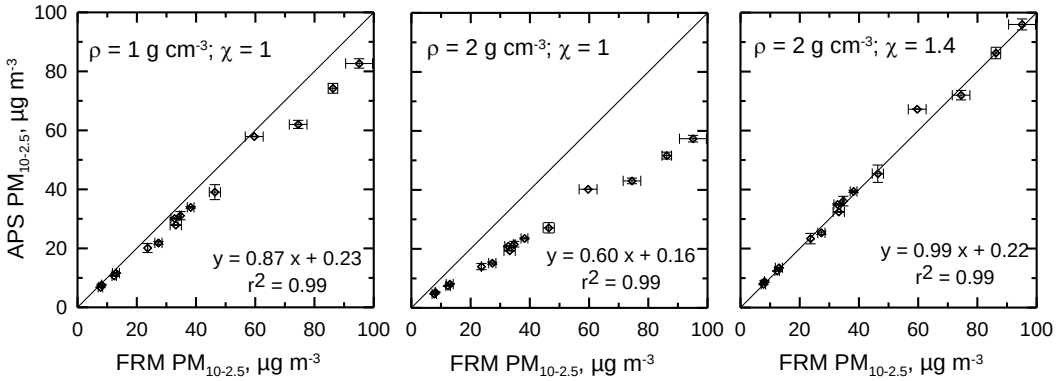

Figure 2. Comparison of $PM_{10-2.5}$ estimated with data from APS™ spectrometer to that measured with filter-based FRM samplers in Phoenix, AZ.

Metered-Dose Inhaler (MDI)

Stein et al. (2003) showed that the MMAD estimated with the Model 3321 APS™ spectrometer agreed well with that measured with an impactor for HFA-134a beclomethsone dipropionate dispersed with an MDI. These results demonstrate that Model 3321 APS™ spectrometer is capable of accurate mass-weighted size distribution measurement of solution MDIs. MMAD data from Andersen Cascade Impactor (ACI) and APS™ spectrometer measurements of HFA solution MDIs are summarized in figure 3. This study also demonstrated the use of APS™ spectrometer to investigate the factors influencing the size distribution from solution MDIs.

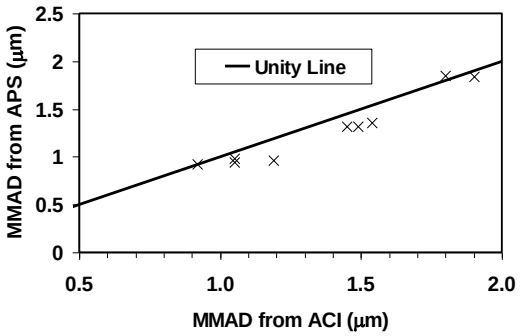

Figure 3. MMAD from size distribution measurements of experimental solution HFA-134a beclomethasone dipropionate MDIs made using the APS 3321 and ACI. The solid line represents a unity relationship. (Adapted from Stein et al., 2003)

Liquid Droplets

Several groups have found mass concentration of oil drops calculated with data from the Model 3321 APS™ spectrometer to be less than that measured with an impactor (Peters and Leith, 2003; Pagels et al., 2005). As discussed earlier, whereas solid particles retain their shape, liquid droplets flatten in the acceleration nozzle of the APS™ spectrometer, experience greater drag than a spherical particle, and are, in turn, sized too small (Baron, 1986). Additionally, droplets may impact on the inner surfaces of the APS™ spectrometer resulting in less than 100% transmission efficiencies (Volckens and Peters, 2005). Both effects result in a lower mass concentration calculated with eq. 3: drop flattening results in D_{ae} being measured too small; and impaction in the nozzle results in dN_{Dae} being measured too small.

Conclusion and future work

The APS™ spectrometer is the only commercially available instrument that measures count-weighted aerodynamic size distributions of particles in real-time. APS™ spectrometer has been shown to perform these measurements rapidly in a matter of few seconds. The applicability of APS™ spectrometer as a surrogate for real-time mass measurement will greatly benefit a variety of applications as reviewed in this application note. Based on the preceding discussion on factors affecting mass estimation using the APS™ spectrometer and a review of selected case studies, it is concluded that the APS™ spectrometer can be used successfully for accurate estimation of real-time mass-weighted aerodynamic size distribution of solid aerosols with known shape factor and density. For liquid droplets having low viscosity and surface tension (like oil), the APS™ spectrometer have been found to underestimate the mass concentration. Investigations conducted so far suggest that a careful characterization of transmission losses by size coupled with an adjustment for droplet flattening is key for using the APS™ spectrometer to measure mass concentration by size for liquid droplets accurately. Future work in understanding these transmission losses will greatly enhance the applicability of APS™ spectrometer for characterizing liquid aerosols.

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