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Size Shifts in Measurements of Droplets with the Aerodynamic Particle Sizer and the Aerosizer

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Observations of the size of liquid droplets using the Aerodynamic Particle Sizer (APS) and the Aerosizer indicated that the measured size was significantly different from the aerodynamic diameter as calculated by measuring droplet settling velocity. The size shifts appeared to be caused by droplet distortion in the detector flow field for the Aerosizer. However, for the APS, droplet sizing was further affected by droplet impaction on the upstream side of the focusing nozzle. It is suggested that liquid accumulated in and constricted the nozzle, resulting in a particle velocity increase at the sensor. The size shift can occur with the deposition of <0.5 μL liquid onto the nozzle; the size shift can occur in 1–10 minutes even at concentrations of 1000 particles/L; and the size shift can disappear after cessation of liquid aerosol sampling. CFD calculations provided information about the amount of velocity increase at the APS sensor for a selected constriction. Both solid and liquid particles are affected by the nozzle constriction, which produces approximately the same percentage size shift throughout the measurement range.

The size shifts (Δ) were related to droplet aerodynamic diameter (μm), viscosity (Pa-s), and surface tension (N/m) by the following empirical equation: $\Delta = -a\ \text{diameter}^b/(\text{surface tension}^c \times \text{viscosity}^e)$. The value of b was arbitrarily set to two. The values for a, c, and e for the APS (including both droplet distortion and nozzle constriction) and for the Aerosizer were determined by a regression analysis of the available data.

INTRODUCTION

The measurement of particle aerodynamic diameter in the range of 0.5 μm to 20 μm is useful for a number of applications where particle settling and impaction are important for understanding particle dynamics. For example, aerodynamic diameter is the equivalent diameter most frequently used for understanding particle deposition in the respiratory tract and in particle filters. Two instruments were developed to measure aerodynamic diameter in real time, namely the Aerodynamic Particle Sizer (APS, TSI, Inc., Shoreview MN) and the Aerosizer (Amherst Process Instruments, more recently from TSI, Inc.). These instruments are in widespread use for environmental and other aerosol measurements and, thus, any potential measurement errors are of interest. Both instruments use particle acceleration through a nozzle and measurement of the particle velocity in the acceleration flow field using a laser velocimeter. The particle size is related to the measured velocity through calibration with unit density spherical particles. In the case of the APS, the particles pass between two laser beams and the time of passage for each particle is recorded in a series of electronic channels (4 ns/channel) called the accumulator. The air velocity through the nozzle of each instrument is high, with the air through the APS acceleration nozzle traveling at about 150 m/s and the air through the Aerosizer nozzle traveling at sonic velocity, i.e., about 340 m/s. The high acceleration through these nozzles mean that some of the particles are measured under non-Stokesian conditions and that the measured sizes are a function of particle density and shape in addition to aerodynamic diameter (Baron et al. 2001).

In the APS, a second nozzle upstream of the acceleration nozzle focuses the particles in a clean sheath air. Some acceleration of the particles occurs in this focusing nozzle (Ananth and Wilson 1988). The Aerosizer does not have such a focusing nozzle.

Several researchers have noted that liquid droplets traveling through the APS nozzle are distorted by the air pressure during acceleration (Baron 1986; Griffiths et al. 1986; Bartley et al. 2000). This distortion causes a flattening of the droplets into an oblate spheroid having increased drag. Hence, the droplets achieve a greater velocity in the sensing zone and their diameters are measured as being shifted to smaller values than those expected for the originally spherical droplets.

The objective of this study was to understand and quantify the size shifts and understand how to eliminate or adjust for

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their presence. A series of measurements of these diameter or size shifts, which represent measurement errors, were made for several liquids using several models of the APS and Aerosizer over about 20 years. The measured sizes from the instruments were compared to aerodynamic diameter as measured by the settling velocity of the droplets (indicating true aerodynamic diameter). The measured size shift of droplets appears to be at least partially caused by droplet distortion as suggested by earlier researchers, but an additional size shift appears to be related to liquid deposition in the APS focusing nozzle.

EXPERIMENTAL PROTOCOL

Oil droplets were generated from an isopropanol solution of the oils indicated in Table 1. The oils included several viscosities of polydimethylsiloxane polymer fluid available from Dow Corning (Midland MI), oleic acid, triethanolamine (TEA), and di-octyl sebacate (DOS) (or (±)-Bis (2-ethylhexyl) sebacate (Aldrich Chemical, Milwaukee, WI). The droplets were generated using a modified vibrating orifice generator (VOMAG, Model 3050, TSI, Inc.). The generator was modified to use a pressurized liquid feed rather than the manufacturer-provided syringe pump feed. Particles from the VOMAG were injected into a vertical cylindrical chamber 0.5 m high with a diameter of 0.2 m to ensure that solvent evaporated from the droplets prior to measurement by the APS and Aerosizer. Concentration of aerosol in the measurement chamber varied with particle size and experiment, but was typically on the order of 10,000 particles/L or less. In each set of experiments, the APS nozzle was cleaned before use. Size calibration of the APS and Aerosizer was achieved using reference polystyrene latex particles of several sizes ranging from 2–20 μm (Bangs Laboratories, Fishers, IN). Aerosols of the reference particles were introduced into the APS and Aerosizer by mechanical fluidization of the particles from a glass vial or by carefully brushing small quantities of powder from a glass slide into the air near the instrument inlet (Blackford and Rubow 1986). Both instruments could easily distinguish between single particles and multiplets, so the relatively crude aerosolization method was effective for size calibration. Instrument calibration was checked before and after each measurement of droplet material and size.

TABLE 1
Properties of liquids used in the study of droplet size shifts

Oil type	Surface tension (N/m)	Viscosity (Pa-s)
200/50[1]	0.0208	0.048
200/100[1]	0.0209	0.0964
200/200[1]	0.021	0.1934
200/500[1]	0.0211	0.4845
Triethanolamine (TEA)	0.0489	0.59
Oleic Acid	0.032	0.0256
Di-octyl Sebacate (DOS)[2]	0.0322	0.027

[1] Designation for Dow Corning Polydimethylsiloxane oils.

[2] Also called (bis)-2-Ethyl Hexyl Sebacate.

The aerodynamic diameter of each droplet type was measured by observing their settling velocity in a vertical 6 mm inner diameter glass tube (John and Wall 1983). Several methods of introducing the droplets into the settling chamber were used, but the most efficient one was to fill a vertical 1.5 cm diameter, 1 m long copper pipe with the droplet aerosol, cap the top of the pipe, and align the pipe vertically over top of the settling chamber. The droplets fell through a 1 mm diameter hole at the top of the settling chamber and were illuminated by a vertical helium-neon laser beam shining through a glass window at the bottom of the chamber. The falling particles were observed in a darkened room. The velocity of the droplets was measured visually by measuring the time taken for the droplet to settle a predetermined distance marked on the settling chamber. About 10 droplets of each size and material were measured to get a precise estimate of settling velocity. Droplets in the range of 5–18 μm aerodynamic diameter were measured in this fashion. When smaller than 5 μm, the droplets were difficult to see and often drifted out of the laser beam before their velocity could be measured, while droplets larger than 18 μm settled so quickly that their transit time in the settling tube could not be measured accurately.

Several measurements of polydisperse oil droplets were made with the APS in order to assess possible measurement artifacts caused by interactions between particles of different sizes. The droplets were produced in approximately the 7–15 μm range by changing the VOMAG drive frequency to produce a polydisperse aerosol (geometric standard deviation approx. 1.2). The size distribution was observed for several oils. A second, lower concentration aerosol of 3.1 μm latex particles was added to the oil droplet aerosol to monitor the shift in particle size for solid particles.

Since its initial introduction in 1983, the Aerodynamic Particle Sizer has undergone development resulting in several models. The data from several instruments used in this study included models 3300, 3320, and 3321 and sometimes included two instruments of the same model. All these instruments used the same nozzle system and the same flow rates through the focusing and acceleration nozzles. Similarly, the Aerosizer passed through several model changes and the Model LD and Model DSP were used in this study. The Aerosizer nozzle and flow rate also did not change with changes in model designation.

COMPUTATIONAL FLUID DYNAMICS SIMULATION

Computational fluid dynamics (CFD) simulations were made of the APS nozzle system using the commercial software Fluent 6.2 (ANSYS-Fluent Inc. Lebanon NH) with nozzle dimensions and appropriate distances within the nozzle sensing system obtained courtesy of TSI, Inc. An axially symmetric 2-D model with 257,286 cells represented the 3-D cylindrical interior and boundaries of the nozzle system and a 1.3 cm long tubular region downstream of the acceleration nozzle. The pressure across an APS nozzle operated at 5 L/min (-1.18×10^4 N/M^2) was used as the outlet boundary condition in this subsonic compressible flow simulation. Controlling this pressure at the outlet of the nozzle system simulates the operation of the APS flow control system. Several rectangular cross-section ring obstructions (reducing the area of the nozzle opening by 10, 20, and 30%) were placed radially inward from the tip of the focusing nozzle to simulate oil accumulation inside the nozzle. The flow field for each simulation was iterated until a normalized residual of 2×10^{-5} was achieved. The conservation equations describing fluid motion (Navier-Stokes equations) were discretized using either a second order or a second order upwind scheme, which was applied from the first iteration until the iterative convergence criterion was met. Under-relaxation parameters included: 0.5 for pressure, 0.4 for momentum, 0.8 for turbulent kinetic energy and dissipation rate, and 1 (no under-relaxation) for turbulent viscosity. Turbulence was approximated using the RNG k-ε model. The air velocity and velocities of several size particles were calculated along the axis of the nozzle system at the sensor location downstream of the acceleration nozzle. The fraction of particles of each size depositing on the nozzle surface was also recorded.

RESULTS

The measurement of settling velocity of a droplet size was typically within ±2%. A measurement of one of the latex reference particles agreed within about 2%. It was difficult to get a more precise estimate of the latex mean size because of the larger geometric standard deviation.

The APS data for size shift as a function of particle size and oil type are presented in Figure 1. Similar data for the Aerosizer are presented in Figure 2. The size shift generally increases with particle size and some oils present a larger particle shift than others. Most of these data were accumulated over a period of about six years with several different experimenters collecting the data. Also included are data from oleic acid measurements taken by Baron (1986). These match later measurements of size shift for the same oil quite well and are included in Figure 1 along with more recent data.

More recent measurements were made immediately after carefully cleaning the nozzles to elucidate the time-dependent size shift in the APS and Aerosizer. The Aerosizer did not show any significant difference from the data indicated in Figure 2. However, the APS showed a time-dependent shift in measured size within several minutes of starting the measurement with a clean focusing nozzle. An example of this shift is presented in Figure 3 for di-octyl sebacate (DOS); the liquid droplet size changed from a measured diameter of 11.47 μm (12.95 μm aerodynamic diameter) to 10.36 μm rather suddenly after about 280 sec of exposure to particle concentration of about 3000 particles/L. The size shift was stable until the injection of droplets into the drying chamber stopped. The measured size drifted upward towards the original size, but did not reach the originally measured size by 580 sec. A small but measurable concentration remained in the chamber for this measurement. A low concentration of latex particles (nominally 8 μm) was measured at the same time and the size shift of these solid particles coincided with that of the liquid droplets; this confirmed that the size shift occurring after several minutes exposure time was not caused by droplet distortion.

Another experiment was performed using the Dow 200/50 oil and carefully cleaning the focusing nozzle before each APS measurement to observe the size shift when the nozzle was clean.

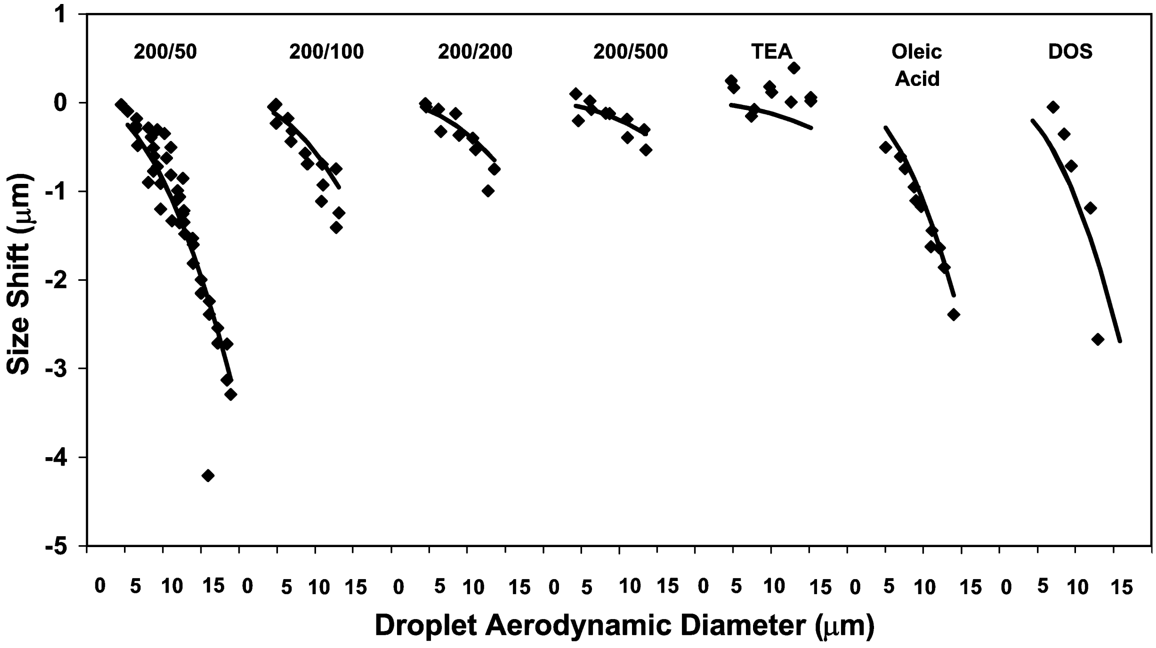

FIG. 1. Size shift data for the APS along with the least squares fit using Equation (1). The largest size shift value for Dow 200/50 was treated as an outlier.

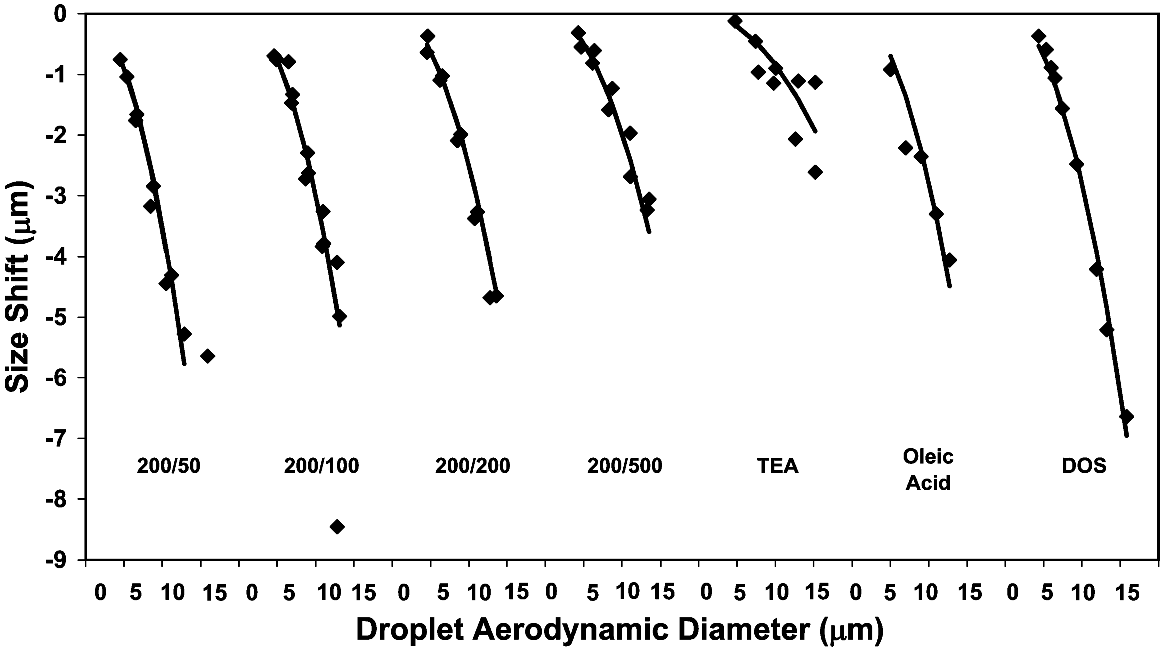

FIG. 2. Size shift data for the Aerosizer along with the least squares fit using Equation (1). The largest size shift values for DOW 200/50 and 200/100 are treated as outliers.

The nozzle was then allowed to load with oil and the maximum shift was recorded. Example results are indicated in Figure 4.

Several measurements were also made using a broader distribution of droplet sizes at several droplet viscosities. These distributions were observed as a function of time. An example of the DOS size distribution shift before and after the inner nozzle of the APS was loaded with oil is shown in Figure 5. Latex spheres with 3.1 μm diameter were also generated during these experiments to estimate the shift for these solid particles (indicated in Table 2). The amount of liquid depositing on the nozzle was estimated using the liquid size distribution and the deposition fraction based on an empirical deposition model for

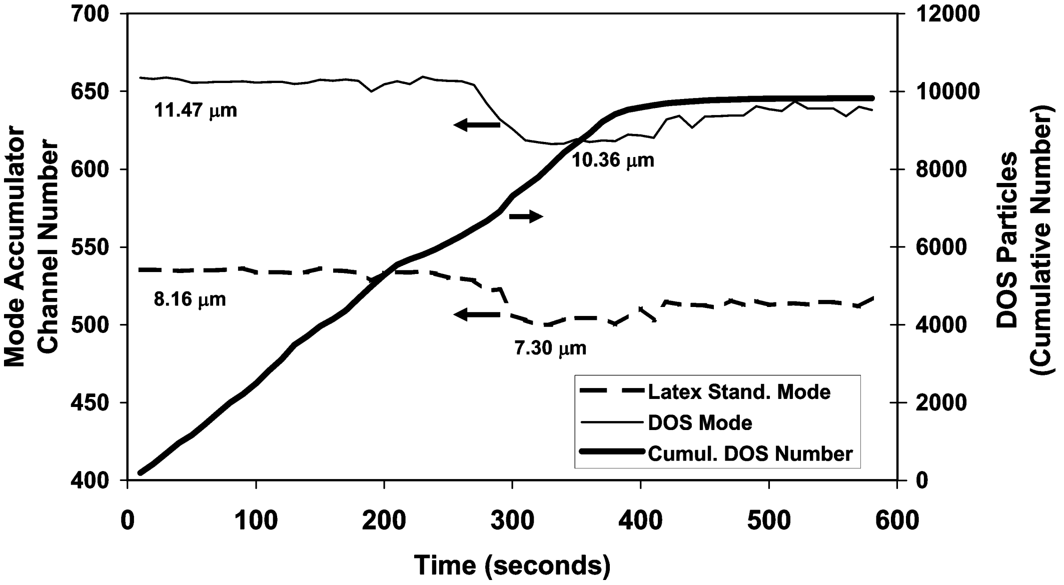

FIG. 3. The size of particles was monitored using the mode peak channel number of the respective particle types. The APS data are presented in terms of the peak concentration (size distribution mode) in the accumulator data. The APS raw size information (accumulator channel number) is directly related to aerodynamic size through the calibration. A time dependent size shift of about 10% observed for both DOS droplets (initially 11.47 μm) and 8.16 μm latex particles at about 280 sec. Both shifts start to decrease after DOS production ceases at about 380 sec. APS measurements were made at 10 sec intervals.

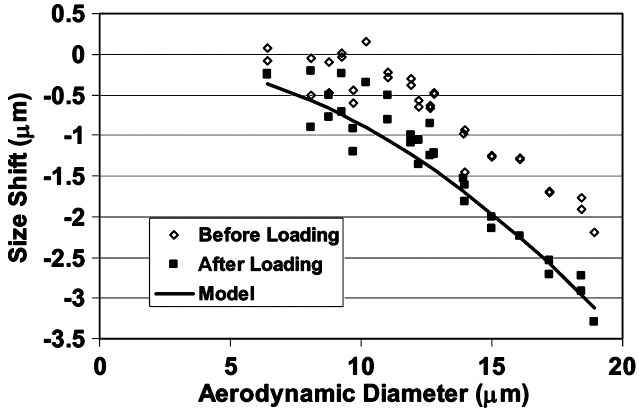

FIG. 4. Comparison of size shift in the APS immediately after cleaning the focusing nozzle (before loading) and the maximum size shift at a later time after the enough DOW 200/50 oil droplets have deposited in the nozzle (after loading).

TABLE 2

Observed parameters for size shift with broad distribution oil aerosols. Experiments for each oil were performed four to six times. The recovery times are approximate because of the slow decay of the peak back to the original size

Liquid type	Fractional size 3.1 μm shift of latex particles	Calculated liquid at time of deposited size shift (μL)	Recovery time (min)
200/200	0.055–0.070	0.045–0.18	15–18
200/100	0.044–0.059	0.042–0.40	15–16
DOS	0.090–0.10	0.22–0.42	15–16

a constriction (Muyshondt et al. 1996). Table 2 indicates the amount of liquid estimated to have deposited at the time of the size shift using the integrated particle concentration measured by the APS. The approximate time it took for the 3.1 μm latex particle size to shift completely back to the original size after APS exposure to liquid droplets ceased was noted and these times are indicated in Table 2. It was further noted that upon re-exposure to liquid droplets, the size shift occurred more rapidly, with estimated liquid deposits of about 0.02 μL.

The axial flow velocity results in the CFD simulation of the APS nozzle system were very similar to those obtained by Ananth and Wilson (1988). The simulated air velocity in the clean nozzle, based on nozzle system and laser beam dimensions provided by TSI, produced a 0.7 μm particle velocity that was within 2% of that indicated by the APS instrument calibration. The axial velocities for several particle sizes for a clean nozzle and 10%, 20%, and 30% area constriction are indicated in Table 3. A smooth polynomial calibration curve was fitted to the data for the unconstricted nozzle (TableCurve 2D, Systat Software Inc., San Jose, CA). This calibration curve was used to calculate the size shift produced by the several nozzle constrictions (Table 3).

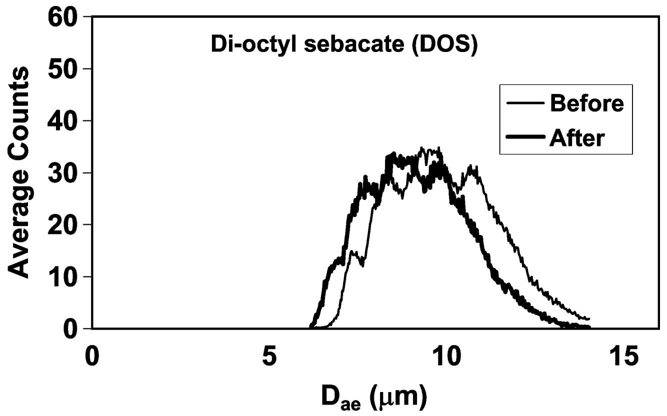

FIG. 5. Example size distributions before and after APS focusing nozzle is loaded with oil droplets of DOS.

EMPIRICAL FIT OF SIZE SHIFT DATA

It was noted that the data appeared to be a function of liquid viscosity and surface tension, as shown by Bartley et al. (2000). Most of the data could be fitted with a simple empirical function including droplet diameter, surface tension, and viscosity.

$$\Delta = -\, a\, d^{b}/(\eta^{c}\sigma^{e}) \qquad [1]$$

where Δ is the size shift in micrometers, η is the liquid viscosity (Pa-s), and σ (N/m) is the liquid surface tension. The data were fitted using a least squares approach by minimizing the sum of the mean squared errors. The diameter exponent was approximately two for all the fits and the goodness of fit did not degrade greatly when it was constrained to be two. Varying the APS fit diameter exponent between 1.7 and 2.9 increased the least squares error by less than 15%. Decreasing the diameter exponent for the Aerosizer data from 2 to 1.74 resulted in a decrease in the least squares error of 15%. To keep the fitting process simpler, subsequent fits were constrained with the diameter exponent equal to 2. In the least squares fits, three data points were excluded; one for the APS data (largest shift value for 200/50 data) and two for the Aerosizer data (the largest shift values for 200/50 and 200/100). For the APS (data from Models 3300, 3320, 3321), the least squares fitted constants were: $a = 2.723 \times 10^{-4}$; $b = 2$; $c = 0.6486$; and $e = 0.3864$ ($n = 105$; mean square error $= 0.0705$). While a wide range of oil viscosities was used, a relatively narrow range of surface tensions was covered by the oils in this study (Table 1). This means that the regression may not be robust with respect to other values of surface tension.

For the Aerosizer (data from Models LD and DSP) the constants were: $a = 4.061 \times 10^{-4}$; $b = 2$; $c = 0.2516$; and $e = 0.9583$ ($n = 67$; mean square error $= 0.116$). The resultant curves for the APS and Aerosizer are plotted with data points in Figures 1 and 2, respectively.

TABLE 3

Air and particle velocities at the APS optical sensor calculated using CFD for a clean nozzle and for area constrictions of 10%, 20%, and 30%. The percent droplets deposited on the upper surface of the focusing nozzle also are indicated in the second column

Particle diameter	Open nozzle velocity m/s (percent deposited)	10% constriction velocity m/s (% size shift)	20% constriction velocity m/s (% size shift)	30% constriction velocity m/s (% size shift)
Air	150.6	152.6 (–2.2)*	154.6 (–4.4)*	156.5 (–6.4)*
0.7 μm	134.2 (0)	136.0 (–8.0)	138.5 (–20)	141.3 (–35)
1 μm	123.1 (0)	125.1 (–5.1)	127.9 (–12)	130.7 (–20)
2 μm	95.1 (0)	97.5 (–6.1)	100.5 (–13)	159.9 (–19)
5 μm	61.0 (6.3)	62.9 (–5.1)	65.4 (–11)	71.5 (–19)
10 μm	40.3 (40.3)	42.0 (–6.6)	43.9 (–13)	48.3 (–17)
15 μm	31.6 (47.5)	33.0 (–6.8)	34.5 (–13)	37.9 (–20)

*The values in parentheses are the number of accumulator channels shifted relative to the unconstricted nozzle air velocity.

DISCUSSION

Early visual observations of droplet distortion (Baron 1986; Baron et al. 2001) supported the assumption that this distortion was entirely responsible for changes in the measured droplet size compared to the true aerodynamic diameter in the APS measurement range. Theoretical work indicated that droplet distortion should occur, producing size shifts on the order of those observed (Bartley et al. 2000). Further visual observations of distorted droplets indicated that distortion does occur under certain circumstances (Secker et al. 2000). The current observations with the APS that liquid and solid particles exhibit a similar time dependent size shift (Figure 3) indicated that the size shift was at least partially caused by something other than droplet distortion. In the APS, the shift to smaller particle size was caused by particles having a higher velocity than expected. Droplet distortion forces the droplets into an oblate spheroid that has higher drag than a sphere of the same volume, hence increasing the acceleration and producing a higher velocity in the measurement zone of the APS (Bartley et al. 2000).

Another mechanism for increasing the particle velocity in the APS measurement zone was needed to explain the size shift for solid particles. It is known that particles will impact in the inner or focusing nozzle of the APS, with the fraction impacted increasing with particle size, especially above 5 μm (Muyshondt et al. 1996; Volckens and Peters 2005). It was found experimentally that oil deposition at the upper surface of the focusing nozzle increased from about 5% at 5 μm to 40% at 10 μm (Volckens and Peters 2005). Note that the CFD calculation (Table 3) closely agrees with these experimental results.

In the past, it was assumed that droplets collecting in the focusing nozzle did not interfere with the acceleration of subsequent particles passing through the nozzle. However, our observations suggest that if sufficient liquid accumulates (on the order of 0.04–0.4 μL), it may flow down the upper surface of the focusing nozzle and constrict the nozzle opening (depicted in Figure 6). Since we observed that the constriction appears to diminish with time after cessation of exposure to droplets, the air flow through the nozzle presumably causes the oil to continue to flow through the nozzle opening and build up on the downstream surface of the nozzle. The liquid flow velocity and shape on the upstream nozzle surface is dependent on the same liquid properties that affect distorting droplets, namely, viscosity and surface tension. Therefore, if nozzle constriction occurs, one might expect that the droplet distortion size shifts observed also have a response that is a function of these properties. This may be why the model represented by Equation (1) appears to fit the APS data as well as that of the Aerosizer. Note that the acceleration system in the Aerosizer does not have the same type of constriction that would cause additional droplet

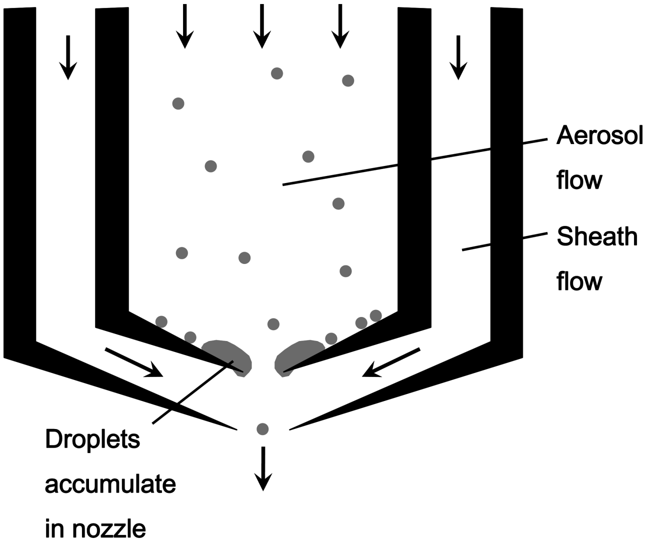

FIG. 6. Schematic of APS nozzles showing suggested mechanism for a size shift when sampling liquid droplets. The droplets impact on the upper surface of the focusing nozzle and partially occlude the nozzle. This increases the velocity of the droplets and is interpreted by the APS as a decrease in measured diameter.

acceleration. In addition, the acceleration flow in the Aerosizer nozzle, being constrained as a sonic nozzle, cannot increase significantly.

The amount of liquid deposited on the focusing nozzle of the APS must be enough to significantly constrict the nozzle to cause the magnitude of the measured size shift (i.e., velocity increase at the laser detector). As indicated above, the minimum amount of oil that appeared to produce the size shift was 0.042 μL. If we assume that this volume of liquid forms a torus around the edge of the focusing nozzle, the nozzle opening will be constricted by approximately 32%. The CFD calculation showed that the increase in velocity at the focusing nozzle required to give a 10% decrease in measured particle size corresponded to a torus with a volume of about 0.009 μL. This indicates that the volume of oil depositing on the nozzle producing the size shift was about four to fifty times greater than the minimum needed to produce that size shift. This suggests that much of the oil either remains on the upper surface of the nozzle or that it passes through the nozzle in rivulets that only minimally affect particle acceleration. Only after sufficient oil accumulates at the nozzle tip to form a complete circumferential constriction in the nozzle tip does the full particle size shift occur. Further, the size shift pattern indicated in Figure 3 suggests that a maximum size shift occurs. Further, the data in Figure 1 indicates that a consistent level of constriction occurs routinely. This indicates there is a maximum constriction, probably related to the balance between the air pressure pushing the liquid through the nozzle and the viscosity of the liquid.

Additional measurements for polydimethylsiloxane 200/50 oil shown in Figure 4 indicate that different size shifts are consistently obtained when the nozzle is clean and when sufficiently loaded with oil. The initial size shift is apparently caused by droplet distortion, while the maximum size shift is a combination of droplet distortion and the nozzle loading effect. We attempted to relate the starting time (time of exposure after starting with a clean nozzle) of the size shift to the total volume of oil likely to deposit on the nozzle but did not obtain consistent results. This is consistent with the above hypothesis that the full constriction producing the size shift only occurs with a relatively large deposit of oil and when individual rivulets have coalesced to form a complete constriction ring around the circumference of the nozzle. This ring formation would depend on the deposit shape and the flow patterns of the oil deposit.

It therefore appears that a significant portion of the size shifts observed in Figure 1 was likely caused by partial and temporary constriction of the APS focusing nozzle. The results indicated in Figure 1 were surprisingly consistent, considering that measurements were made by several different investigators over several years and with different instruments. For this reason, we feel that the equations that fitted the data and Figures 1 and 2 may be useful to others. However, the equation for the APS should be used with some caution. When the nozzle is clean, the size shift will not be nearly as large as perhaps it would be several minutes later, depending on droplet concentration. In addition, Griffiths et al. (1986) performed similar experiments to ours and found size shifts that were similar in magnitude and also dependent on particle diameter, but had much less dependence on surface tension and viscosity. At this point, we are at a loss to explain the difference in the results of the two studies.

Observation of broader distribution oils with time indicated that the nozzle constriction dependent size shift (observed using 3.1 μm latex particles) was somewhat different for different oils, but not in an apparently consistent manner (Table 2). The size shift was between 5–10% of the reference particle size. The estimate of the amount of oil deposited for these distributions ranged from 0.042 μL to 0.45 μL, with most values in the higher range. It is possible that the nozzle may not have been completely cleaned in some cases, resulting in the lower values. After removing the oil exposure and allowing the nozzle to clear itself (about 15 min), it usually took only about 0.02 μL deposit to re-establish the full size shift, indicating that some oil remained on the nozzle tip, allowing the constriction to readily reform. The observed size shifts with the broader distribution oils were consistent with the size shifts observed for monodisperse oils and the amount of oil deposited, indicating that no interaction occurred between droplets of different sizes that changed the degree of size shift.

The CFD simulations provide information on the effect that a constriction of the focusing nozzle has on the velocity of particles in the detection region of the APS. The simulations indicate that when a size shift occurs caused by nozzle constriction, the percent size shift is similar for all particle sizes in the measurement range, increasing only slightly from 1 μm to 15 μm. The size shifts for 0.7 μm are larger, probably because of the slight air velocity increase with increased nozzle constriction. Submicrometer particles are more sensitive to small changes in air velocity than larger particles because of the slope of the calibration curve in this range. Table 2 indicates that the size shift can range from 4.5% to 10% for several oils. If we select a size shift of 10%, the information in Table 3 can be used to estimate the decrease in focusing nozzle area needed to produce this size shift, namely about 16%.

One way to deal with the issue of APS nozzle constriction caused by droplet deposition is to estimate the time before cleaning is needed. For example, if we sample monodisperse 10 μm droplets at a concentration of 10,000 particles/L, we can estimate the deposited fraction in the nozzle to be 0.68 (Muyshondt et al. 1996; Baron 2001). The time to deposit 0.04 μL on the focusing nozzle is estimated to be 1.5 min, a conservative estimate of the time at which the nozzle should be cleaned. However, under ideal conditions, the time before cleaning is required may be 10 time greater. Estimating the deposition time for a more complex size distribution requires calculation of the amount deposited in each size bin and integrating the volume.

For APS users, there are at least two options for determining whether or not a nozzle constriction size shift has occurred. It was noted in the CFD calculations that the air velocity changed by an amount that would produce a shift in the smallest particles

($<0.5\ \mu$m) of about 1.5 to 2.5 channels in the accumulator distribution of the APS. This may not be a reliable indicator, however, if the shape and concentration of the smallest particle side of the distribution changes significantly during the measurements. Another option might be to generate a small concentration of reference particles at intervals during the measurements to indicate whether or not a size shift has occurred and the magnitude of the size shift.

Once the occurrence and magnitude of the size shift has been observed, the APS user has to determine whether the particles being detected are primarily solid particles or liquid particles. If they are liquid particles, then Equation (1) may be used to correct the size distribution. Alternatively, if the liquid properties are unknown, one could apply the reference particle percent size shift to the entire distribution as a first order correction. Note that this latter approach is likely to lose accuracy with increasing particle size, since it would not include the effect of droplet distortion.

It should be noted that the size shift for all liquids may not follow Equation (1). Equation (1) fits to the TEA and DOS data were not as good as for other oils. These oils have the largest and lowest viscosities of the oils tested, respectively. Thus, extrapolating Equation (1) to liquids with viscosity outside the range of values listed in Table 1 may be problematic. For example, it was observed that water droplet size shift from distortion (Bartley et al. 2000) is much less than predicted by this equation. Our investigation only included a relatively small range of surface tensions and so Equation (1) may not be very sensitive to surface tension values outside this range.

The present observations have been made under conditions where the concentration was relatively low, measured as being on the order of 10,000 particles/L or less. We have not measured higher concentrations under carefully controlled conditions and do not know what the effect of much higher concentrations of oil droplets might be on sizing. We have assumed that the size shift is produced by a small but self-limiting constriction of the nozzle and that higher concentrations will not change this situation. Further testing is required to confirm this.

If solid particles are sampled at a similar or higher concentration compared to the liquid droplets, the combined deposit of these materials on focusing nozzle may exhibit considerably different flow behavior than indicated by just the liquid properties. Normally, oils behave as Newtonian fluids, with shear stress proportional to applied shear rate, as produced by air flow in the focusing nozzle. When a significant concentration of solid particles are added to the fluid droplets, the deposited solid/liquid mixture can become a Bingham fluid, in which there must be a minimum shear stress before the fluid will flow (Chhabra 1993). This could allow a significantly greater and longer term buildup of deposit in the nozzle.

The presence of oils or liquids in a measured size distribution may not only affect the shape of the distribution, but may also cause further error if the distribution is used to estimate mass concentration. For instance, a 10% decrease in all particle sizes will decrease the calculated mass by nearly 30% (Reid and Peters 2007). Obviously, the instrumental response for any given aerosol will be a complex function of time, droplet sizes and concentration, and solid particle sizes and concentration. It may be helpful to use a 5 μm (or smaller) impactor in front of the APS inlet to remove larger droplets. This will reduce the rate of droplet deposition on the focusing nozzle and allow a longer time to obtain accurate results in the lower size range of the APS.

Frequent cleaning of the APS focusing nozzle appears to be warranted to improve size measurement accuracy when sampling liquid droplets, especially those larger than about 5 μm. The deposited liquid may not only affect the focusing nozzle, but may eventually shed or spray off the back of the nozzle and end up depositing on the acceleration nozzle as well. If liquid subsequently sprays off the back of the acceleration nozzle, it may cause particle deposition on the optics. We have not observed effects of this at the low concentrations measured in this study, but suspect that it may occur at high concentrations of large droplets. In the past, this was noted as occurring in the APS 3300 at high concentrations of water droplets and necessitated cleaning the optics and re-calibrating the instrument. The improved capture efficiency of the nozzle in the APS 3321 may reduce this optics contamination, but again, frequent cleaning appears to be a prudent approach to prevent problems caused by droplet deposition.

The droplet size shift data for the Aerosizer are more consistent than for the APS, are fitted well using Equation (1), and appear not to have a time dependent component. This indicates that the size shifts are caused solely by droplet distortion. The Aerosizer size shifts are larger than for the APS because of the higher velocity in the Aerosizer sensor nozzle.

CONCLUSIONS

Measured droplet sizes in both the APS and the Aerosizer exhibit size shifts from true aerodynamic diameter for liquids with a range of viscosities and surface tensions. The APS size shifts appear to be caused by a combination of droplet distortion and focusing nozzle loading, especially for droplets larger than about 5 μm. When the nozzle is clean, the size shift is solely caused by droplet distortion, but can change in minutes to a combination of droplet distortion and nozzle loading even at relatively low concentrations. The Aerosizer size shifts are larger than those for the APS because of the higher acceleration in the sensor and can be reliably predicted using Equation (1). The Aerosizer size shifts appear to be entirely caused by droplet distortion in the sensor flow field.

REFERENCES

Ananth, G. and Wilson, J. C. (1988). Theoretical Analysis of the Performance of the TSI Aerodynamic Particle Sizer: The Effect of Density on Response, *Aerosol Sci. Technol.* 9(3):189–200.

Baron, P. A. (1986). Calibration and Use of the Aerodynamic Particle Sizer (APS 3300), *Aerosol Sci. Technol.* 5:55–67.

Baron, P. A. (2001). Aerosol Calculator (Computer Spreadsheet). www.bgiusa.com, BGI, Inc.

Baron, P. A., Mazumder, M. K., and Cheng, Y. S. (2001). Direct-Reading Techniques Using Particle Motion and Optical Detection. *Aerosol Measurement: Principles, Techniques and Applications*. P. A. Baron and K. Willeke, eds. John Wiley & Sons, New York, 495–535.

Bartley, D. L., Baron, P. A., Martinez, A. B., Secker, D. R., and Hirsch, E. (2000). Droplet Distortion in Accelerating Flow, *J. Aerosol Sci.* 31(12):1447–1460.

Blackford, D. B. and Rubow, K. L. (1986). *A Small-Scale Powder Disperser*. World Congress Particle Technology, Nuremberg, West Germany, TIZ-Fachberiche.

Chhabra, R. P. (1993). *Bubbles, Drops, and Particles in Non-Newtonian Fluids*. CRC Press, Inc., Boca Raton, Florida.

Griffiths, W. D., Iles, P. J., and Vaughan, N. P. (1986). The Behaviour of Liquid Droplet Aerosols in an APS 3300, *J. Aerosol Sci.* 17(6):921–930.

John, W. and Wall, S. M. (1983). Aerosol Testing Techniques for Size-Selective Samplers, *J. Aerosol Sci.* 14(6):713–727.

Muyshondt, A., McFarland, A. R., and Anand, A. K. (1996). Deposition of Aerosol Particles in Contraction Fittings, *Aerosol Sci. Technol.* 24(3):205–216.

Reid, J. S. and Peters, T. M. (2007). Update to "Reconciliation of Coarse Mode Sea-Salt Aerosol Particle Size Measurements and Parameterizations at a Subtropical Ocean Receptor Site" Regarding the Use of Aerodynamic Particle Sizers in Marine Environments, *J. Geophys. Res.* 112:D04202.

Secker, D. R., Kaye, P. H., Greenaway, R. S., Hirst, E., Bartley, D. L., and Videen, G. (2000). Light Scattering from Deformed Droplets and Droplets with Inclusions. I. Experimental Results, *Appl. Optics* 39(27):5023–5031.

Volckens, J. and Peters, T. M. (2005). Counting and Particle Transmission Efficiency of the Aerodynamic Particle Sizer, *J. Aerosol Sci.* 36(12):1400–1408.